SOME ASPECTS OF QUANTUM PHASE TRANSITION IN INCIPIENT FERROELECTRICS

By

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Recommendations of the Viva Voce Board

As members of the Viva Voce Board, we recommend that the dissertation prepared by **Nabyendu Das** entitled "Some Aspects of Quantum Phase Transition in Incipient Ferroelectrics" may be accepted as fulfilling the dissertation requirement for the Degree of Doctor of Philosophy.

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DECLARATION

I, hereby declare that the investigation presented in the thesis has been carried out by me. The work is original and the work has not been submitted earlier as a whole or in part for a degree/diploma at this or any other Institution or University.

Nabyendu Das

To my family

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Synopsis

The thesis contains some theoretical studies on the low temperature dielectric properties of incipient ferroelectrics such as $SrTiO_3$, $KTaO_3$, $EuTiO_3$ etc. in the vicinity a of a quantum phase transition. Studies are motivated by experimental findings on the low temperature dielectric behavior of these incipient ferroelectrics or quantum paraelectrics under various external perturbations. These materials are perovskites and are known to remain paraelectric down to any experimentally accessible low temperature. In these materials the $q = 0$ optic mode which consists of Ti or Ta motion along a [100] axis against oxygen octahedra becomes nearly unstable as temperature approaches to zero. As such the instability of this soft optic mode would lead to a ferroelectric transition as occurs in other structurally similar materials like $BaTiO_3$. However, because of neighborhood of the instability, these materials end up in a state of incipient ferroelectric, characterized by a very high, temperature independent static dielectric constant ($\mathcal{O}(10^4)$ for $SrTiO_3$) and no spontaneous polarization at low temperature ($< 10K$ for $SrTiO_3$). It is apparent that the low temperature dielectric behavior of these systems are dominated by soft transverse optic mode fluctuations near $q = 0$. Since the zone center mode has is nearly vanishing frequency at low temperature, a theoretical understanding of the dielectric behavior of these materials needs a proper account of quantum fluctuations near an instability point and its effect on the finite temperature dielectric properties. We work with simple models in each case of pure quantum paraelectrics, its coupling with anti-ferromagnetic fluctuations, strain and disorder. Then we develop a self-consistent mean-field approximation and scaling arguments, to explain some experimental findings and make various predictions about these materials.

In the first chapter some generic feature of a quantum phase transition which are relevant for these materials are introduced. In the second chapter we explore the possible consequences of quantum fluctuations in the low temperature dielectric behavior of these materials. To do that a semi-phenomenological Landau-Ginzburg theory is used. We restrict ourselves to a one component model to make our analysis simpler. Moreover anisotropy induced by the dipolar interaction in the transverse optic modes is neglected with suitable justifications. Within a mean field description with some self-consistency condition, we are able to describe the effects

of quantum fluctuations in the low temperature dielectric behavior of pure $SrTiO_3$, a well-known quantum paraelectric material. The same analysis is extended to predict its dielectric behavior when it is tuned to a quantum critical point. A prediction about the $1/T^2$ behavior in contrast to the usual Curie-Weiss behavior is made.

A recent spectroscopic experiment reports that SrTiO3 shows phase separation near its quantum critical point. This Raman scattering experiment at low temperature reports simultaneous responses from both the paraelectric and the ferroelectric phase near a quantum critical point in O^{18} doped $SrTiO_3^{16}$. The intensity of the scattered light from the ferroelectric phase is reported to be very weak and becomes weaker as one moves away from the quantum critical point. The coexistence of a quantum paraelectric phase with a quantum ferroelectric phase in O^{18}-exchanged SrTiO3 provides strong evidence for a first order phase transition. Moreover owing to the low intensity of the scattered light from ferroelectric phase, the nature of the transition can be called a weak first order where many features of a continuous transition remain unaltered. This experiment is performed at zero electric field, constant pressure and there is no report of electro-magnetic coupling in this materials. Thus one can attribute the first order nature of the quantum paraelectric to a ferroelectric phase transition to the coupling between the critical mode with non-critical strain fluctuations. Motivated by this experiment, we make an attempt to discuss the effects of the strain fluctuations in a quantum critical paraelectric in the third chapter. In our theory strain fluctuations are integrated out resulting to a long range interaction among paraelectric fluctuations. In a pure mean field scenario, a weak first order transition occurs when the effective quartic coupling of the paraelectric action is negative and close to zero. In this case one can add a higher order term with positive coefficient in the paraelectric action and make some mean field prediction about the transition. We emphasis that in such a case one should consider fluctuation effects in the quartic coupling, namely four point vertices and show that fluctuation effects can stabilize the system without invoking higher order terms. The crucial role played by long range interaction mediated by the strain fluctuations in this process is also explained. A self consistent parquet approximation is used to take care of leading order fluctuation effects. The experimental observation that the presence of the finite temperature restores the second-order nature of the transition near a quantum phase transition is also

captured in this theory.

In the fourth chapter we focus on an incipient ferroelectric $EuTiO_3$ where ferroelectric fluctuations are coupled to anti-ferromagnetic fluctuations. We consider a case where this material is tuned to ferroelectric or anti-ferroelectric quantum critical points by some non-thermal parameter. We write an action where paraelectric fluctuations are coupled to anti-ferromagnetic fluctuations in a bipartite lattice and and in presence of non-zero magnetic field. The action is used to discuss the static dielectric behavior of this system both in presence and absence of uniform magnetic field. Again a self-consistent mean field approach and scaling arguments are invoked. A new power law behavior of the static dielectric constant, namely a $T^{-\frac{3}{2}}$ variation, in presence of small non-zero magnetic field is predicted. It is in contrast of the $1/T^2$ behavior of the quantum critical paraelectric and has already got attentions of the experimental community.

Finally we look for the effects of quenched disorder in quantum critical paraelectrics using a replica formalism in the fifth chapter. Here the coupling between a random T_c type disorder with energy density is considered. Near quantum criticality in these systems, a bare power counting scheme predicts such disorder effects to be marginally relevant. A classical replica formalism with broken replica symmetry at the vector level predicts inhomogeneous solutions in these system. Gaussian fluctuations around such solutions in case of a classical phase transition were studied earlier. In their static limit the correlator of the Gaussian fluctuations due to such inhomogeneous solutions are found to independent of their sizes and a single instability was predicted. We consider the tunneling of such solutions in the quantum limit and consider a quantum phase transition in terms of the instability of Gaussian fluctuations around them. A broad power law distribution of the quantum critical points is predicted. Its consequences of the static dielectric behavior at finite temperature is also emphasized.

In conclusion, in this work the effects of quantum fluctuations on finite temperature properties of some dielectric materials are studied. Possible power law behavior of static dielectric constant at finite temperature in various materials at various external conditions are predicted using a minimal model in each case. Moreover emergent new physics near a pure quantum critical point due to the coupling with strain fluctuations and magnetic fluctuations in different materials is discussed. The effects of disorder induced inhomogeneity along with their dy-

namics at low temperature are addressed, occurrence of a mixed phase, a broad power law distribution of instability points and its consequence on the temperature dependence of the static dielectric constant is predicted. A contact with experimental scenario is made whenever possible.

List of Publications/Preprints

- **On the possibility of mixed phases in disordered quantum paraelectrics: N Das, arXiv:1104.1692**
- **Quantum critical behavior of magnetic quantum paraelectrics: N Das, arXiv:0910.4374**
- **Weak first order phase transitions in quantum ferroelectrics: N Das and S G Mishra, arXiv:0906.0944**
- **Fluctuations and criticality in quantum paraelectrics: N Das and S G Mishra, arXiv:0707.2634 , J. Phys.: Cond. Mat 21 (2009) 095901**

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1 Introduction

The basic motivation of modern condensed matter physics is to explore new physics that emerges out of complexity in a collection of large number ($\sim$ Avogadro number$\sim 10^{23}$) of interacting non-relativistic particles. The "fundamental Hamiltonian" in a condensed matter system is usually known. It consists of a collection of atoms interacting via coulomb interaction. But when a system with such a large number of particles is exposed to thermal fluctuations or enters in a quantum domain or both and when the system parameters are tuned to certain values, many novel features can emerge. Formation of crystal structure, superfluity, superconductivity are few examples of such novel phenomena. Such emergent behavior may not be adiabatically connected to the phases that appear in either side of those special points in a parameter space[1, 2]. It is difficult to capture such novel behavior in a standard perturbation theory and one needs a new mechanism, such as spontaneous symmetry breaking for phase transition[3] and new calculational scheme, such as renormalization group[4] scheme for critical phenomena to explain such behavior. Such a scenario is observed when the system undergoes a transition between two phases at zero temperature as a result of changes in some non-thermal parameter and is dubbed as *quantum phase transition*[5]. At a certain value of a non-thermal tuning parameter where a quantum phase transition of a continuous kind occurs is called a quantum critical point. At a quantum critical point system properties even at finite temperature, are governed mainly by quantum critical fluctuations. Finite temperature properties near a quantum critical point shows novel power law behaviors which are beyond the realm of any zero temperature limit of a classical theory. Such an emergent scenario drew lot of attentions in the past

and has been experimentally observed in case of itinerant magnets, He^3, etc.[6]. As a result of experimental findings, most of the earlier works in this area were directed mostly toward quantum phase transition either in quantum spin systems or in metallic magnets. Though classical phase transitions in insulating dielectric materials are well studied, there has not been any study in context of quantum phase transition. In this thesis we emphasis that the effect of quantum fluctuations can as well be observed in case of certain insulating dielectric materials, namely quantum paraelectrics such as $SrTiO_3$, $KTaO_3$ etc. In these materials a quantum phase transition occurs as a result of isotopic substitution and it involves softening of an optical mode. Thus a quantum generalization of the soft mode picture of phase transition in classical ferroelectrics is realized in these materials. A theory of quantum phase transition in these materials can be described by a continuum model which includes transverse optical modes near zone center as the most relevant degrees of freedom. It shares some similarity with the effective theory of spin fluctuations in metals. At the technical level the effective theory for quantum paraelectric to ferroelectric transition is similar to a undamped Bosonic version of the effective theory of quantum paramagnet-ferromagnetic transition in a metallic magnet. Theoretically these systems are much simpler than their magnetic counterparts to deal with, and moreover many features are experimentally observable. Thus these systems can become good playgrounds for studying some general aspects of quantum phase transitions such as finite temperature properties, disorder effects etc.

Before going into the details of the issues related to the quantum phase transition in the above mentioned materials, a brief excursion through these concepts would set a background for the present study. More detail discussions on some of the concepts if needed, will be presented in the corresponding chapter.

1.1 Quantum phase transition

Quantum phase transition is a phase transition induced by quantum fluctuations at zero temperature. Unlike the classical phase transition where the tuning parameter is temperature, one looks for a quantum phase transition by tuning a non-thermal parameter such as hydrostatic pressure, impurity concentrations, etc. Whereas a

classical phase transition corresponds to non-analyticity in the free energy as a function of temperature, a quantum phase transition involves non-analyticity in the ground state energy as a function of some non-thermal external parameter. In a strict sense, a quantum phase transition is defined only at zero temperature. However it affects finite temperature properties of a systems over a finite temperature range. Though the temperature range over which quantum critical fluctuations dominate is specific to the system, the emergent behavior at a quantum critical point is universal. The finite temperature properties depend on system dimension, order parameter dimension and symmetry, range of interaction and the *dynamic scaling exponent*. The concept of dynamic scaling exponent will be introduced in the next section. Consequences of being in the vicinity of a quantum phase transition has been observed in many systems like itinerant magnets, He^4, ferroelectrics etc. and are argued in other systems like high T_c superconductors. Before going into the detail of issues of a quantum phase transition relevant to the system we study, we now briefly introduce a path integral formulation of quantum statistical mechanics.

1.1.1 Quantum Statistical Mechanics

To calculate any physical properties of a statistical system in equilibrium we need to know its Partition function. In a statistical system quantum fluctuations become important when its temperature is much lower than its characteristic energy scale. In a quantum domain many system can be described by a Hamiltonian in operator form as $\hat{H} = \hat{T} + \hat{V}$. Where $\hat{T}$ and $\hat{V}$ are the kinetic and potential energies respectively in the operator form. With this Hamiltonian, its partition function at a finite temperature can be written as,

$$\mathcal{Z} \;=\; Tr \exp(-\beta \hat{H}). \tag{1.1}$$

Here β is the inverse temperature. If we describe the state of system in terms of a complete set of eigen states $\{|\phi_i>\}$ of some operator $\hat{\phi}$, with corresponding eigen values $(\{\phi_i\})$, then the partition function in the path integral formalism[7] can be

re-written as,

$$\mathcal{Z} \;=\; \int d\phi_a < \phi_a| \exp(-\beta \hat{H})|\phi_a > . \tag{1.2}$$

On the other hand in quantum mechanics, the transition amplitude for a system in returning to its initial state ϕ_a after a time t is given by,

$$< \phi_a| \exp(-it\hat{H})|\phi_a >= \int_{\phi(x,0)=\phi_a(x)}^{\phi(x,t)=\pm\phi_a(x)} d\phi e^{i\int_0^t dt \int dx \mathcal{L}(\phi, \frac{\partial \phi}{\partial t})}. \tag{1.3}$$

Here $\mathcal{L}$ is the Lagrangian of the system. The boundary conditions are periodic for Bosons and anti-periodic for Fermions. It is now quite evident that one can write the expression for the partition function (eqn. (1.2)) using the expression for quantum mechanical transition amplitude (eqn. (1.3)) through an Wick rotation of the time axis to the imaginary direction. This leads to the following correspondence,

$$\beta = \frac{1}{T} \equiv it. \tag{1.4}$$

At $T = 0$, i.e., at $\beta = \infty$ system acquires a complete "extra dimension". Above mathematical correspondence has interesting consequences. Firstly an equilibrium quantum statistical system in d-dimension is mapped on-to a $d + 1$-dimensional classical statistical system. Secondly the information about the dynamics of a classical system enters into the description of the corresponding quantum system. Given the knowledge of dynamics of the system, quantum-classical mapping is extremely helpful in calculating quantum fluctuations in a systematic manner. When the interaction part contains terms beyond quadratic form, an exact calculation of a quantum partition function (eqn. (1.2)) is not possible. One needs to use some systematic and controlled truncation scheme.

1.1.2 Quantum critical point

Quantum phase transition follows a similar classification as thermal phase transition. A point in parameter space where a continuous phase transition occurs at zero temperature is called a quantum critical point. Near this point the system is describable by a vanishing characteristic energy scale or a diverging correlation

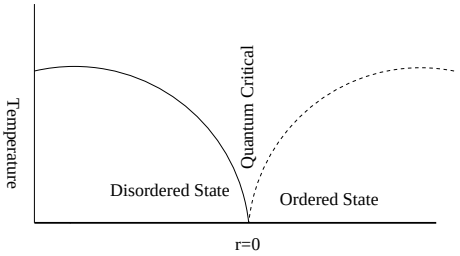

Figure 1.1: Schematic phase diagram for a quantum phase transition. r is a non-thermal parameter.

length which becomes the only relevant length scale near this point. As a result, at or near the quantum critical point various physical quantities follow power laws and such behavior can be explained by scaling arguments similar to that of the classical phase transitions[8, 9].

Dynamic scaling at $T = 0$: In general, an action in the path integral representation of a partition function can have terms with different powers of time derivative and space derivative of the field configurations. Thus in general, the scaling behavior of the characteristic time scale (τ) and the correlation length (ξ) becomes anisotropic near a quantum critical point. Near a quantum critical point a new quantity, namely *dynamic scaling exponent* z needs to be introduced. Dynamic scaling exponent characterizes the scaling behavior of the characteristic time scale (τ) with the correlation length (ξ) and is defined as

$$\tau \sim \xi^z, \tag{1.5}$$

with z positive[1] but not necessarily $= 1$. Such anisotropic scaling is used in other systems also. In case of dynamic critical phenomena where one studies the dynamics of a system near a critical point, one needs to consider such anisotropic space-time scaling to find out scaling behavior of various time dependent quantities near a critical point[10]. Similar situation also arises in case of a Lifsitz transition. In that case one considers critical phenomena in an anisotropic system and the critical properties depends on anisotropic scaling in the different directions[11]. Introduction of a dynamic scaling exponent has many consequences. A system in $d-$spatial dimension and near a quantum phase transition can be thought of as a

[1]Negative z would mean smaller relaxation time for larger size system which is unphysical.

classical system of dimension $d+z$. The *upper critical dimension*[2] of the quantum system is reduced by z, fluctuation effects becomes less relevant and the theory becomes more mean field like. However in a critical system whether it is classical or quantum, fluctuation corrections are always important and a naive perturbation theory can not have the correct answers. One needs to go beyond that and needs to invoke ideas like scaling hypothesis, various self-consistent scheme etc. In the scaling hypothesis that holds in case of a second order phase transition, any physical quantity near a quantum critical point can be written in a scaled form as follows,

$$\mathcal{O}(k,\omega,T=0) \quad = \quad \xi^{\nu}\mathcal{F}(k\xi,\omega\xi^{z},0). \tag{1.6}$$

where $\mathcal{O}$ is some physical quantity, observed at a momenta k and frequency ω. The correlation length ξ is the only important length scale in this hypothesis. $\mathcal{F}$ is the scaling function and ν is the scaling exponent. It is to be noted that though the scaling exponent is universal, the scaling function is not. Predictions based on such scaling hypothesis can be established by various theoretical schemes like self-consistent mean field theory, renormalization group theory etc.

Finite size scaling at $T \neq 0$: Since in a strict sense, a quantum critical point is defined only at zero temperature, it is not experimentally observable. However, a quantum critical point has its effects at finite T also. At a low but non-zero temperature, any physical quantity of a quantum critical system should obey power law behavior in temperature and such behavior can be obtained using the previous scaled form as shown in equation (1.6). At a non-zero temperature, a quantum critical system behaves like a finite size system of size T^{-1} in the "time direction". In this case the correlation length can not diverge but can be extended up-to a size T^{-z}. Thus we can put $\xi = T^{-z}$ in the expression eqn. (1.6) and thus in the limit $T \to 0$,

$$\mathcal{O}(\omega=0,k=0,T) \quad = \quad T^{-z\nu}\mathcal{F}(0,0,1). \tag{1.7}$$

[2]Upper critical dimension is defined as the critical value of the space dimension above which fluctuation effects does not play any major role and a mean field theory gives sufficiently correct result.

Above expression is the most experimentally relevant feature of a quantum critical point and is experimentally observed in many systems like itinerant magnets, He_4, ferroelectrics etc. The case of quantum criticality in ferroelectrics will be discussed in the next chapter where these power laws will be derived in a self-consistent mean field scheme.

1.1.3 First order quantum phase transition

Like classical phase transitions, a quantum phase transition can become first order because of the coupling to other degrees of freedom, disorder etc. A first order quantum phase transition can be observed through discontinuity of the order parameter at the transition point. A system undergoing a first order transition, evolves from its parent phase to resulting phase through a metastable state. In the intermediate phase a system shows coexistence of both the parent phase and the final phase. Classic example of a phase coexistence is the water-vapor phase transition and can be detected by some light scattering experiment. Thus in some sense a phase coexistence is also a signature of a first order transition. Many features of a first order transition can be described in Landau mean field description. In this case the free energy density for an one component system can be written as a variational form as,

$$f(\phi, r, \{\lambda_i\}) = r\phi^2 + \lambda_3\phi^3 + \lambda_4\phi^4 + \lambda_5\phi^5 + \lambda_6\phi^6 + ... \tag{1.8}$$

Here ϕ is the expectation value of some field configuration whose fluctuations are neglected completely. Such an approximation works well when the system is above its upper critical dimension and/or when order parameter dimensionality is very high. If one has the privilege to do so, the free energy density is dictated only by the symmetry of the system. The above expression is a small ϕ expansion of the variational form and the actual free energy can be found by minimizing the above expression with respect to ϕ followed by a substitution of the corresponding value of ϕ in the expression for variational ansatz. If the transformation which changes ϕ to $-\phi$ is a symmetry of the system, then terms with odd powers of ϕ are not allowed and one can truncate the free energy density at the lowest i^{th} order term

with positive λ_i. In case of positive λ_4, the Landau free energy density looks like

$$f(\phi, r, \lambda_4) = r\phi^2 + \lambda_4\phi^4. \tag{1.9}$$

Above free energy density shows a continuous phase transition at $r = 0$. For $r < 0$, order parameter is non-zero and is given by $\sqrt{\frac{-r}{2\lambda_4}}$ which smoothly goes to zero at the phase transition point.

On the other hand if it turns out that $\lambda_4 < 0$, one needs to truncate Landau free energy density at some higher power of ϕ with a positive coefficient. Let us consider the simplest possible case when $\lambda_6 > 0$. In this case the Landau free energy density looks like ,

$$f(\phi, r, \lambda_4, \lambda_6) = r\phi^2 + \lambda_4\phi^4 + \lambda_6\phi^6. \tag{1.10}$$

In this case a non-zero value of the order parameter corresponding to a metastable

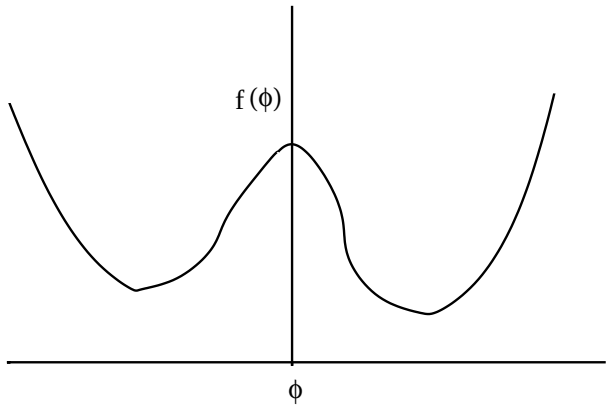

Figure 1.2: Typical Landau free energy profile for a first order transition. Eqn. (1.8) corresponds to such free energy profile when $0 \neq \lambda_3 < 0$.

minima of the Landau free energy profile develops at some positive value of r. At the phase transition point i.e. $r = 0$ order parameter has a discontinuity in its non-zero value $\sim \sqrt{\frac{-\lambda_4}{\lambda_6}}$ which corresponds to a first order transition.

However using Landau theory for a first order transition has many limitations. Firstly the Lanadu expansion is appropriate for small values of order parameter. Thus it can not incorporate the case of strong first order transition where the order parameter exhibits a large discontinuity at the phase transition point. On

the other hand for *weak first order transition*, i.e. in the limit $\lambda_4 \to 0$ one needs to consider the fluctuation corrections in the effective coupling constant for the quartic term which is also missing in a traditional Landau expansion. The later case will be considered here and will be discussed in detail in this thesis in context of weak first order transition in ferroelectrics.

1.1.4 Effects of disorder

Introduction of disorder makes a system inhomogeneous. Coupling constants in a disordered system varies from point to point. As a result system becomes a collection of ordered and non-ordered regimes. Thus one is interested in average behavior of various physical properties with a meaningful averaging scheme. If in a particular scheme, the mean of the averaged physical quantities are greater than their variances, we can say that the averaging scheme is meaningful. In such a situation, a single large system is sufficient to represent the whole ensemble and is called self-averaging. At a pure critical point randomness is classified as relevant if it leads to a change in the critical behavior (i.e., the critical exponents) of the pure system. Such systems are non self-averaging with respect to a pure critical point scenario. The relevancy of disorder for a pure critical point can be estimated using a field theoretical language as follows. Let us consider a disordered parameter $\delta r(x)$ of quenched type (no dynamics) which has a Gaussian distribution with variance g, couples to some field variable $\mathcal{O}(x,\tau)$ with scaling dimension η_0 as

$$\int d^d x d\tau \delta r(x) \mathcal{O}(x,\tau). \tag{1.11}$$

Integration of the Gaussian disorder will generate a term

$$g^2 \int d^d x d\tau_1 d\tau_2 \mathcal{O}(x,\tau_1)\mathcal{O}(x,\tau_2). \tag{1.12}$$

The above term will generate the effective disorder effect in a disordered system. Now if we use a dimensional analysis to look for the relevancy of the above term near a critical point, we see that at the zeroth order perturbation theory the coupling constant g^2 has the scaling dimension $d + 2z - 2\eta_0$. Thus near a critical point where low energy and long wave length fluctuations are most dominant, the

coupling constant g^2 becomes relevant if

$$d + 2z - 2\eta_0 > 0. \tag{1.13}$$

This is the criteria for relevancy of certain kind of disorder in a quantum phase transition. When disorder couples to energy density whose scaling dimension of the associated coupling constant is $1/\nu$, and so the dimension of the energy operator is $\eta_0 = d + z - 1/\nu$. Thus the criterion for its relevance becomes above relation becomes

$$\nu < \frac{2}{d+z}. \tag{1.14}$$

In literature this relation is known as Harris criteria[5]. Above criterion is derived on the basis of a dimensional analysis which neglects the effects of the interaction and the effects of spatial inhomogeneity as well. However it sets a criterion for the breakdown of a pure critical behavior. When certain kind of disorder is found to be relevant, one needs to consider the spatial inhomogeneity which is not included in a theory of critical phenomena in a pure system. Such a consideration needs some technique beyond standard perturbation theory. Vector breaking of the replica symmetry is such a candidate and is used to analyze the effects of disorder in ferroelectrics near a quantum phase transition in this thesis.

1.2 Quantum paraelectrics

In the previous section we have introduced some basic ideas regarding the quantum statistical mechanics and the quantum phase transitions. Now we introduce some dielectric materials where those theoretical concepts can be experimentally observed. Insulating materials such as $SrTiO_3$ and $KTaO_3$ are ABO_3 type perovskites and have interesting dielectric behavior. They are known to remain paraelectric down to any experimentally accessible low temperature. However, the $q = 0$ optic mode which consists of Ti or Ta motion along a diagonal of the cubic perovskite unit cell against oxygen octahedra becomes very nearly unstable as temperature approaches to zero. Perfect softening of this optic mode would lead to a ferroelectric transition as occurs in other structurally similar materials like $BaTiO_3$. Thus at low temperature ($< 10K$ for $SrTiO_3$), these materials end up in a state

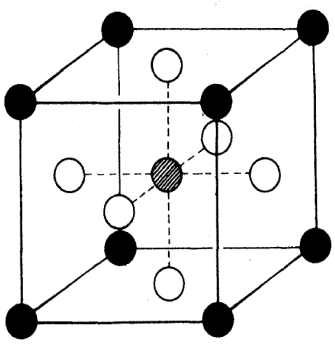

Figure 1.3: An unit cell of a ABO_3 type perovskite structure. Filled black circles are A-atom at the corners, circle filled with dashed line is the B-atom at the center and Oxygen atoms are shown with un-filled circles.

of incipient ferroelectrics, characterized by a very high, temperature independent static dielectric constant ($\mathcal{O}(10^4)$ for $SrTiO_3$) and no spontaneous polarization. Dielectric properties of these materials are being studied since long ago and they are widely known as quantum paraelectrics in the literature[16]. The correspondence between the high static dielectric susceptibility and the softening of an optic mode is also confirmed by the neutron scattering experiments[17, 18]. Thus it is evident that the quantum paraelectric systems are of displacive type and the dielectric behavior of these systems are dominated by a nearly soft $q = 0$ transverse optic mode fluctuations. A theoretical understanding of the dielectric behavior of these materials needs a proper account of not only thermal fluctuations but also quantum fluctuations arising from the optic modes near zone center. The lack of physical content of earlier theoretical works on this material, particularly regarding its vicinity to a quantum phase transition is one of the motivations for recent studies on this materials. In an earlier attempt to explain the dielectric behavior of such systems, Barrett [19] proposed a semi-phenomenological theory, which essentially recasts the Curie-Weiss formula with a replacement of temperature T there, by average energy, thereby the inverse of dielectric susceptibility could be written as, $\chi^{-1} \propto T_1 \coth(T_1/T) - T_c$, where T_c is classically calculated critical temperature and T_1 is a quantum scale $\sim (\hbar/\text{mass})$. This theory, in the high temperature limit, reproduces the Curie Weiss law. To match experimental data in $SrTiO_3$ the Barretts' formula has been found inadequate as one single constant quantum scale T_1 can not trace the full curve. The formula has since been modified

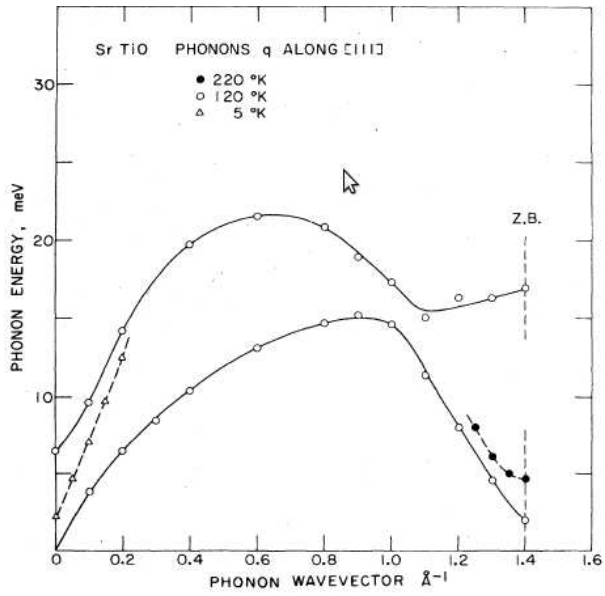

Figure 1.4: Temperature-dependent phonon modes in $SrTiO_3$ measured by Shirane G and Yamada Y, Phys. Rev. 177, 858 (1969). The 111^oK transition is caused by the soft mode at the zone boundary. Soft mode near the origin is due to incipient ferroelectricity.

in various ways, for example, by introducing an extra exponent [20], that is, by writing χ^{-1} as $(T_1 \coth(T_1/T) - T_c)^{-\nu}$, and by making T_1 temperature dependent with an extra scale [21], to take care of various "anomalies", for example the one near 40K. There has been a proposal of attributing this extra energy scale to the structural transition which occurs at 110K [22]. These proposals either follow an order parameter expansion similar to the Landau expansion or some modifications thereof, hence they do not introduce any new microscopic description. Moreover these attempts considers fluctuations arising from $q = 0$ mode only and misses a fact that the characteristic energy scale in this systems are very low, i.e. these systems are near a quantum phase transition. We assume that the would be quantum phase transition from a paraelectric phase to a ferroelectric phase in this materials to a continuous transition and will show that analysis based on such a view point can capture many features of the dielectric behavior of these system which were untouched by the previous theories.

We analyze the fluctuation effects in such systems within a self consistent mean field approximation. The theory involves a lowest order perturbation expansion of

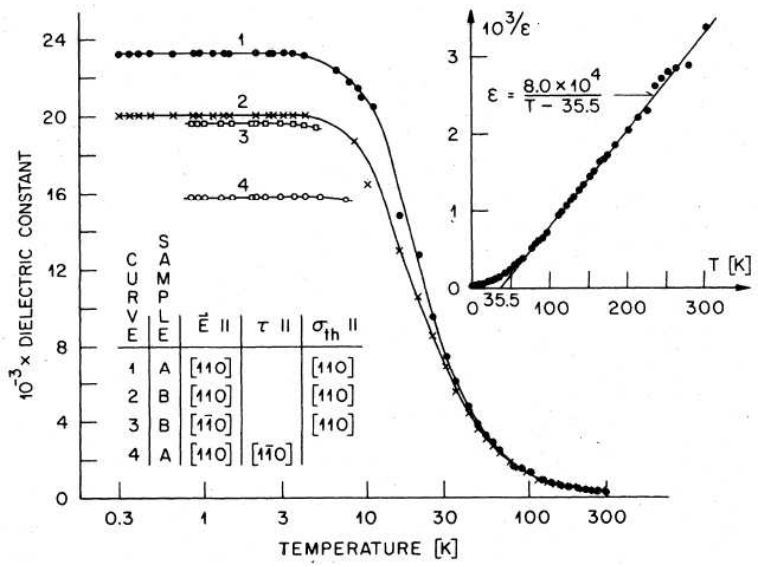

Figure 1.5: Static dielectric constants ϵ_{110} and $\epsilon 1\bar{1}0$ of the monodomain $SrTiO_3$ samples. Inset: $10^3/\epsilon \, vs \, T$. Reference[16].

a continuum theory with a momentum cut-off and self consistent conditions[23]. Results depend on the choice of the cut-off and we have shown that a choice of temperature dependent cut-off at/near a quantum critical point can lead to a novel T^{-2} behavior of the static dielectric susceptibility which is argued by scaling analysis[24] and also verified by recent experiment[25]. Motivated by the success of the assumption of nearness of these materials to a quantum critical point we discuss the effects of strain coupling, magneto-electric coupling and the quenched disorder in these materials. Each case will be analyzed by a minimal action and suitable mean-field scheme. Analysis are mostly analytic and are motivated to capture the basic physics rather than exact matching with the experimental data. Moreover due the universality of the behavior near a quantum critical point many of these analysis will also be useful for analyzing the quantum critical behavior of a large class of Bosonic systems with undamped dynamics.

The thesis is organized in the following manner. In this chapter, basic concepts related to quantum phase transitions, phase transitions in ferroelectrics and some experimental facts about some incipient ferroelectrics such as $SrTiO_3$ and $KTaO_3$ are introduced. In the next chapter we will explore the possibility of quantum critical phenomena and its consequences in these materials. Attempts are made

to analyze low temperature behavior of these system with the assumption of its nearness to a quantum critical point. It is followed by a theory of weak first order quantum phase transition observed in $SrTiO_3$ which is assumed to be a result of the strain coupling. Analysis is based on a self-consistent scheme for vertex corrections at non-zero polarization. Predictions are made on the discontinuity in the non-zero polarization both at zero and non-zero temperature and are in accord with experiments. Chapter four is an account of dielectric behavior of an incipient ferroelectric $EuTiO_3$ where ferroelectric fluctuations are coupled to antiferromagnetic fluctuations. Invoking a self-consistent scheme similar to the first chapter in presence of magneto-electric coupling and external magnetic field, predictions are made about new power law behavior of the static dielectric behavior at finite temperature. Predictions are new and worth further experimental investigations. Next chapter is an account of the effects of disorder in quantum critical paraelectrics. Discussions are based on semi-phenomenological Ginzburg-Landau theory with self-consistent mean field analysis. In this case a replica formalism is invoked to take account of fluctuations from locally ordered regimes. A prediction broad power law distribution of the instability points and its consequences on the temperature dependence of the static dielectric behavior are made.

2

Quantum criticality in ferroelectrics

2.1 Introduction

In this chapter we discuss the low temperature dielectric properties of quantum paraelectrics like $SrTiO_3$ as a result of their vicinity to a transition from a paraelectric phase to a ferroelectric phase at zero temperature or a ferroelectric quantum phase transition. These materials are introduced in the previous chapter. A ferroelectric transition in these materials can be induced by tuning non-thermal parameters such as doping concentration by isotopic substitution, which to a good approximation can be assumed as of continuous type[1]. Thus a theory of the low temperature dielectric behavior of such systems needs proper account of the fluctuations near a quantum critical point. As a matter of fact, ferroelectric transition in materials like $SrTiO_3$ involves softening of a transverse optic mode. This kind of phase transition also occurs in case of classical ferroelectric phase transition in $BaTiO_3$ and is called displacive transition. Unlike the case of order-disorder transition where a local moment is always present, in this case the moment formation and their ordering take place simultaneously. Phase transitions in such system can not be described by an Ising Hamiltonian, which is usually invoked for a system going through order-disorder transition. Dielectric behavior of these systems are governed by collective oscillations of coupled dipoles and the phase transition is described by softening of the corresponding optical mode due to ther-

[1]Experimental results suggest that the case of $SrTiO_3$ is of weak first order type[37]. Many of the discussions in context of a quantum critical point also hold in case of an weak first order transition and will be discussed in the next chapter.

mal fluctuations[13, 14]. Due to the importance of both the collective behavior as well as the quantum fluctuations, in these cases a proper quantum generalizations of the classical soft mode concept is needed to describe various aspects of low temperature behavior. Moreover as a system approaches a quantum critical point the interactions between the zone-center critical mode and other modes near it becomes increasingly important which also need proper considerations.

2.2 Mean Field Analysis

The low temperature physics of these systems is dominated by fluctuations of transition metal ions from their equilibrium position (center of the unit cell) in the background of other ions. The action for such interacting ions is modeled in terms of local displacements of the fluctuating transition metal ions with a nearest neighbor harmonic interaction[28],

$$\mathcal{A} = \sum_{\mathbf{l}} \left\{ \frac{\dot{\phi}_{\mathbf{l}}^2}{2} + \frac{1}{2}\omega_0^2\phi_{\mathbf{l}}^2 + \frac{1}{4}\lambda\phi_{\mathbf{l}}^4 \right\} - \frac{1}{2}\sum_{\mathbf{l}\mathbf{l}'} v\phi_{\mathbf{l}}\phi_{\mathbf{l}'}. \tag{2.1}$$

Here $\phi_{\mathbf{l}}$ displacement of the transition metal ions in the $\mathbf{l}$-th unit cell and $\dot{\phi}_{\mathbf{l}}$ is the time derivative of $\phi_{\mathbf{l}}$. For simplicity we consider $\phi_{\mathbf{l}}$ to be one component. The constants λ and v are assumed to be positive and mass taken as unity. For $|v| << |\omega_0^2|$ and $\omega_0^2 \ll 0$, the above action describes two local minima with a nearest neighbor coupling v. In that case it mimics a two state Ising system with Gaussian fluctuations around one of the local minima. When $|v| \sim |\omega_0^2|$, there is a possibility of large tunneling between these minima. In this regime the system has to be described in terms of its collective behavior. Such system is called displacive system and the limit $|v| \to |\omega_0^2|$ is called Displacive limit. In momentum space,

$$\mathcal{A} = \sum_{\mathbf{q}} \frac{1}{2}\dot{\phi}_{\mathbf{q}}^2 + \frac{1}{2}\sum_{\mathbf{q}}(\omega_0^2 - v\delta \sum_{i=x,y,z} \cos q_i a)\phi_{\mathbf{q}}\phi_{-\mathbf{q}} + \frac{1}{4}\lambda \sum_{\mathbf{q}_1,\mathbf{q}_2,\mathbf{q}_3} \phi_{\mathbf{q}_1}\phi_{\mathbf{q}_2}\phi_{\mathbf{q}_3}\phi_{-\mathbf{q}_1-\mathbf{q}_2-\mathbf{q}_3} \tag{2.2}$$

Here δ is the coordination number and a is the lattice spacing. In Fourier space $p_{\mathbf{q}}(\omega) = \imath\dot{u}_{\mathbf{q}}(\omega) = -\imath\omega\phi_{\mathbf{q}}(\omega)$ contributes a $\omega^2\phi_{\mathbf{q}}(\omega)\phi_{-\mathbf{q}}(\omega)$ in the kinetic energy term. We decouple the quartic term which creates interactions among the harmonic

or free phonons in a quasi harmonic approximation as follows,

$$\sum_{\mathbf{l}} \phi_{\mathbf{l}}^4 \approx 6N(\sigma + \langle\phi\rangle^2) \sum_{\mathbf{q}_1} \phi_{\mathbf{q}_1}\phi_{-\mathbf{q}_1} \tag{2.3}$$

where $< .... >$ denotes a thermal averaging and σ is defined as

$$\sigma = \sum_{\mathbf{q}} \langle T\phi_{\mathbf{q}}(0)\phi_{-\mathbf{q}}(0^+)\rangle. \tag{2.4}$$

Finally the action for quasi-harmonic phonons can be written as,

$$\mathcal{A} = \frac{1}{2}\sum_{\mathbf{q}} (\omega_{\mathbf{q}}^2 - \omega^2)\phi_{\mathbf{q}}\phi_{-\mathbf{q}} \tag{2.5}$$

where $\omega_{\mathbf{q}}$ is the renormalized value of the oscillator frequency and for isotropic case, is given as

$$\omega_q^2 = \omega_0^2 - v\delta\cos qa + 3\lambda\sigma \simeq \omega_0^2 - v + v\delta a^2q^2 + 3\lambda\sigma, \tag{2.6}$$

for small q. Such a truncation to the lowest order contribution from spatial variations is quite justified for a near critical system where only low energy and long wavelength fluctuations are important. We are interested in the paraelectric phase of the system, that is, where $< \phi >= 0$. Since the system is at low temperature and the dielectric constant has an enhanced value, $< \phi^2 >$ need not vanish, however. The purpose of present work is to present a self consistent calculation of $< \phi^2 >$ in classical as well as in the quantum regime. In the previous chapter we have discussed how a quantum statistical system can be mapped onto a dynamical model. All one need is to consider the dynamics in the imaginary time. The frequencies corresponding to the imaginary time in Fourier space is called Matsubara frequencies (ω_n). Owing to the different statistics of the Bosons and the Fermions, $\omega_n = 2n\pi T$ and $(2n+1)\pi T$ for these two case respectively where n is an integer and T is the temperature of the system[29]. The susceptibility, which is related to $< \phi^2 >$, is essentially the phonon propagator corresponding to the action for quasi-harmonic phonons (eqn. (2.5)) and can be written in Matsubara frequency

as

$$\chi(\mathbf{q},n) = -\frac{1}{(\imath\omega_n)^2 - \omega_q^2}, \quad \omega_n = 2n\pi T. \tag{2.7}$$

Above propagator depends of $< \phi^2 >$ or σ and the dependence is included in the expression for ω_q in equation(2.6). Using the definition of sigma (eqn. (2.4)) we have a self consistent equation,

$$\begin{aligned} \sigma &= \sum_{\mathbf{q}} \langle T\phi_{\mathbf{q}}(0)\phi_{-\mathbf{q}}(0^+)\rangle = \frac{1}{\beta}\sum_{\mathbf{q},n} \chi_{\mathbf{q}n} e^{\imath\omega_m 0^+} \\ &= \frac{1}{\beta}\sum_{\mathbf{q},n} \frac{1}{\omega_n^2 + \omega_q^2} = \sum_{\mathbf{q}} \frac{1}{2\omega_q} \coth\left(\frac{\omega_q}{2T}\right). \end{aligned} \tag{2.8}$$

The solution of this equation will determine σ at zero temperature as well as its temperature dependence at finite temperature. Above equation in its asymptotic forms reduces to,

$$\sigma = \begin{cases} \sum_{\mathbf{q}} \frac{T}{\omega_q^2} \sim \int_0^\Lambda dq\ q^2 \frac{T}{\omega_0^2 - v + v\delta q^2 + 3\lambda\sigma} & (\mathrm{T} >> \omega_\Lambda) \\ \sum_{\mathbf{q}} \frac{1}{\omega_q} \sim \int_0^\Lambda dq\ q^2 \frac{1}{\sqrt{\omega_0^2 - v + v\delta q^2 + 3\lambda\sigma}} & (\mathrm{T} << \omega_\Lambda). \end{cases} \tag{2.9}$$

The integrals can be performed analytically and are cut-off (Λ) dependent. We need to impose such cut-off to avoid ultraviolet divergences in the integrals. However in condensed matter system there is always a natural ultraviolet cut-off which determines the maximum momentum scale up-to which a continuum description is valid. For any fluctuations in a ordinary periodic solid inverse lattice spacing is an example of such a ultraviolet cut-off. In case a system is far away from quantum criticality, one can divide a high temperature ($T >> \omega_\Lambda$) and a low temperature ($T << \omega_\Lambda$) regime using such a ultraviolet cut-off. However in case of a quantum critical system, we see that (fig. (2.2)) such a demarcation is also governed by temperature itself. Thus for calculating leading order temperature dependent contribution from the fluctuation integral to the dielectric susceptibility, we use a high temperature expansion as above with a temperature dependent cut-off in this thesis. Before we go into details of the temperature dependence of static dielectric susceptibility, we need to define some dimensionless parameters such as, $\Delta = (\omega_0^2 - v)/\omega_0^2$, $\sigma_c = (\omega_0^2 - v)/3\lambda$, $\eta = \hbar/(2\omega_0\sigma_c)$ and $\hbar$ is taken as unity for the rest of the discussions. The parameter Δ describes the effective stiffness for collec-

tive modes at harmonic level. The strength of coupling between various modes near $q = 0$ is determined by σ_c^{-1} while the parameter η tells us about the vicinity to the quantum limit in the system. Introducing normalized temperature $x = T/m\omega_0^2\sigma_c$ and using the previously defined parameters, we rearrange the equation (2.6) as follows

$$\frac{\omega_q^2}{\omega_0^2} = \frac{v\delta a^2 q^2}{m\omega_0^2} + \Delta(\frac{\sigma}{\sigma_c} + 1) \tag{2.10}$$

where

$$\frac{\sigma}{\sigma_c} = \sum_{\mathbf{q}} \frac{\eta\omega_0}{\omega_q} \coth\left(\frac{\eta\omega_q}{\omega_0 x}\right). \tag{2.11}$$

A self-consistent solution of these equations will determine the inverse dielectric susceptibility which using eqn. (2.7) and (2.10) can be written as,

$$\chi(0,0)^{-1} \propto \Delta(\frac{\sigma}{\sigma_c} + 1). \tag{2.12}$$

A numerical calculation of the self-consistent equations (2.10, 2.11) is presented in

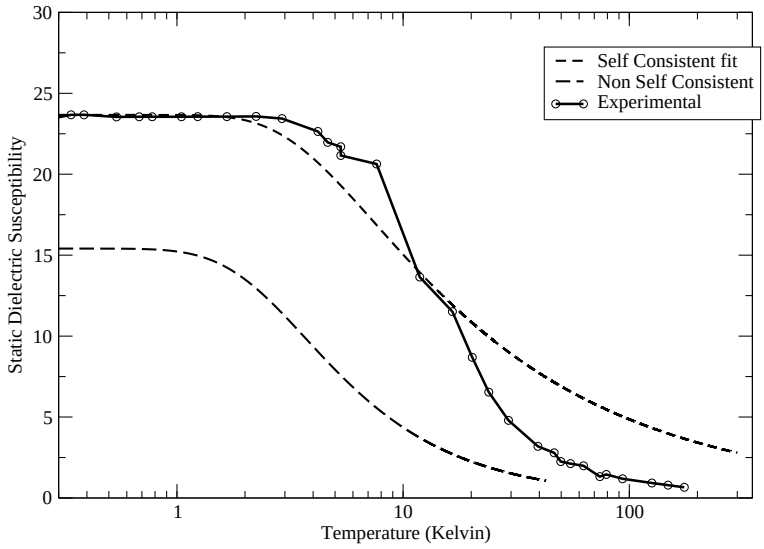

Figure 2.1: Numerical solution shows saturation in Static susceptibility (in units of 10^4) vs Temperature curve. This curve is in good agreement with Muller's experiment in low temperature side, with $\frac{v\delta a^2}{\omega_0^2} = 1, \Delta = 0.0025, \eta = 1/\Delta, q_{max} = 0.1$ and at the end χ and T are rescaled with $0.4/\Delta$ and 30Δ respectively. The lower curve is the non-self consistent fit with the same parameters as the upper one but with rescaling of χ and T by 9.75 and 100 respectively.

the figure 2.1. From the numerical calculation we learn that the high value of static dielectric susceptibility of $SrTiO_3$ is fitted with the dimensionless parameter $\Delta = 0.003$, which is indeed a small number. This gives us another justification to treat this system to be near a quantum critical point. The static dielectric susceptibility data of $SrTiO_3$ remind us of the behavior of itinerant Fermionic systems near quantum phase transition point and fluctuation regime around that. There the (staggered) magnetic susceptibility diverges for (anti-)ferromagnetic transition as the coupling constant crosses a critical value[30]. The case of $SrTiO_3$ is similar to that of liquid Helium-3 [31], where the magnetic susceptibility gets enhanced, as large as ten times, depending upon pressure, from its free Fermionic value.

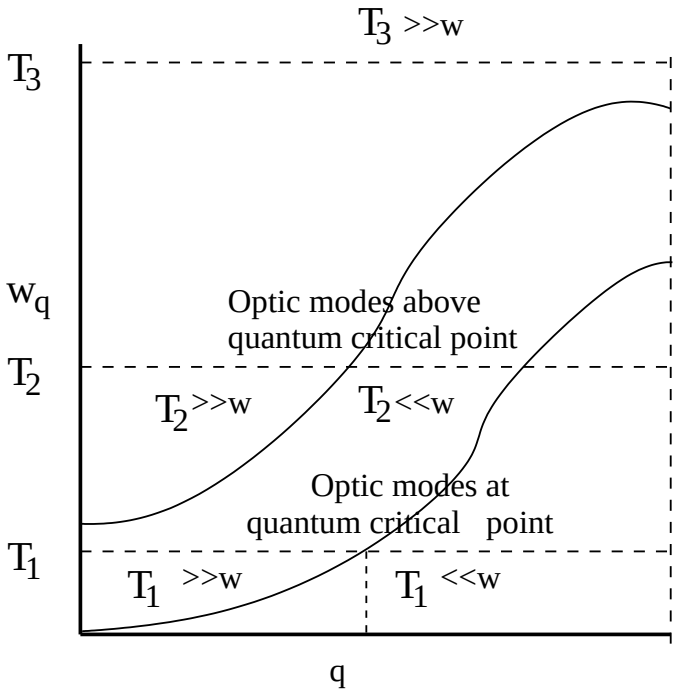

Figure 2.2: Schematic diagram showing how the momentum cut-off becomes temperature dependent at a quantum critical point. At very high temperature (T_3) the momentum cut-off is always temperature independent. At lower temperature a temperature dependent momentum cut-off demarcates between the high temperature and the low temperature regime. Only in the case of a quantum critical branch, where the energy gap for $q = 0$ mode vanishes has a momentum cut-off $\sim T$.

A system far above its quantum critical point has non-zero energy gap(Δ) for

$q = 0$ mode. For low enough temperature i.e. $T << \Delta$, quantum fluctuations dominate the low temperature physics. In this case the momentum cut-off in the integral (eqn. (2.9)) is temperature independent as shown by T_1 in figure 2.2 and so is σ. At a higher temperature as shown by T_2 in the same figure, there is a crossover from a quantum domain to classical one at a cut-off determined by the energy gap of the $q = 0$ mode. Again the cut-off is weakly temperature dependent ($\Lambda \sim (T - \Delta)^{1/2}$) and equation (2.9) tells that $\sigma \sim T$ in the leading order. The mode coupling would give corrections higher order in temperature, and T_c would be proportional to Δ. On the other hand as Δ become smaller and η becomes larger, the system move towards the quantum criticality. When Δ or T_c becomes identically zero we have quantum critical point. At this point the zero temperature static dielectric susceptibility diverges and because of quantum critical fluctuations it shows novel power law behavior at low but finite temperature. Interestingly the $\Delta = 0$ or $\omega_0^2 = v$ limit is the *displacive limit*, well known in the structural transition literature. Owing to the vanishing Δ, the momentum cut-off (Λ) in the integral (2.9) becomes strongly temperature dependent at quantum critical point and the dispersion relation (2.6) tells that $\Lambda \sim T$. A non-self-consistent estimate with ($\Lambda \sim T$), which neglects $3\lambda\sigma$ in the right hand side of the equation (2.9) tells that σ and hence the inverse of the static dielectric constant follows a T^2 behavior at any finite temperature up-to the Debye temperature. Though the Debye energy scale is system specific, the exponent is same for other systems with same dispersion relation. Such estimate is essentially an outcome of the lowest order perturbation theory which gives quite correct result when the system is far away from the quantum critical point i.e. $|(\omega_0^2 - v)/3\lambda\sigma| << 1$. At quantum critical point, an estimation of the self-consistent correction by putting $\sigma \sim \lambda T^2$ in the right hand side of the equation (2.9) is found also $\sim T^2$. Thus self-consistency condition in this case changes the coefficient of T^2 only. Thus as far as the basic physics is concerned, a non-self consistent prediction is sufficient for this material. However to fit an experimental data self-consistent calculation becomes important.

2.3 Phase diagram and Hydrostatic Pressure at QCP

Based on the previous discussions, we now focus on a possible phase diagram for the dielectric systems near a quantum phase transition. If we focus on the phase diagram these materials near the ferroelectric quantum critical point where the power law behavior of the dielectric constant etc is valid. In our estimate T^2 is the leading order correction to the paraelectric gap near the quantum critical point. A more sophisticated calculation can lead to a slight deviation from T^2 corrections but the basic physics will remain the same. In the regime $\Delta \leq 0$ self consistency

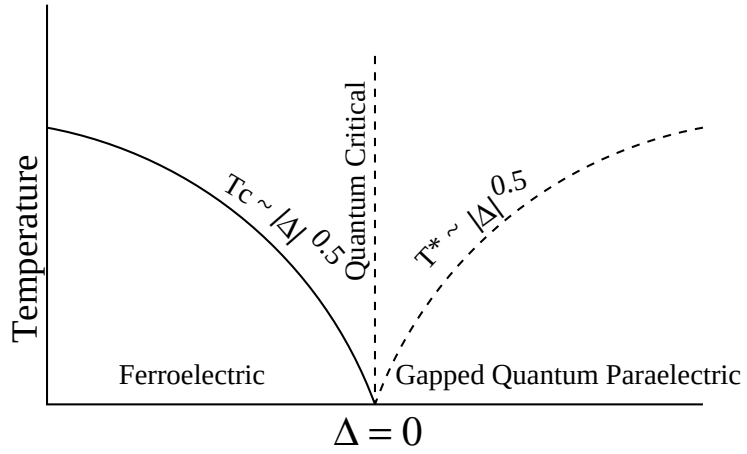

Figure 2.3: Schematic phase diagram of a typical quantum paraelectric system.

in fluctuation breaks down, system seeks ordering and hence an expansion about the non-zero $< u >$ is required. A similar analysis in this regime will also lead to a T^2 corrections. As a result the transition temperature T_c determined by the solution of the equation $\Delta + \lambda T^2$, namely the gap equation, $\sim |\Delta|^{\frac{1}{2}}$. On the other hand, in $\Delta \geq 0$ regime the system can not have any ordering and its behavior has to described by self consistent fluctuations as done in the previous section. There is a characteristic temperature (crossover temperature in modern parlance [5]) $T^* \sim \Delta^{\frac{1}{2}}$ which demarcates the boundary between the low temperature *gapped quantum paraelectric* behavior and the classical behavior. In case of $SrTiO_3$, the plateau in the susceptibility vs temperature curve is the signature of gapped quan-

tum paraelectric behavior. There is no transition in this system. But there is a crossover from low temperature quantum to high temperature classical behavior at the crossover temperature T^* ($\sim 10K$). This is exactly the temperature where plateau ends and the susceptibility curve stars following a Curie behavior. One can now hope to reach at $\Delta = 0$ through tuning some parameters like pressure, impurity etc. The width of this plateau regime vanishes at this point and the system becomes *quantum critical*. At this point thermodynamics will be described by power laws in temperature (e. g. $\chi(0,0)^{-1} \sim T^{-2}$) and the system will show some non trivial dynamics. The later is beyond the scope of the present work. It is quite evident here as the controlling factor $v/m\omega_0^2$ strongly depends upon structural aspects and hence this quantum-ness in $SrTiO_3$ can be properly understood through some intrinsic mechanism which give rise to such large tunneling. The importance of the vicinity to a quantum critical point in determining the low temperature properties of a quantum paraelectric shows the limitation of the Barrett's analysis and its variants. Clearly it can not capture the consequences of the quantum critical fluctuations. That formula is essentially attempted at mimicking the quantum fluctuations in a single mode theory, which would fail near the quantum critical point as many modes and their coupling would dominate the behavior of system there. This necessitates a self-consistent calculation for quantum paraelectrics near its quantum critical point.

A possibility of exploring the physics near such quantum critical point is through application of hydrostatic pressure. Such a technique is already used in case of ferroelectrics and quantum paraelectrics long ago [32] and more recently [33] in different contexts. We found that those experimental results can be discussed more interestingly as is done in the context of itinerant magnetic system[34]. Application of hydrostatic pressure will couple to optical mode via its coupling to the acoustic mode. In this case the starting action takes the form

$$\begin{aligned}\mathcal{A} &= \frac{1}{2}\int dq \left[p_q^2 + \left(\omega_0^2 - v\delta \sum_{i=x,y,z} \cos q_i a\right) u_q u_{-q}\right] + \frac{\lambda}{4}\int \Pi_{i=1}^4 (dq_i u_{q_i}) \delta(\sum_i q_i) \\ &\quad + g\int dk\ dq\ \epsilon(k)\ u_q u_{k-q} + \frac{K}{2}\int dq \epsilon^2(q) - p\epsilon(0). \qquad (2.13)\end{aligned}$$

Here last three terms are results of applications of pressure, in lowest possible

order. The parameter "g" couples strain fields to unit cell displacement related to optic mode and "K" is the force constant for harmonic acoustic phonons, and the last term shows the coupling of the hydrostatic pressure "p" to the static strain with some unit strength. Now if the pressure is strong enough ϵ has a minima at $\epsilon = \epsilon(0)$ and is given by

$$\epsilon(0) = p/K. \tag{2.14}$$

substituting the above relation in equation (2.13) and neglecting the strain fluctuations, we get an effective action

$$\begin{aligned} \mathcal{A} &= \int dq \left[\frac{1}{2} p_q^2 + \frac{1}{2} \left(\omega_0^2 + gp - v\delta \sum_{i=x,y,z} \cos q_i a \right) \phi_q \phi_{-q} \right] \\ &+ \frac{1}{4} \lambda \int \Pi_i dq_i \phi_{q_1} \phi_{q_2} \phi_{q_3} \phi_{-q_1-q_2-q_3}. \end{aligned} \tag{2.15}$$

Again we write a self consistent equation for paraelectric fluctuations as,

$$\sigma = \int d^d q \frac{1}{\omega_q} \coth \left(\frac{\omega_q}{T} \right). \tag{2.16}$$

Here the renormalized value of the optical mode frequencies are given as

$$\omega^2(q) = 3\Delta\lambda(1 + p/p_0) + v\delta q^2 a^2/2 + 3\lambda_R \sigma \text{ and } p_0 = \frac{3K\Delta\lambda}{g}. \tag{2.17}$$

Up to this point result is just a renormalization of the factor Δ as $\Delta(1 + p/p_0)$ and it becomes an experimentally controllable parameter. And the behavior of susceptibility at different values of Δ is shown in the figure 2.4. It is visible in this figure that as Δ decreases, the saturated value of the static dielectric constant increases, the curve becomes a straight line down to zero temperature, signaling a power law variation over the whole temperature range. In this proposal we assume strain fluctuations to be negligible. However strain fluctuations can generate long range interactions among the harmonic paraelectric fluctuations, i.e. a quartic term with a coupling constant $\sim g^2/K$. And in certain situation this coupling constant can become negative and its magnitude can become comparable to λ. In that case it is quite possible that the transition will be first order and such scenario is discussed in the next chapter. However in real situation one can try to induce the

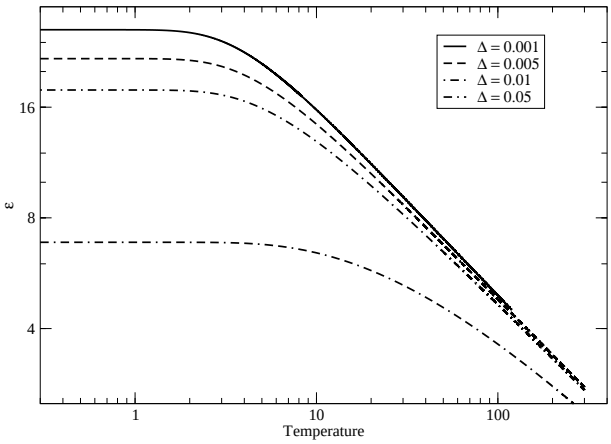

Figure 2.4: Temperature variation of Susceptibility at different values of Δ and the log-log plot of the same.

effect of negative pressure required in these systems to achieve a quantum critical point through some homogeneous effects of non-polar impurity. But in either case nature of the transition can be modified because of strain coupling or disorder respectively.

2.4 Discussion

We have shown that a mean field theory for quantum paraelectric fluctuations within a quasi harmonic approximation reproduces the low temperature behavior of the static dielectric susceptibility of a quantum paraelectric. The qualitative behavior of susceptibility is reproduced as well as a new insight gained into the quantum critical behavior of such systems. A mismatch in theory and experiment for the static dielectric constant at high temperature can be attributed to the effect of structural transition which occurs at higher temperature (i.e. at 110 K in $SrTiO_3$), such discrepancy is irrelevant for the present discussion which refers mainly to the low temperature regime. The short range model studied here is justified since only transverse optical modes are involved in the quantum paraelectric fluctuations. In presence of a long range dipolar interaction longitudinal mode becomes stiff and only transverse modes can get soft. The dipolar interaction induces a certain amount of anisotropy to the transverse modes which can certainly change

the critical behavior, however, only with a fairly large value of dipolar contribution to anisotropy in the quadratic term [26]. Usually such anisotropy parameters are picked up from ab-initio band structure calculations. We are not aware of such ab-initio band structure results for anisotropy parameters in case of $SrTiO_3$ or $KTaO_3$. However, the band structure calculations support our choice of parameter for the effective stiffness. Compared to $BaTiO_3$ it is about twenty times smaller (Table V in ref [36]) for $SrTiO_3$, which makes it more near the quantum domain. On the other hand the lattice induced anisotropy in the quartic term is of the same order of magnitude and it would not play a key role in distinguishing the low temperature behavior in these systems. We leave discussions on anisotropy dependence for the future work and stick to an isotropic short range model. It is also clear that there is no need to introduce "anomalous" regime as proposed earlier. That proposal might be due to the insistence on comparing experimental results with Barrett's formula and its extensions. The experimental behavior is well accounted for in the quantum region and at high temperature the susceptibility smoothly crosses over to the classical behavior. The structural aspects and anisotropy effects are not attempted here.

3

Weak first order transition in quantum paraelectrics

3.1 Introduction

Previous chapter sets up a basic theoretical ground for discussing the low temperature dielectric properties of quantum paraelectrics along with some predictions about their quantum critical behavior. With this background we focus on understanding the detailed experimental observations on various quantum paraelectrics. In this context a recent spectroscopic experiment reports some interesting behavior of quantum critical $SrTiO_3$. This experiment[37] indicates that $SrTiO_3$, one of the member in the quantum paraelectric family, shows phase separation near its quantum critical point. This Raman scattering experiment at low temperature reports simultaneous responses from both the paraelectric and the ferroelectric phase near a quantum critical point in O^{18} doped $SrTiO_3^{16}$. The intensity of the scattered light from the ferroelectric phase is reported to be very weak and becomes weaker as one moves away from the quantum critical point. The coexistence of a quantum paraelectric phase with a ferroelectric phase in O^{18}-exchanged $SrTiO_3$ provides strong evidence for a first order phase transition. Moreover owing to the low intensity of the scattered light from ferroelectric phase, the nature of the transition can be called a weak first order type where many features of a continuous transition remain unaltered. This experiment is performed at zero electric field, at a constant pressure and there is no report of electro-magnetic coupling in this materials. Thus one can safely attribute this first order nature of the quantum

paraelectric to a ferroelectric phase transition to the coupling between the critical mode with non-critical strain fluctuations. Such a coupling is quite common in classical ferroelectrics and has been studied both theoretically as well as experimentally in earlier literature. Earlier experiments show that the application of hydrostatic pressure moves these systems away from criticality and the possibility of phase transition is suppressed. One needs to apply, what is termed as, *a negative pressure* to induce phase transition in these materials. One way to simulate negative pressure is to put non-polar impurities which create local pressure deficiencies. In this context, experimentally [38] one finds $T_c \sim (n - n_c)^{\frac{1}{2}}$ (where n is the average impurity concentration and n_c is the critical value, typically 33%) which matches well with the theoretical estimated[23] transition temperature for pressure induced transition. In this case, the mean field $T_c \sim (p + p_c)^{\frac{1}{2}}$ (where p is hydrostatic pressure and p_c is the critical value). The exponent $\frac{1}{2}$ is obtained when thermal fluctuations are treated at the Gaussian level. The similarity between effects of pressure and the impurity in the transition temperature can be attributed to the high density of impurity concentrations in these cases. Here the disorder effects seem to be small and a non-polar impurity essentially induces an internal pressure. This motivates us to develop a description, suitable for the properties of the pressure induced phase transition, which can be used to understand the occurrence of the phase separation mentioned above in the ferroelectric transition near the ferroelectric quantum critical point. In this chapter we look for a weak first order transition scenario where correlation length is large enough to adopt a continuum model. We therefore, start with an effective one component model with short range interaction without dipolar anisotropy and try to explore the results of order parameter fluctuations with an effective long range interactions among them, mediated by strain fluctuations. We look for the fluctuation effects in four point vertices. To retain the leading fluctuation effects in the vertex function along with their dependencies on the non-zero polarization, we calculate the free energy using a set of renormalization group equations. This fluctuation renormalized free energy is used to explore the possibility of a first order transition at zero temperature as well as at a finite temperature.

3.2 Summary of the mean field analysis

We assume that a fluctuating strain field $\epsilon_{ij}(\tau) \sim (\nabla_i u_j(\tau) + \nabla_j u_i(\tau))$ couples to a bi-linear form of the optical modes fluctuations as $g\epsilon_{ij}(\tau)\phi_i(\tau)\phi_j(\tau)$. Here $u_i(\tau)$ represents the displacement due to acoustic mode fluctuations at i-th site in real space and g is the opto-elastic coupling. We consider the Gaussian fluctuations of the strain fields and consider the system to be at a constant pressure. Integrating out the strain fluctuations completely, we get an effective long range interactions among the optical mode fluctuations of the form $v\phi_i^2\phi_j^2$ in real space. Here $v \propto g^2$ and depends on various elastic constants depending on which it can be either negative or positive. A naive quantum generalization of such interaction would lead to a term like $v\phi_i^2(\tau)\phi_j^2(\tau)$, where τ is time. Such a term indicates field variables at two different position interact at same point with same interaction strength. This clearly violates causality and we need to introduce non-locality in time in such interaction. Thus we the resulting interaction to be $v\phi_i^2(\tau_i)\phi_j^2(\tau_j)$ which is found to consistent with the quantum-classical mapping of our strain coupled system. Since we consider a weak first order transition a-priori, we assume v to be negative and leave the detailed discussions on its dependence on various elastic constants. In a Fourier space our effective action describes only polarization fluctuations transverse to the momentum vector with strain induced long range interactions among them and takes the following form,

$$\begin{aligned}\mathcal{A} &= \frac{1}{\beta}\sum_q \frac{1}{2}(\omega_n^2 + r + cq^2)\phi_q\phi_{-q} + \frac{1}{4!\beta}\sum_{q_1,q_2,q_3} u\phi_{q_1}\phi_{q_2}\phi_{q_3}\phi_{-q_1-q_2-q_3} \\ &\quad + \frac{vL^{-d}}{4!\beta}\sum_{q_1,q_2}\phi_{q_1}\phi_{-q_1}\phi_{q_2}\phi_{-q_2}. \end{aligned} \tag{3.1}$$

Here $\phi_{q_i} = \phi(\mathbf{q}_i, \omega_{n_i})$ describes the Fourier transform of local transverse polarization, $\mathbf{q}$ is the field momentum, $\omega_n = 2\pi n/\beta$ is the Matsubara frequency for Bosonic excitations, r and u are the coupling constants for quadratic and the anisotropic short range quartic interactions respectively. The parameter v is the coupling constant for isotropic long range part of the quartic coupling induced by strain and L^d is the system volume in d-spatial dimension. Hydrostatic pressure, as well as the non-polar impurity, couples to the optical mode via strain. It shifts the bare

quadratic and quartic coupling r_0 by $r = r_0(1+p/p_0)$, where p is the homogeneous pressure and p_0 is a constant. Strain fluctuations induce a long range attractive interaction between the dipoles and is denoted by the effective quartic coupling v. We focus on a weak first order transition near a quantum critical point where ϕ acquires a non-zero value. Thus in a mean field approximation near such a transition point ϕ can be decomposed into two parts, P the static mean field part and $\psi(\mathbf{q},\omega)$, the fluctuating part as follows,

$$\phi_q = P\delta(\mathbf{q},\omega) + \psi(\mathbf{q},\omega) \tag{3.2}$$

It is assumed that $< \psi(q,\omega) >= 0$. In this approximation our starting action (3.1) can be rewritten as

$$\begin{aligned}\mathcal{A} &= \frac{r}{2}P^2 + \frac{(u+v)}{4!}P^4 + \frac{1}{2\beta}\sum_q(\omega_n^2 + r + cq^2 + (u/2+v/6)P^2)\psi_q\psi_{-q} \\ &+P\frac{u}{3!\beta}\sum_{q_1,q_2}\psi_{q_1}\psi_{q_2}\psi_{-q_1-q_2} + \frac{u}{4!}\sum_{q_1,q_2,q_3}\psi_{q_1}\psi_{q_2}\psi_{q_3}\psi_{-q_1-q_2-q_3} \\ &+\frac{vL^{-d}}{4!\beta}\sum_{q_1,q_2}\psi_{q_1}\psi_{-q_1}\psi_{q_2}\psi_{-q_2}.\end{aligned} \tag{3.3}$$

Here we use the notation $\psi_{q_i} = \psi(\mathbf{q}_i,\omega_{n_i})$. It is to be noted that the term $P^2\psi_q\psi_{-q}$ has a coefficient $3u+v$ which can remain positive even when $u+v<0$ and it has important consequences which will be discussed later. Technically the long range part of the action with vertex v contributes to such term two possible ways whereas the short range part with vertex u contributes in six (C_2^4) possible ways and the difference lies in their range of interactions. With this action we can study the thermodynamics of the system by constructing a free energy which is defined as the logarithm of a functional integral over $\mathcal{A}(\psi,P)$, i.e.

$$\mathcal{F} = -\frac{1}{\beta}\log\left(\int \mathcal{D}\psi e^{-\mathcal{A}(\psi,P)}\right). \tag{3.4}$$

The value of P is to be determined by minimizing the free energy $\mathcal{F}$. Stability of a thermodynamic system requires the free energy to be positive. In a Landau theory, which neglects fluctuations completely, stability criteria requires the coefficient of the quartic term, i.e. $(u+v)$ to be positive. In that case, for $r>0$, the free energy

will be minimized for $P = 0$ resulting in a second order transition. On the other hand for $(u + v) < 0$ stability criteria in a mean field theory requires a higher order term with positive coefficient which results a first order transition with a non-zero $P \sim |u + v|$. We consider a limiting situation where $|u + v| \approx 0$ which corresponds to a weak first order transition. In this regime a proper account of the fluctuation corrections should be taken and it will be shown that fluctuation corrections alone can stabilize the system without invoking a higher order term in the starting action. We will discuss the effects of fluctuations in four point vertices near a weak first order transition in the next section.

3.3 Fluctuation corrections to the free energy at zero temperature

In the previous section we discussed importance of the coefficients of quartic term to determine the nature of phase transition. In a field theory description these coefficients are called vertex functions. Under certain circumstances they can get heavily renormalized by order parameter fluctuations and an weak first order transition is such an event. In this case there is a competition between order parameter fluctuations and a non-zero value of order parameter to stabilize a thermodynamic system. To quantify the effects of the competition between the order parameter fluctuations and a non-zero value of order parameter we calculate the fluctuation re-normalized four point vertex functions. Then a fluctuation renormalized free energy is constructed using them. We use renormalization group equations for four point vertices obtained in the lowest order perturbation theory. Such equations were derived earlier by Gadeker and Ramakrishnan[39, 40] in a *parquet approximation*. It is assumed that near a weak first order transition, a system acquires a small but non zero polarization P, the polarization fluctuation near such a phase transition becomes gapped, with the gap being proportional to P^2. Thus near such a phase transition the free optical phonon propagator which is the inverse of the coefficient of the quadratic term of the fluctuating part in the mean field action (eqn. (3.3)), is given by,

$$G^{-1}(q, \omega_n) = r + cq^2 + \omega_n^2 + (u/2 + v/6)P^2. \tag{3.5}$$

In a paraelectric phase with $u + v > 0$, $P = 0$. A bare theory predicts that at

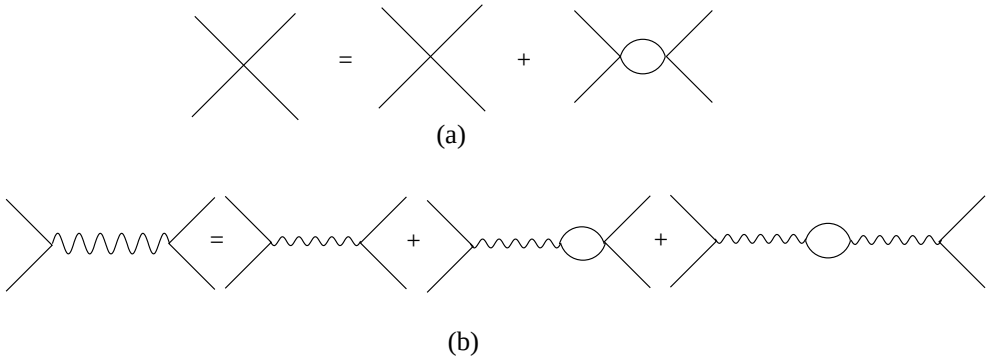

Figure 3.1: Parquet diagrams for the fluctuation corrections to the short interaction vertex u (a) and the long range vertex v(b) are shown at the lowest order. Here the curly line corresponds to the long range vertex and the solid line corresponds to the propagator given by the equation (3.5).

$T = 0$, the static susceptibility $\chi(0,0) \sim G(0,0) \sim 1/r$. Thus $r = 0$ is a point of instability in the paraelectric phase, and when there is no discontinuity in the order parameter ($u+v > 0$) at that point, it can be identified as a quantum critical point. In the vicinity of the critical point, correlation length becomes large, leading to dominance of the order parameter fluctuations. Moreover when $|u + v| \approx 0$, fluctuation corrections to the four point vertices become important. We will try to discuss the effects of fluctuations in the vicinity of the limit $|u + v| \to 0$, in developing spontaneous non-zero value of the order parameter near the transition point. Since in the case of a week first order transition, initially correlation length grows significantly, the bare vertices get strongly re-normalized. Now using the bare propagator for the order parameter fluctuation (eqn. (3.5)) we find that the leading order contribution from the second diagram of the figure 3.1(a) to the renormalization vertex function with a momentum cutoff Λ is given as,

$$\delta u = -u^2 \sum_n \int_0^\Lambda d^d q G^2(q, \omega_n). \tag{3.6}$$

At zero temperature the frequency summation becomes an integral and in this case, the combination of the frequency sum and the d-dimensional integral can be

replaced by a $d+1$ dimensional integral. Hence in three dimension,

$$\begin{aligned} \delta u &\sim -u^2 \int_0^{\Lambda} d^{3+1} q G^2(q) \text{ which} \\ &\sim -u^2 K_4 \log \frac{\Lambda}{(r + (u/2 + v/6)P^2)^{\frac{1}{2}}}. \end{aligned} \tag{3.7}$$

Here K_4 is a constant which is related to the surface area of a 4-dimensional sphere of unit radius. Surface area of a d-dimensional hyper-sphere is given by,

$$K_d = \left(2^{d-1}\pi^{\frac{d}{2}}\Gamma(d/2)\right)^{-1}. \tag{3.8}$$

$K_d = \frac{1}{8\pi^2}$ and $\frac{1}{2\pi^2}$ in d=4 and d=3 respectively. The correction to four point vertex δu has a logarithmic divergence as $r \to 0$ and $P \to 0$. We define the diverging logarithmic part as a new cut-off variable

$$x = \log \frac{\Lambda}{(r + (u/2 + v/6)P^2)^{\frac{1}{2}}}. \tag{3.9}$$

We define the re-normalized proper four point vertices Γ_4 and Δ_4, with their bare values given as,

$$\Gamma_4^0 = u \text{ and } \Delta_4^0 = v. \tag{3.10}$$

Considering the lowest order corrections, we get the following renormalization group equations in terms of the cut-off variable x,

$$\frac{d\Gamma_4}{dx} = -\frac{3}{2} K_{d+1} \Gamma_4^2(x), \tag{3.11}$$

$$\frac{d\Delta_4}{dx} = -K_{d+1}\Gamma_4(x)\Delta_4(x) - \frac{1}{6} K_{d+1}\Delta_4^2(x). \tag{3.12}$$

The above equations can also be obtained in a parquet re-summation scheme by summing leading order diagrams up-to infinite order as shown in figure 3.1. However the solutions of these equations can be written as,

$$\begin{aligned} \Gamma_4 &= \frac{\Gamma_4^0}{1 + \frac{3}{2} K_{d+1} \Gamma_4^0 x}, \\ \Delta_4 &= \frac{3\Gamma_4\Delta_4^0}{\Delta_4^0 + (3\Gamma_4^0 - \Delta_4^0)(1 + \frac{3}{2} K_{d+1}\Gamma_4^0 x)^{-1/3}}. \end{aligned} \tag{3.13}$$

In this derivation the contributions from the third order term, i.e. from $P\psi\psi\psi$ is neglected. In a perturbative theory, this term contribute nothing at the first order. It contributions to the higher order. But those corrections are less divergent compared to the contributions coming from quartic terms. It is to be noted that the cut-off variable x contains Γ_4 and Δ_4. Thus the set of equations (3.13) defines coupled equations for Γ_4 and Δ_4. They need to be solved self-consistently to find out their dependencies on r and P. From Γ_4 and Δ_4 thus obtained, we can calculate the fluctuation re-normalized free energy using the relation,

$$\frac{\partial^4 F}{\partial P^4} = \Gamma_4 + \Delta_4. \tag{3.14}$$

We need to integrate (with proper boundary conditions) the above equation four times with respect to P to get an expression for the free energy. Integrating the equation (3.14) once, we get

$$\begin{aligned} \frac{\partial^3 F}{\partial P^3} &= \int_0^P (\Gamma_4 + \Delta_4) dP' \\ &= P(\Gamma_4 + \Delta_4) + \int_0^P P' \frac{\partial}{\partial P'}(\Gamma_4 + \Delta_4) dP' + c(r). \end{aligned} \tag{3.15}$$

Here $c(r)$ is a constant independent of P and can be equated to zero using the symmetry constraint $\partial^3 F/\partial P^3|_0 = 0$. The second term in the right hand side of the above equation takes care of the P dependence of Γ_4 and Δ_4. To make our calculations simpler, we will neglect that term at this stage. Before doing so, we make an estimate of the corresponding error. The integral reads as,

$$\int_0^P \frac{2K_{d+1}(3\Gamma_4 + \Delta_4)^3 P'^2 dP'}{6r + [(3\Gamma_4 + \Delta_4) - \frac{K_{d+1}}{6}(3\Gamma_4 + \Delta_4)^2]P'^2}. \tag{3.16}$$

Here $K_{d+1} = \frac{1}{8\pi^2}$ in $d = 3$. In order to make our lowest order perturbation theory valid, we choose $(3\Gamma_4 + \Delta_4) \sim \mathcal{O}(10)$. Thus $K_{d+1}(3\Gamma_4 + \Delta_4) \sim \mathcal{O}(10^{-1})$. The contribution from the P-dependence of Γ_4 and Δ_4 to the integral(3.16) becomes,

$$\begin{aligned} & \frac{K_{d+1}}{3}(3\Gamma_4 + \Delta_4)^3 \int_0^P \frac{P'^2 dP'}{6r + (3\Gamma_4 + \Delta_4)P'^2} \\ \approx\ & \frac{K_{d+1}}{3}(3\Gamma_4 + \Delta_4)^2 P \approx 10^{-2}(3\Gamma_4 + \Delta_4)P. \end{aligned} \tag{3.17}$$

We see that, the P-dependence of Γ_4 and Δ_4 contributes two order of magnitude less compared to the other terms in the free energy. Thus we neglect the P dependence of Γ_4 and Δ_4 at this stage in calculating F. Integrating two more times, we get,

$$\frac{\partial F}{\partial P} = rP + \frac{1}{3!}(\Gamma_4 + \Delta_4)P^3. \tag{3.18}$$

To obtain a form of F suitable to describe the first order transition we need to retain the P dependence of Γ_4 and Δ_4 at this stage and thus,

$$\begin{aligned} F &= \int_0^P (rP' + \frac{1}{3!}(\Gamma_4 + \Delta_4)P'^3)dP' \\ &= \frac{1}{2}rP^2 + \frac{(\Gamma_4(P) + \Delta_4(P))}{4!}P^4 \\ &\quad -\frac{1}{4!}\int_0^P P'^4 \frac{d}{dP'}(\Gamma_4(P') + \Delta_4(P'))dP'. \end{aligned} \tag{3.19}$$

To evaluate the above integral, we make the following substitution(using eqn. (3.9))

$$P^2 = \frac{\Lambda e^{-x} - r}{\frac{\Gamma_4}{2} + \frac{\Delta_4}{6}}. \tag{3.20}$$

Contribution from the integral part in the previous equation is given by

$$\begin{aligned} &- \frac{1}{4!}\int_{\log(\Lambda/r)}^{\log \frac{\Lambda}{r+(\frac{\Gamma_4}{2}+\frac{\Delta_4}{6})P^2}} \left(\frac{\Lambda e^{-x} - r}{(\frac{\Gamma_4}{2} + \frac{\Delta_4}{6})}\right)^2 \times \frac{d}{dx}(\Gamma_4(x) + \Delta_4(x))dx \\ &= \frac{K_{d+1}}{4}\int_{\log(\Lambda/r)}^{\log \frac{\Lambda}{r+(\frac{\Gamma_4}{2}+\frac{\Delta_4}{6})P^2}} (\Lambda^2 e^{-2x} - 2r\Lambda e^{-x} + r^2)dx \\ &= \frac{K_{d+1}}{4}\left(\frac{r}{2}(\Gamma_4 + \Delta_4)P^2 - \frac{1}{2}\left(\frac{\Gamma_4}{2} + \frac{\Delta_4}{6}\right)^2\right)P^4 \\ &\quad + r^2 \log \frac{r}{r + (\frac{\Gamma_4}{2} + \frac{\Delta_4}{6})P^2}). \end{aligned} \tag{3.21}$$

Thus we get the following expression for the free energy at zero temperature

$$\begin{aligned} F &= \frac{K_{d+1}}{4}r^2 \log \frac{6r}{6r + (3\Gamma_4 + \Delta_4)P^2} + \frac{1}{2}rP^2\left(1 + \frac{K_{d+1}}{2}\left(\frac{\Gamma_4}{2} + \frac{\Delta_4}{6}\right)\right) \\ &\quad + P^4\left(\frac{\Gamma_4 + \Delta_4}{4!} - \frac{K_{d+1}}{8}\left(\frac{\Gamma_4}{2} + \frac{\Delta_4}{6}\right)^2\right). \end{aligned} \tag{3.22}$$

where Γ_4 and Δ_4 are P-dependent and are to be determined from the set of equations (3.11,3.12). We notice that only the following combinations of Γ_4 and Δ_4 appear in all the calculations,

$$\gamma_1 = 3\Gamma_4 + \Delta_4, \gamma_2 = \Gamma_4 + \Delta_4. \tag{3.23}$$

Here the bare value of γ_2 (γ_2^0)is the coefficient for the quartic term in mean field approximation. On the other hand non-zero polarization enters into the fluctuation propagator with a coupling constant γ_1. In case of $\gamma_2^0 < 0$, a mean field picture requires an additional $|P|^6$ term with positive coefficient for the stability of the system. However a fluctuation corrected scenario can ensure stability without such a term, provided $\gamma_1 > 0$. With the above definitions, the set of equations (3.13) becomes a single self-consistent equation for γ_1

$$\gamma_1 = \frac{3\Gamma_4^0}{1+\frac{3}{2}K_{d+1}\Gamma_4^0 x}\left(1+\frac{\Delta_4^0}{\Delta_4^0+(3\Gamma_4^0-\Delta_4^0)(1+\frac{3}{2}K_{d+1}\Gamma_4^0 x)}\right). \tag{3.24}$$

Here x contains γ_1 only. Solving the above equation, γ_2 can be found from (eqn. 3.12)

$$\frac{d\gamma_2}{dx} = -\frac{K_{d+1}}{6}\gamma_1^2. \tag{3.25}$$

Below $\gamma_2 = 0$, phase transition in this system will be first order. We are interested in the phase transition near $\gamma_2 = 0$, where fluctuation effects in four point vertices are important. If we limit ourselves to the region $|\Delta_4| < 3\Gamma_4$, the leading order behavior of γ_1 is same as that of Γ_4 and is given by

$$\gamma_1 \approx \frac{3\Gamma_4^0}{1+\frac{3}{2}K_{d+1}\Gamma_4^0 x} = \frac{3\Gamma_4^0}{1+\frac{3}{2}K_{d+1}\Gamma_4^0 \log\frac{6\Lambda}{\sqrt{6r+\gamma_1 P^2}}}. \tag{3.26}$$

Assuming the bare value $\Gamma_4^0 \sim \mathcal{O}(10)$, so that $\frac{\Gamma_4^0}{4!} < 1$ which validates perturbation theory, gives $6\Gamma_4^0 \sim \mathcal{O}(10^2)$. If we define a parameter $a = \frac{3}{4}K_{d+1}\Gamma_4^0$, then within the validity regime of perturbation theory $a \sim \mathcal{O}(10^{-1})$. Thus for an wide range of γ_1 e. g. $\gamma_1 \sim \mathcal{O}(1) - \mathcal{O}(10^2)$, we find $|a\log\gamma_1| < |\frac{3\Gamma_4^0}{\gamma_1}|$. For small r, γ_1 and γ_2

can be estimated as

$$\gamma_1 \sim \frac{3\Gamma_4^0}{1 - a\log(\gamma_1 P^2)} \approx \frac{3\Gamma_4^0}{1 - 2a\log P}. \tag{3.27}$$

This is essentially a non-self-consistent solution for γ_1. Variation of γ_1 at zero temperature is shown in figure 3.2. From the figure it is visible that, at $P = 0$ because of the quantum critical fluctuations there is a strong reduction of γ_1 from its bare value and a non-zero P restores it to its bare value. A non-zero polarization has similar effects on the γ_2 which leads to a first order transition.

From the equation (3.25), we get,

$$\gamma_2 = \gamma_2^0 + (-9(\Gamma_4^0)^2 + \frac{\gamma_1}{3}) \tag{3.28}$$

$$= \gamma_2^0 - \frac{9(\Gamma_4^0)^2 K_{d+1}}{6a}\left(1 - \frac{1}{1 - 2a\log P}\right) \tag{3.29}$$

where γ_2^0 is the bare value of γ_2. Since the corrections due to self-consistency $\sim \log\gamma_1$, the above estimate breaks down near $P \sim \exp(1/2a)$. Except in that regime, the non-self-consistent result is expected to give good result.

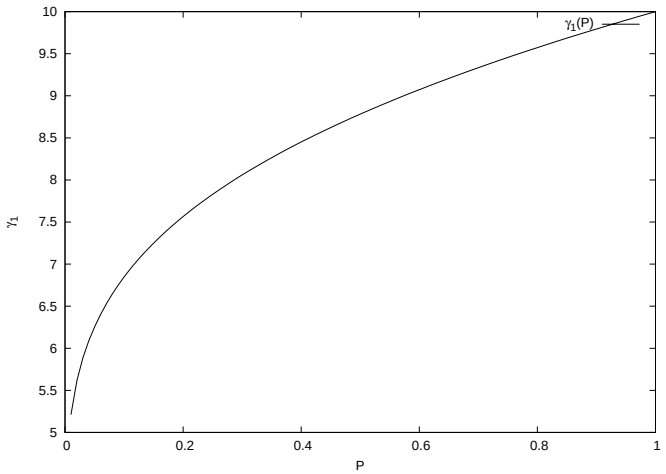

Figure 3.2: Asymptotic evolutions of γ_1 with P at $T = 0$. Parameter values are chosen as $3\Gamma_4^0 = 10$ and $a = 0.1$ in an arbitrary scale.

Free energy at zero temperature: Using the asymptotic behavior of the four point vertices (eqn. (3.27)) and using equation (3.22) defining $\tilde{\gamma}_2^0$ as $\gamma_2^0 - \frac{9(\Gamma_4^0)^2 K_{d+1}}{6a}$, we can write the following asymptotic expression for the free energy at

zero temperature

$$F = \frac{K_{d+1}}{4} r^2 \log \frac{6r}{6r + P^2 \frac{3\Gamma_4^0}{1-2a\log P}} + \frac{1}{2} r P^2 \left(1 + \frac{K_{d+1}}{12} \frac{6\Gamma_4^0}{1-2a\log P}\right) + \frac{P^4}{4!}\left(\left(\tilde{\gamma}_2^0 + \frac{K_{d+1}}{6a}\left(\frac{9(\Gamma_4^0)^2}{1-2a\log P}\right)\right) - \frac{4! K_{d+1}}{36\times 8}\left(\frac{3\Gamma_4^0}{1-2a\log P}\right)^2\right) \quad (3.30)$$

Here the first term $\sim \mathcal{O}(r^2)$, hence negligible compared to the other terms in the vicinity of a quantum critical point($r \to 0$). The second term is a standard quadratic term with fluctuation corrections. The third term, describes the appropriate physics of the problem. The coefficient of the quartic term contains three terms. First one is a constant and can take either positive or negative but small values. Now there is a competition between the second and the third term. The second term tries to make the free energy positive while the third term tries to make it negative. However unless γ_2^0 is a sufficiently large negative number, coefficient of the quartic term is positive in the parameter regime of our interest.

To have a phase transition, the following equation must be satisfied,

$$\frac{\partial F}{\partial P} = rP + \frac{1}{3!}(\Gamma_4 + \Delta_4) P^3 = 0 \quad (3.31)$$

For positive $r \approx 0$, the above equation can be satisfied only if $\Gamma_4 + \Delta_4$ is negative. Corresponding value of the P is given as,

$$P_0 = \exp\left(\frac{1}{2a} + \frac{9K_{d+1}(\Gamma_4^0)^2}{12a\tilde{\gamma}_2^0}\right) = \exp\frac{1}{2a}(1+\mu) \quad (3.32)$$

with $\mu = \frac{9K_{d+1}(\Gamma_4^0)^2}{6\gamma_2^0}$. Since $|\mu| >> 1$, the parameter $\tilde{\gamma}_2^0 > 0$ corresponds to the very large values of P_0. Since the scheme presented here, is valid for small P, we exclude this possibility in this discussion. On the other hand, $\tilde{\gamma}_2^0 < 0$ corresponds to the finite value of P_0 and there is a possibility of a first order transition at $r = r_0 > 0$. As $r \sim P^2$, $r_0 \sim \exp\frac{1}{a}(1+\mu)$. This is in sharp contrast to the mean-field prediction. In the later case a discontinuity in the order parameter is predicted to be $\sim \gamma_2^0 = |u + v|$, while such discontinuity has a non-analytic dependence on γ_2^0 in a fluctuation induced and order parameter limited transition in our theory. If we consider the Gaussian thermal fluctuations near the instability point and

neglect the temperature dependencies of the vertices, then the thermal corrections to r and $P^2 \sim T^2$ and P_0^2 should vanish above a temperature $T \sim \exp \frac{1}{2a}(1+\mu)$. However the fluctuation cut-off for the renormalized vertices and hence the form of fluctuation re-normalized free energy should get changed at finite temperature. In the next section we will discuss the finite temperature case in detail.

3.4 Fluctuation corrections to the free energy at finite temperature

Figure 3.3: Diagrammatic representation of gap renormalization up-to one loop.

Here we consider the system to be at a low but non-zero temperature as well as near a mean field quantum critical point with a zero temperature negative gap(i. e. $r = -r_0$, $r_0 \geq 0$). Near $r_0 = 0$, fluctuation corrections at finite temperature to it leads to

$$r(T) = -r_0 + K_3 \left(\frac{u}{2} + \frac{v}{6}\right) T^2. \tag{3.33}$$

Here the thermal fluctuations are considered up-to the Gaussian level as done in the chapter 2. Above expression for $r(T)$ without any correction to four point vertices predicts a second order transition with a transition temperature $T_c \sim \sqrt{r_0}$ for $u + v > 0$. Again we will look at the correction to the four point vertices in the limit $|u + v| \to 0$ but at non-zero temperature. Using the same procedure as used for the zero temperature case, we now deduce the parquet equations for the four point vertices and hence the free energy at finite temperature. At non-zero temperature, fluctuation corrections to the four point vertices takes the form

$$\begin{aligned} \delta u &\sim -u^2 T \int_0^{\Lambda} d^3 q G^2(q,\ \omega_n = 0) \\ &\sim -u^2 K_3 \frac{T}{(r(T) + (u/2 + v/6)P^2)^{\frac{1}{2}}}, \end{aligned} \tag{3.34}$$

in three dimension. Near the critical point, $r(T) \sim r + K_3(u/2+v/6)T^2$. Thus at finite temperature, we can define finite temperature fluctuation cut-off as

$$x_T = \frac{T}{(r+(u/2+v/6)(P^2+K_3T^2))^{\frac{1}{2}}}. \tag{3.35}$$

In this case, fluctuation corrections to the free energy in terms of γ_1 and γ_2 (equation 3.19) becomes,

$$\begin{aligned} I_{fluc} &= -\frac{1}{4!}\int_{x_T(0)}^{x_T(P)} \left(\frac{T^2/x^2 - r}{\gamma_1/6} - K_3T^2\right)^2 \frac{d\gamma_2(x)}{dx}dx \\ &= \frac{1}{4}[T[(r+\frac{\gamma_1}{6}K_3T^2)^{\frac{3}{2}} - (r+\frac{\gamma_1}{6}(P^2+K_3T^2))^{\frac{3}{2}}] \\ &\quad +r^2[(r+\frac{\gamma_1}{6}K_3T^2)^{-\frac{1}{2}} - (r+\frac{\gamma_1}{6}(P^2+K_3T^2))^{-\frac{1}{2}}] \\ &\quad +T^4\gamma_2^2[(r+\frac{\gamma_1}{6}K_3T^2)^{-\frac{1}{2}} - (r+\frac{\gamma_1}{6}(P^2+K_3T^2))^{-\frac{1}{2}}] \\ &\quad -Tr[(r+\frac{\gamma_1}{6}K_3T^2)^{\frac{1}{2}} - (r+\frac{\gamma_1}{6}(P^2+K_3T^2))^{\frac{1}{2}}] \\ &\quad +2rT^2\gamma_2[(r+\frac{\gamma_1}{6}K_3T^2)^{-\frac{1}{2}} - (r+\frac{\gamma_1}{6}(P^2+K_3T^2))^{-\frac{1}{2}}] \\ &\quad +T^4\gamma_2[(r+\frac{\gamma_1}{6}K_3T^2)^{-\frac{1}{2}} - (r+\frac{\gamma_1}{6}(P^2+K_3T^2))^{-\frac{1}{2}}]]. \end{aligned} \tag{3.36}$$

Here $x_T(P) = \frac{T}{(r+\frac{\gamma_1}{6}(P^2+K_3T^2))^{\frac{1}{2}}}$ is used as polarization dependent fluctuation cut-off at non-zero temperature. In performing the above integral, x_T-dependence of Γ_4 and Δ_4 are neglected as that would lead to sub leading corrections. For the systems near $r \to 0$ limit, retaining only the terms lowest order in T and r, the free energy can be truncated as

$$\begin{aligned} F &= \frac{1}{2}rP^2 + P^4\left(\frac{\Gamma_4+\Delta_4}{4!}\right) + \frac{T}{4}[\left(r + K_3(\frac{\Gamma_4}{2}+\frac{\Delta_4}{6})T^2\right)^{\frac{3}{2}} \\ &\quad - \left(r+(\frac{\Gamma_4}{2}+\frac{\Delta_4}{6})(P^2+K_3T^2)\right)^{\frac{3}{2}}]. \end{aligned} \tag{3.37}$$

For small r and T, the third term is of the $\mathcal{O}(T^4)$, hence is negligible. Thus the free energy in the leading order can be further truncated as

$$F = \frac{r}{2}P^2 + P^4\left(\frac{\Gamma_4+\Delta_4}{4!}\right) - \frac{T}{4}\left(\frac{\Gamma_4}{2}+\frac{\Delta_4}{6}\right)^{\frac{3}{2}}P^3. \tag{3.38}$$

In the above equation $P \equiv |\vec{P}|$ and the cubic term which is a result of small P expansion, does not violate the symmetry of the problem. A finite temperature version of the equation (3.27), i.e. the asymptotic form of the self consistent equation for γ_1, one of the important combinations of the four point vertices reads,

$$\gamma_1 = \frac{6\Gamma_4^0}{1 - \frac{6aT}{(\gamma_1(P^2+K_3T^2))^{1/2}}}. \tag{3.39}$$

Non-zero solution of the above equation tells

$$\gamma_1 = \frac{18a^2T^2 + K_3\Gamma_4^0T^2 + \Gamma_4^0P^2}{P^2 + K_3T^2} \pm \frac{6aT\sqrt{9a^2T^2 + \Gamma_4^0P^2 + \Gamma_4^0K_3T^2}}{P^2 + K_3T^2}. \tag{3.40}$$

In deriving the equations at finite temperature we have limited ourselves in the low temperature region i.e. a^2T^4 is neglected compared to K_3T^2. Moreover γ_1 should be strongly suppressed due to critical fluctuations at zero P and should go towards its bare value with increasing P. Hence the part is remaining,

$$\gamma_1 \approx \Gamma_4^0 - \frac{6a\sqrt{\Gamma_4^0}T}{\sqrt{P^2 + K_3T^2}}. \tag{3.41}$$

Asymptotic evolution of γ_1 with P for two different temperature is shown in figure 3.4. In this figure we find that qualitative nature of the curve is similar to that of the zero temperature case except that at $P = 0$, reduction of γ_1 at a finite temperature is lower than that of it at a zero temperature. Since at a finite temperature the quantum critical fluctuations are suppressed, this is an expected result.

However from equation (3.28) we get

$$\gamma_2 = \gamma_2^0 + \left(\frac{2\Gamma_4^0}{3} + \frac{2a\sqrt{\Gamma_4^0}T}{\sqrt{P^2 + K_3T^2}}\right). \tag{3.42}$$

Free energy at finite temperature: Again using the asymptotic behavior of the four point vertices (3.41) and using equation (3.38), we can write the following

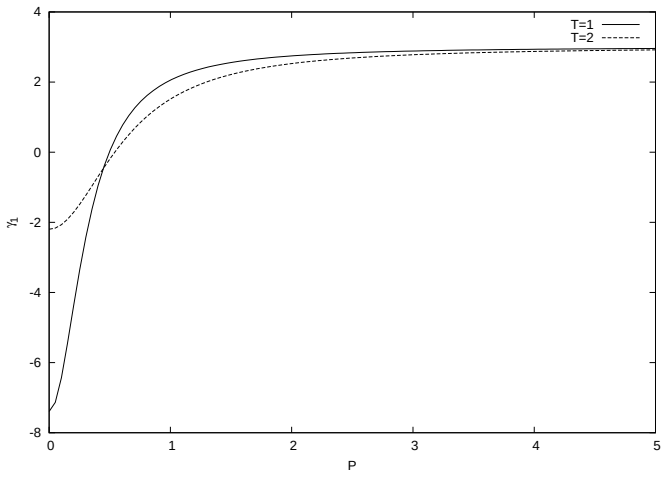

Figure 3.4: Asymptotic evolution of γ_1 with P at $T \neq 0$. Two curves are drawn at two different temperatures with $\Gamma_4^0 = 10/3$, $a = 0.1$ and $K_d = 0.1$.

asymptotic expression for the free energy at non zero temperature

$$\begin{aligned} F &= \frac{r(T)}{2}P^2 + \frac{P^4}{4!}\left(\gamma_2^0 + \left(\frac{2\Gamma_4^0}{3} + \frac{2a\sqrt{\Gamma_4^0}T}{\sqrt{P^2+K_3T^2}}\right)\right) \\ &\quad -\frac{T}{4}P^3\left(\frac{\Gamma_4^0}{6} - \frac{a\sqrt{\Gamma_4^0}T}{\sqrt{P^2+K_3T^2}}\right)^{\frac{3}{2}}. \end{aligned} \tag{3.43}$$

Gap renormalization up-to one loop(figure 3.3) at finite temperature near $r = 0$, tells,

$$r(T) = -r_0 + K_3\left(\frac{\Gamma_4^0}{6} - \frac{a\sqrt{\Gamma_4^0}T}{\sqrt{P^2+K_3T^2}}\right)T^2. \tag{3.44}$$

Neglecting the term of $\mathcal{O}(T^{-3})$ in the coefficient of P^2, the expression for free energy becomes

$$\begin{aligned} F &= \frac{1}{2}\left(-r_0 + \frac{K_3\Gamma_4^0}{6}T^2\right)P^2 \\ &\quad +\frac{1}{4!}P^4\left(\gamma_2^0 + \left(\frac{2\Gamma_4^0}{3} + \frac{2a\sqrt{\Gamma_4^0}T}{\sqrt{P^2+K_3T^2}}\right)\right) \\ &\quad -\Gamma_4^0\frac{T}{4}P^3\left(\frac{1}{6} - \frac{3aT}{2\sqrt{\Gamma_4^0(P^2+K_3T^2)}}\right). \end{aligned} \tag{3.45}$$

In the limit $P^2 >> K_3T^2$, it takes the form[1]

$$F = \frac{1}{2}\left(-r_0 + \frac{K_3\Gamma_4^0}{6}T^2 + \frac{3aT^2\sqrt{\Gamma_4^0}}{2}\right)P^2 - \frac{T}{24}P^3\left(\Gamma_4^0 - \frac{a}{2}\sqrt{\Gamma_4^0}\right) + \left(\frac{\frac{2\Gamma_4^0}{3} + \gamma_2^0}{4!}\right)P^4. \quad (3.46)$$

Above equation tells that, the solution of the equation $\partial F/\partial P = 0$ will result a nonzero value of $P_0 \sim T_0$, even if $\gamma_2^0 > 0$. Here T_0 is the second order transition temperature for the mean field theory. Since near quantum critical point, $T_0 \sim \sqrt{r_0}$, P_0 is also $\sim \sqrt{r_0}$. If we compare this results with that of the zero temperature case, we see that in the finite temperature case, the discontinuity in the order parameter near the transition point $\sim \sqrt{r_0}$ while it is independent of r_0 in the zero temperature case. Since r_0 is the smallest scale(near mean field quantum critical point, $r_0 \to 0$) in this system, first order transition as a result of the order parameter fluctuations at finite temperature near the mean field quantum critical point is weaker than that of the zero temperature case. This result is consistent with some experimental fact as reported in [37] and established the importance of the quantum critical fluctuations in this regards.

3.5 Discussions

Occurrence of first order transition due to coupling between the order parameter fluctuations and soft modes is a well studied problem in many classical systems[42, 43, 44, 45] and recently studied in context of quantum phase transitions in electronic systems[46]. To capture the basic physics near a weak first order transitions, one needs to study the effects of fluctuations on the proper four point vertices. Bare perturbation calculations in this system show vertex corrections to be logarithmically singular at zero temperature and with zero polarization in three dimensions due to quantum critical fluctuations. To include the effects of the singular contributions, we use the lowest order renormalization group equations to derive a set

[1]This assumption does not contradict K_3T^2 contribution to the gap renormalization. Such contribution appeared with the assumption that the lowest non-zero Matsubara frequency, i.e. $2\pi T > \sqrt{\frac{\gamma_1}{6}}P$. This assumption holds good even if $P^2 >> K_3T^2$.

of recursion relations for four point vertices. Moreover to stabilize the system a non-zero polarization is assumed. Here the re-normalized vertices crucially depend on the non-zero polarization. Using these relations, the expressions for the free energy both at zero temperature and at finite temperature are derived. The relevant quantities like transition temperature and the discontinuity in the order parameter at the transition point turn out to be small but finite. We mainly concentrate on the phenomena near a *quantum critical point* predicted by a mean field theory. We found stronger possibility of first order transitions at $T = 0$ than at any finite temperature transition near quantum critical point which is in accord with recent experimental finding. From our analysis it is clear that this fact can be attributed to the critical fluctuations near a quantum critical point. It is found that a finite polarization is required to suppress the effects of the critical fluctuations. Since the effects of quantum critical fluctuations get reduced at a finite temperature, a lower value is sufficient to stabilize the system. Thus the phenomena observed in the experiment[37] is clearly a consequence of the quantum critical fluctuations. Apart from that, unlike the standard renormalization group approach, which considers first order transition as just the inability of the system to reach an unstable fixed point, the present approach makes qualitative predictions about the magnitude of the discontinuity in order-parameter near the transition point. Similar phenomena occurs in low T_c itinerant magnets because of the coupling between order parameter and other soft modes[47]. The effects of fluctuations in the discontinuity in order-parameter has been estimated earlier using different theoretical set up, namely loop corrections to the free energy, hence an non-analytic Landau expansion. In those works the occurrence of first order transition at low temperature is found to be more susceptible to the effects of generic scale invariance than their classical counterparts. We have considered the constant pressure case only, where strain fluctuations are completely integrated out. One can also consider a situation where system volume is constant[48] and a proper quantum generalization of such case should certainly be addressed.

4

Quantum criticality in magnetic quantum paraelectrics

4.1 Introduction

In this chapter we focus on some novel behavior of $EuTiO_3$ at low temperature. This material can certainly be considered as a good addition in the list of quantum paraelectric materials exhibiting macroscopic quantum phenomena in ferroelectrics and multiferroics. As far as the structure and the gross features of the static dielectric behavior are concerned, this material is similar to other quantum paraelectrics with perovskite structure, like $SrTiO_3$, $KTaO_3$, etc. In the study of quantum criticality in ferroelectrics, $EuTiO_3$ certainly adds a new dimension. This material contains Eu ions with spin 7/2, and undergoes anti-ferromagnetic order at $T_N \sim 5.3K$[49]. As one lowers the temperature a sharp decrease in the static dielectric susceptibility is observed below the Neel temperature. Moreover presence of an external magnetic field reduces the suppression of the dielectric constant by Neel order. At a critical value of the external magnetic field $\sim$ 1Tesla, which suppresses the effects of the Neel order completely, the static dielectric constant of this material attains a quantum paraelectric behavior, with $\epsilon_0 \sim \mathcal{O}(10^2)$ at zero temperature[50]. The dielectric susceptibility starts getting saturated at a crossover temperature (defined in chapter 2 and ref. [23]) $\sim 30K$. An experimentally observed dielectric behavior of this material is shown in figure 4.1. Thus it is almost evident that the magnetic order couples to the polarization fluctuations in this material. In previous theoretical attempts, a mean field theory with such

a scenario[51, 52] was found quite successful in describing many aspects of the dielectric properties of this system. Like other quantum paraelectrics, electric polarization in this material is due to the variations of Ti-O bond-lengths from their equilibrium values. However the collective behavior of such interacting stretched bonds does not lead to a ferroelectric state even at zero temperature. Let us now analyze the above experimental findings in context of quantum phase transition in ferroelectrics as discussed in our second chapter. In that line of thought, we see that $EuTiO_3$ has a much lower value of static dielectric constant at low temperature than other quantum paraelectrics and thus it is far away from a ferroelectric quantum critical point than them. However this material can be tuned to a ferroelectric quantum critical point by changing some non-thermal parameter, such as replacement of a certain amount of O^{16} by O^{18}. On the other hand its Neel temperature is low enough to consider it as near a anti-ferro magnetic quantum critical point. Thus it is quite sensible to speculate that this material can be a good play ground for observing an interplay of two different kind of critical fluctuations at low temperature.

In this chapter we consider a non-thermal parameter tuned $EuTiO_3$ near both

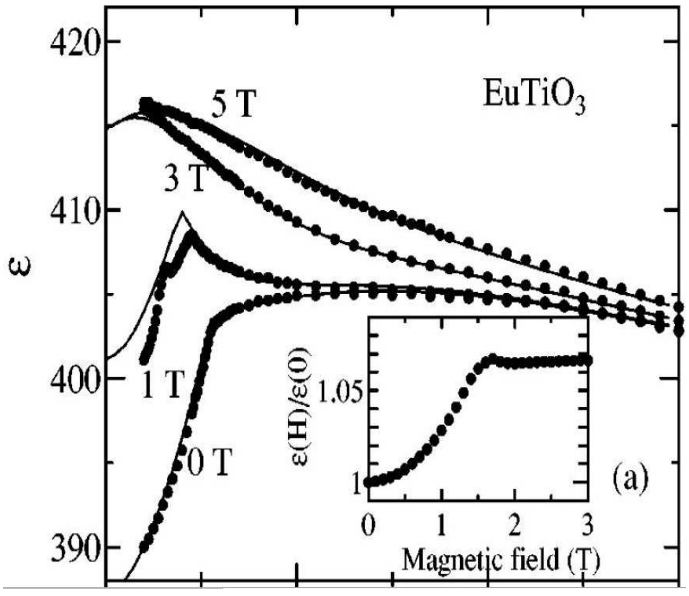

Figure 4.1: Variation of the static dielectric constant with temperature in $EuTiO_3$ as found in the experiment at different values of the uniform external magnetic field.

ferroelectric and anti-ferromagnetic quantum critical points. The ferroelectric sub system is considered as displacive type, i.e. corresponding order parameter ($\vec{\phi}$) fluctuations are represented by the fluctuations in two transverse optic branches. Ferroelectric transition in this case is due to the softening of the optic phonons at the zone center. The effects of dipolar interaction is considered as the stiffening of longitudinal branch, and thus longitudinal fluctuations are not taken into account. Magnetic sector, i.e. the collection of interacting Eu spins ($\vec{S}$) in a cubic perovskite environment and in absence of external magnetic field, is represented by Heisenberg anti-ferromagnetic order parameter ($\vec{m}$) which is a vector spin with three components with short range interactions. In presence of an external magnetic field a ferromagnetic component will grow and will compete with the anti-ferromagnetic component to restore the quantum paraelectric behavior. Following the previous theoretical works[51, 52], we consider a coupling of the form $-\frac{w}{2}|\vec{\phi}|^2|\vec{S}|^2$ with coupling constant $w > 0$, between them and focus on the dependencies of the static dielectric susceptibility on temperature and external magnetic field below T_N. In earlier works which was directed towards explaining experimental findings in this material, the effects of magneto-electric coupling on the thermodynamic behavior of this material has been described at off-critical regime without considering quantum fluctuations. No attention has been paid to the possible quantum critical behavior of this system. We will try to explore the static dielectric behavior in this material near both the anti-ferromagnetic quantum critical point and ferroelectric quantum critical point regime. A scheme for systematic analysis of quantum fluctuations in this regime is proposed and the possibility of realizing such a limit in experiments will be discussed in the following sections.

4.2 Mean field theory

We consider this material in the vicinity of both anti-ferromagnetic and ferroelectric quantum critical points. In this regime both the magnetic fluctuations and the paraelectric fluctuations can be described by a continuum theory. To study its dielectric properties in this regime, a Landau-Ginsburg-Wilson action for a magnetic quantum paraelectric system, in terms of the sub-lattice magnetization $\vec{m}$ and the

electric polarization $\vec{\phi}$ (soft mode coordinates) can be written in the following form,

$$\begin{aligned}\mathcal{A} &= \int d^dx \int_0^\beta d\tau [\frac{r_e}{2}(\vec{\phi}\cdot\vec{\phi}) + (\partial_\tau\vec{\phi})^2 + \frac{c_e}{2}(\nabla\cdot\vec{\phi})^2 + \frac{u}{2}(\vec{\phi}^2)^2 \\ &+\frac{r_m}{2}(\vec{m}\cdot\vec{m}) + (\partial_\tau\vec{m} - i\vec{h}\times\vec{m})^2 + \frac{c_m}{2}(\nabla\cdot\vec{m})^2 + \frac{v}{2}(\vec{m}^2)^2 \\ &-\frac{w}{2}|\vec{\phi}|^2|\vec{m}|^2]. \end{aligned} \tag{4.1}$$

Here τ is the imaginary time, β is the inverse temperature, and $\vec{h}$ is the applied external uniform magnetic field. Above action contains a dielectric part which is identical to one used in our study of quantum criticality in ferroelectrics in the chapter 2. The magnetic part of the action is derived with the consideration of small ferro-magnetic component which can be integrated out, as well as the bipartite structure of the $EuTiO_3$ lattice that supports a Neel order below the transition point. The detailed derivation of the anti-ferro magnetic part is given in reference [5]. In three dimension topological terms associated with quantum anti-ferromagnetic fluctuations are not important and hence neglected. The coupling between the staggered-magnetization and the uniform magnetic field has some important consequences and hence its origin deserves some comments. At a microscopic level, an uniform magnetic field couples only to the uniform component of a Heisenberg spin. If we invoke a continuum description and integrate out the uniform component with a constraint of vanishing scalar product between the uniform and the staggered components, such term results. Here r_e and r_m are the non-thermal parameters which can be tuned to zero to have ferroelectric and anti-ferromagnetic instabilities respectively. The coupling constants of the quartic terms are positive, i. e. u, $v > 0$, to ensure the stability of the system. Since in a quantum picture, statics and dynamics are coupled, an applied magnetic field induced precession of the magnetic vectors also play an important role in the study of phase transitions in quantum magnets. Now we start our discussions with the following mean field approximations.

$$\begin{aligned}\vec{m}(\vec{q},\omega) &= m_0\hat{z}\delta(\vec{q})\delta(\omega) + \vec{m}'(\vec{q},\omega), \\ <\vec{m}'> &= 0, <\vec{\phi}>=0 \end{aligned} \tag{4.2}$$

Here $\vec{m}(\vec{q},\omega)$ is the Fourier transform of $\vec{m}(\vec{x},\tau)$. The above approximations, along with the quasi-harmonic decoupling of the quartic terms as done in chapter 2, lead to the following mean field action,

$$\begin{aligned}\mathcal{A}_{MF} &= \frac{r_m}{2}m_0^2+\frac{v}{2}m_0^4 \\ &+\int d^dq\frac{1}{\beta}\sum_n[(\frac{r_e}{2}+\omega_n^2+\frac{c_eq^2}{2}+\frac{u}{2}\lambda_e)(\vec{\phi}\cdot\vec{\phi})+(\omega_n\vec{m}^{'}-i\vec{h}\times\vec{m}^{'})^2 \\ &+(\frac{c_mq^2}{2}+\frac{r_m}{2}+\frac{v}{2}(2m_0^2+\lambda_m))(\vec{m}^{'}\cdot\vec{m}^{'}) \\ &-\frac{w}{2}(m_0^2+\vec{m}^{'}\cdot\vec{m}^{'})\vec{\phi}\cdot\vec{\phi}]. \end{aligned} \tag{4.3}$$

In the above expression we use the following notations,

$$\begin{aligned}\vec{X}\cdot\vec{X} &= \vec{A}(\vec{q},\omega_n)\cdot\vec{A}(-\vec{q},\omega_n), \\ \lambda_{(e,m)} &= \int d^dq\frac{1}{\beta}\sum_n\chi_{(e,m)}(\vec{q},\omega_n), \\ \chi_e(\vec{q},\omega_n) &= <\vec{\phi}(\vec{q},\omega_n)\cdot\vec{\phi}(-\vec{q},\omega_n)>, \\ \chi_m(\vec{q},\omega_n) &= <\vec{m}^{'}(\vec{q},\omega_n)\cdot\vec{m}^{'}(-\vec{q},\omega_n)>. \end{aligned} \tag{4.4}$$

Zero magnetic field ($h=0$): In a zero external magnetic field, there is no ferromagnetic component and there is a competition between paraelectric fluctuations and anti-ferromagnetic fluctuations. The self consistent equations for polarization and magnetic fluctuations are,

$$\begin{aligned}\chi_e(\vec{q},\omega_n) &= <\vec{\phi}(\vec{q},\omega_n)\cdot\vec{\phi}(-\vec{q},\omega_n)> \\ &= \frac{1}{\frac{r_e}{2}+\frac{c_eq^2}{2}+\omega_n^2-\frac{w}{2}(m_0^2+\lambda_m)+\frac{u}{2}\lambda_e}, \end{aligned} \tag{4.5}$$

and

$$\begin{aligned}\chi_m(\vec{q},\omega_n) &= <\vec{m}^{'}(\vec{q},\omega_n)\cdot\vec{m}^{'}(-\vec{q},\omega_n)> \\ &= \frac{1}{\frac{r_m}{2}+\frac{c_mq^2}{2}+\omega_n^2-\frac{w}{2}\lambda_e+\frac{v}{2}(2m_0^2+\lambda_m)} \end{aligned} \tag{4.6}$$

respectively. The above two equations should be supplemented by the following expression for the *magnetic free energy* (within one loop correction) to determine

m_0 in the magnetically ordered phase,

$$f_m = \frac{r_m}{2}(\vec{m}_0)^2 + \frac{v}{2}|\vec{m}_0|^4 - \frac{1}{2}Tr\log(\chi_m(\vec{q},\omega_n)). \tag{4.7}$$

We need to know m_0, λ_e, and λ_m as a function of the temperature at various values of the system parameters, using equations (4.5-4.7). The extremization of f_m with respect to m_0 gives,

$$r_m m_0 + 2vm_0^3 + vm_0 \int d^dq \frac{1}{\beta}\sum_n \chi_m(\vec{q},\omega_n) = 0. \tag{4.8}$$

Non-zero solution of m_0 reads as,

$$m_0^2 = \frac{-r_m - v\lambda_m}{2v}. \tag{4.9}$$

Here we emphasize that at the critical value of the magnetic field, where the magneto-electric coupling is believed to be very small, the static dielectric constant for $EuTiO_3$ reaches a value $\mathcal{O}(10^2)$. Thus according to the classifications of various quantum paraelectrics the ferroelectric subsystem falls into the category of the *gaped quantum paraelectrics*, and is much more away from the ferroelectric quantum critical point than $SrTiO_3$. Such a dielectric state is characterized by a *crossover temperature* $T^* \sim \sqrt{r_e}$ as explained in chapter 2. For pure $EuTiO_3$, we see that the ferroelectric crossover temperature T^* is much higher than T_N, the Neel temperature. Low T_N implies that, the system is in the vicinity of anti-ferromagnetic quantum critical point. Owing to the large T^*, λ_e the fluctuation corrections to the ferroelectric gap is temperature independent. Thus the temperature dependence in the static dielectric constant at low temperature comes only from the magnetic fluctuations through magneto-electric coupling. We consider the temperature dependence of λ_m near as well as away from the magnetic critical point. Near a anti-ferro magnetic quantum critical point momentum cut-off becomes temperature dependent. Since in this material the dispersion relation for anti-ferromagnetic fluctuations is similar to that of the ferroelectric one the momentum cut-off at the critical point also $\sim T$. Thus within a non-self consistent

estimate we get,

$$\lambda_m \sim \begin{cases} T\int_0^{T/\sqrt{c_m}} \frac{q^2 dq}{c_m q^2} = c_m^{-\frac{3}{2}} T^2 \sim T^2 & \text{near AFM-QCP} \\ T\int_0^{\Lambda} \frac{q^2 dq}{r_m} = \frac{T\Lambda^3}{r_m} \sim T & \text{away from QCP .} \end{cases} \tag{4.10}$$

Due to the magneto-electric coupling in this material, the above temperature dependence of λ_m enters into the static dielectric constant and results in the following temperature dependence of inverse static dielectric constant

$$\chi_e^{-1}(0,0) \sim \begin{cases} \tilde{r_e} - w c_m^{-\frac{3}{2}} T^2 \ (\text{near AFM} - \text{QCP}) \\ \tilde{r_e} - \frac{w\Lambda^3}{r_m} T \ (\text{large } T_N). \end{cases} \tag{4.11}$$

Here $\tilde{r_e}$ is the re-normalized value of r_e which includes the temperature independent contribution from paraelectric fluctuations. Thus we see that, dielectric measurements can be considered as an indirect thermodynamic probe for magnetic systems in a magnetic quantum paraelectrics. It is to be noted that unlike the similar discussions in our second chapter, we do not equate c_m to unity here. We will see in the subsequent discussions that along with quartic couplings and magneto-electric couplings, c_m/c_e will be an important parameter to determine the dominance between the magnetic fluctuations and the paraelectric fluctuations to contribute to the static dielectric susceptibility of this material in certain appropriate circumstances. In next subsection we will consider change in the dielectric behavior in this system in case of field induced transition in the magnetic subsystem.

Non-zero magnetic field ($h \neq 0$): Non-zero h modifies anti-ferromagnetic order, develops a ferromagnetic order along the direction of the field and the resulting magnetic configuration becomes *canted*[5]. Firstly it is apparent from our starting action (eqn.(4.1)) that in case of a non zero h along the z-direction, anti-ferromagnetic gap in the transverse plane (with respect to the field) changes to,

$$r_m \sim r_m(0) - h^2. \tag{4.12}$$

Where $r_m(0) < 0$ is the value of r_m at zero magnetic field. Thus h reduces the gap in the transverse directions. In the regime $r_m < 0$ and $|r_m| > h^2$, m_0 is still

non-zero and it increases with increasing h in the following manner,

$$m_0 \sim (h^2 - r_m(0))^{\frac{1}{2}} \sim (h - h_0)^{\frac{1}{2}}, \tag{4.13}$$

where $h_0 = \sqrt{r_m(0)}$. But the ferromagnetic order along the direction along the field grows more rapidly with applied magnetic field as follows,

$$m_{fm} = -\frac{\partial F}{\partial h} \sim \frac{h(h^2 - r_m(0))}{v}. \tag{4.14}$$

A schematic phase diagram for field induced transition in the magnetic subsystem is shown in figure (4.2). In our case $r_m(0)$ is negative and we consider the external magnetic field induced modification of the anti-ferromagnetic -ground state to a canted state with partial ferromagnetic order and its effect on the static dielectric susceptibility. An experimentally observed fact is that the increase in anti-ferromagnetic component results in the suppression of dielectric constant while the roll of the uniform component is just opposite to it. If we assume both the components couple to the polarization in the same fashion, we can make an estimate of the critical value of the magnetic field (h_c) which exactly nullifies the effects of magnetic order on the static dielectric constant, in the following way. Using eqn. (4.13) and (4.14) we get,

$$\begin{aligned} (h_c^2 - r_m(0)) &= \frac{1}{c} \times h_c^2 (h_c^2 - r_m(0))^2 \\ \Rightarrow h_c &= \frac{r_m(0) \pm \sqrt{r_m^2(0) + 4c}}{2}. \end{aligned} \tag{4.15}$$

Where c is a non-universal constant and so is h_c. Thus at $r_m(0) = 0$ i. e. at anti-ferromagnetic quantum critical point, $h_c \sim \sqrt{c}$. Apart from this, external magnetic field has one more effect on quantum criticality. In case of field induced transition, the finite temperature behavior near quantum critical point will also be different. If we look back the action(4.1), we see that the magnetic field adds a new dynamic term $\sim -i\vec{h} \times \vec{m}' \cdot \partial_\tau \vec{m}'$ which is linear in ω_n. Thus the dynamic exponent $z = 2$ and the temperature dependent momentum cut-off for magnetic

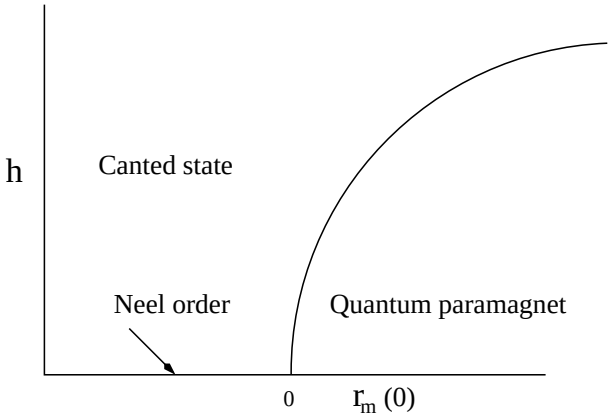

Figure 4.2: Figure shows the schematic phase diagram for a field induced transition in insulating Heisenberg anti-ferromagnet at zero temperature. At $h = 0$, $r \leq 0$ indicates a Neel order and $h \neq 0$, $r < 0$ region represents Canted state with both ferromagnetic and anti-ferromagnetic order[5].

excitations $\Lambda \sim \sqrt{\frac{T}{hc_m}}$ in this case. For small h, i. e. when $m_{fm} << m_0$,

$$\lambda_m \sim T \int_0^{\sqrt{T/hc_m}} \frac{q^2 dq}{c_m q^2} = c_m^{-\frac{3}{2}} h^{-\frac{1}{2}} T^{3/2}. \tag{4.16}$$

Thus one would expect a $T^{3/2}$ contribution, from the magnetic subsystem to the inverse static dielectric susceptibility at low temperature near anti-ferromagnetic quantum critical point and

$$\chi_e^{-1}(0,0) \sim \tilde{r_e} - w\lambda_m = \tilde{r_e} - w c_m^{-\frac{3}{2}} h^{-\frac{1}{2}} T^{3/2}. \tag{4.17}$$

Here we assume that the applied field is small enough to induce a meta-electric transition.

Near ferroelectric quantum critical point: So far, we have considered the dielectric subsystem as a spectator with a temperature independent dielectric susceptibility at low temperature. However, one can make T^* closer or smaller than T_N through doping. A generic possibility is replacing O^{16} by O^{18} in $EuTiO_3$, as is done in case of $SrTiO_3$[53]. Such a doping can create a reduced crossover temperature $\tilde{T}^*(x) \sim (1-x)^{\frac{1}{2}} T^*$, (where x is the impurity concentration) and move the system towards ferroelectric quantum critical point without affecting the mag-

netic subsystem. At finite temperature near ferroelectric quantum critical point, dipolar contribution to the inverse static dielectric constant is $\sim uc_e^{-\frac{3}{2}}T^2$ which will compete with negative contribution ($\sim -T^\nu$, $\nu = (1, 2, 3/2)$), coming from the coupling with magnetic subsystem. If we assume that these two quantum critical point do not affect each other, then considering the leading order temperature dependence to the static dielectric susceptibility, we can write,

$$\chi_e^{-1} = \alpha + \gamma_e T^2 - \gamma_\nu T^\nu. \tag{4.18}$$

Where α, γ_e, γ_ν with γ_e, $\gamma_\nu > 0$ are constant which varies from system to system and γ_e and γ_ν are proportional to $uc_e^{-\frac{3}{2}}$ and w respectively and γ_ν for different values of ν is given as follows

$$\gamma_\nu \sim \begin{cases} w\frac{\Lambda^3}{r_m} \text{ for } \nu = 1 \\ wc_m^{-\frac{3}{2}}h^{-\frac{1}{2}} \text{ for } \nu = 3/2 \\ wc_m^{-\frac{3}{2}} \text{ for } \nu = 2. \end{cases} \tag{4.19}$$

Among all these values, except $\gamma_{3/2}$ other γ_νs are fixed by system parameters and can not be controlled externally. Since $\gamma_{3/2}$ depends on the external magnetic field, the temperature scale up-to which a $T^{3/2}$ behavior of the dielectric susceptibility should be observed is also depends on it and can be tuned externally in an experimental situation. However for $\nu = 1$ or $3/2$, the temperature dependence of static dielectric susceptibility at low temperature will be dominated by anti-ferromagnetic quantum critical point with ($1/T^\nu$ increase) and there will be a maxima at a temperature $T_{max} = (\frac{2\gamma_e}{\nu\gamma_\nu})^{\frac{1}{\nu-2}}$ as shown in figure 4.3. For $\nu = 2$, there will be a competition between anti-ferromagnetic quantum critical point and ferroelectric quantum critical point and depending on the values of γ_2/γ_e an anti-ferromagnetic quantum critical point dominated behavior with $1/T^2$ increase or a ferroelectric quantum critical point dominated behavior with $1/T^2$ decrease can be found in the static dielectric constant as shown in the figure 4.4.

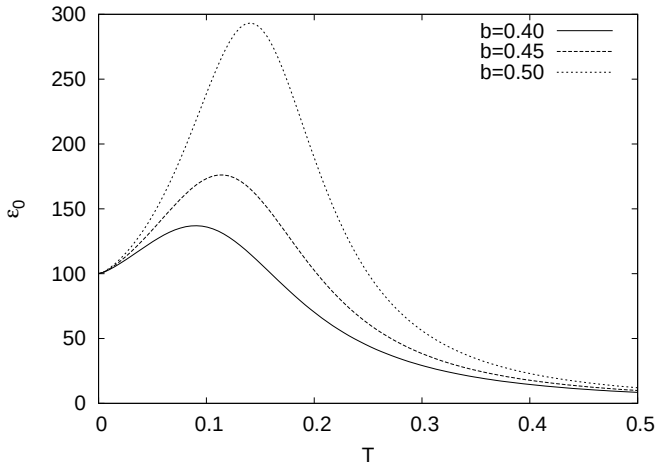

Figure 4.3: Figure shows the temperature dependence of static dielectric susceptibility near both the anti-ferromagnetic and ferroelectric quantum critical point for various values of the parameter $b = \frac{\gamma_\nu}{\beta}$. Here $\nu = 3/2$, and $\alpha = 0.01$ and ϵ_0 and T are plotted in arbitrary scale.

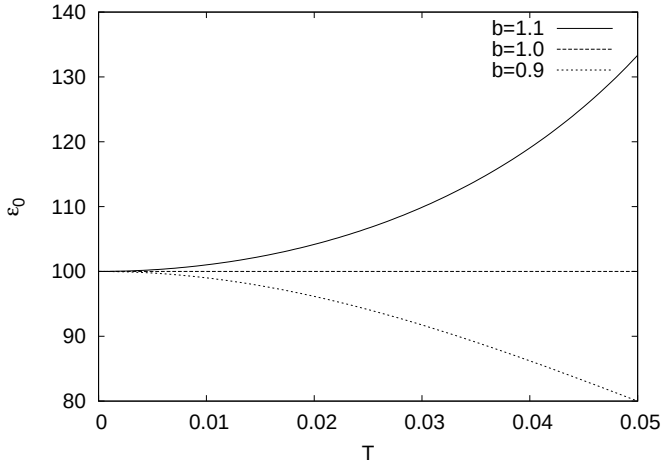

Figure 4.4: Figure shows the temperature dependence of static dielectric susceptibility near both the anti-ferromagnetic and ferroelectric quantum critical point for various values of the parameter $b = \frac{\gamma_2}{\beta}$. Here $\alpha = 0.001$ and ϵ_0 and T are plotted in arbitrary scale.

4.3 Discussion

In this work we presented a mean field theory to discuss the temperature and the magnetic field dependence of the static dielectric susceptibility of a magnetic quantum paraelectric at low temperature. In this material anti-ferromagnetic fluctuations are coupled to the polarization fluctuations and their interplay can lead to many interesting thermodynamic consequences when some non-thermal control parameters of both fluctuations are tuned to near critical values. We focus on the behavior of the system in the vicinity of two such quantum critical points both in absence and in presence of an external magnetic field. Based on scaling argument near quantum critical points, we predict that there is a possibility that the low temperature suppression of the static dielectric susceptibility due to magnetic order can be compensated by polarization fluctuations and the static dielectric susceptibility would take a $1/T^2$ form as predicted for quantum critical ferroelectrics[23]. On the other hand because of magneto-electric coupling there is a possibility of new power law behavior of the static dielectric susceptibility in presence of an external magnetic field and is predicted to be $1/T^{3/2}$ in this case. At present, up-to our knowledge, there is no report on experimental investigations on the simultaneous effects of two such quantum critical points. Hence fitting some experimental data through the numerical solutions of self-consistent equations is not tried here. Rather possible new features in this multi-ferroic material near various quantum critical points are explored. Moreover this system, in many aspects shares similarity with the systems where anti-ferromagnetic order parameter is coupled to superconductivity[54]. Thus apart from being important in their own rights, further studies in this material can be beneficial to other systems and of-course the field of quantum critical phenomena in solids in general.

5

Disorder in quantum paraelectrics

5.1 Introduction

In this chapter we focus on the effects of disorder in quantum critical paraelectrics. In previous chapters we developed a theoretical set-up for such materials which shares many similarities with systems like itinerant magnets and other strongly correlated systems near a quantum phase transition. However all those discussions were devoted to pure systems where effects due to disorder were completely neglected. It is quite justified to speculate that like other systems with which a quantum critical paraelectric shares many similarities, can show many disorder induced novel features which are beyond the realm of theory of pure systems. We follow the recent progress in the understanding of quantum phase transitions in strongly correlated systems with disorder[55, 56, 58, 47] and make an attempt to develop a theory of disordered quantum critical paraelectrics. We restricts the discussions to the case of quenched disorder of random T_c type, i.e. the disorder parameter couples to the energy density and is frozen in time. Relevancy of a particular type of disorder can be tasted using Harris criteria which was introduced in the first chapter. This criteria tells us that the kind of disorder we consider here can destroy a quantum critical point of a pure system if $\nu < \frac{2}{d+z}$. Here ν, d, and z are the correlation length exponent, dimensionality of the system and the dynamic scaling exponent respectively. In case of a paraelectric near a quantum critical point, $d = 3$ and $z = 1$ and a mean field theory predicts $\nu = 1/2$. Thus according to the Harris criteria this particular type of disorder is marginally relevant for this system and can not be neglected. This necessitates a theoretical descrip-

tion beyond a standard mean field theory. In general, a small amount of quenched impurity and associated disorder, can create locally ordered regions(droplets) even above the transition point of the corresponding pure system. Near a phase transition, large size droplets become more probable and their slow dynamics becomes an important factor to determine the nature of quantum phase transition in the disordered system. Experiments on various disordered paraelectrics also support the occurrence of locally ordered regime and glassy behavior. Most of the disordered quantum paraelectrics show *relaxor behavior* which is often described as a classical glassy behavior of a dipolar system[62, 63]. With these motivations we focus on the effects of disorder in quantum critical behavior of certain ferroelectrics in this chapter. Such issues were addressed in case of classical critical behavior earlier[64] and some attempts to make a quantum generalization of it in context of itinerant magnets have been proposed in the recent past[56, 57, 58, 47]. We use some of the earlier results and develop a new mean field description of the possible low temperature behavior of a disordered quantum critical paraelectric.

5.2 Mean field Theory

Following our earlier discussions on quantum paraelectrics without disorder, we start with a one component Landau-Ginzburg-Wilson quadratic action.

$$\mathcal{A}_{pure} = \frac{1}{2\beta}\sum_{n,\mathbf{q}}(\omega_n^2+\mathbf{q}^2+r)|\phi(\omega_n,\mathbf{q})|^2+\frac{u}{4!}\int d\mathbf{x}\int_0^\beta d\tau\phi^4(\mathbf{x},\tau). \quad (5.1)$$

The parameter r determines the gap in polarization fluctuations in absence of the interaction and $r=0$ is the mean field quantum critical point of the pure system. Disorder is introduced into the problem as a random variation of the non-thermal tuning parameter r in real space and the disorder contribution to the above action is given by,

$$\mathcal{A}_{dis} = -\frac{1}{2}\int d\mathbf{x}\int_0^\beta d\tau\delta r(\mathbf{x})\phi^2(\mathbf{x},\tau). \quad (5.2)$$

A Gaussian probability distribution of $\delta r(\mathbf{x})$ with variance g is assumed to be

$$P(\delta r(\mathbf{x}))\propto exp\{-\frac{1}{4g}\int d\mathbf{x}\delta r^2(\mathbf{x})\}, \quad (5.3)$$

so that $\overline{\delta r(\mathbf{x})} = 0$ and $\overline{\delta r(\mathbf{x})\delta r(\mathbf{y})} = g\delta^d(\mathbf{x}-\mathbf{y})$. For a single realization of disorder configuration, the partition function, hence the thermodynamic properties can be calculated using the total action $\mathcal{A}_{pure} + \mathcal{A}_{dis}$. It is also apparent that to consider the effects of disorder we need to do a proper averaging either at the level of partition function or at the level of free energy. Now the question is which one will lead to the correct physical behavior? In order to obtain the physical free energy (self averaging) we need to average the free energy or the logarithm of the partition function over all possible disorder configurations. To calculate the disorder averaged free energy, we use the identity $\log x = \lim_{n\longrightarrow 0} \frac{x^n-1}{n}$ and write the average free energy in terms of n^{th} power of the partition function as

$$\overline{F} = -\frac{1}{\beta}(\overline{Z^n} - 1)/n, \tag{5.4}$$

taking $n \longrightarrow 0$ at the end of the calculation. We change the status of the one component field ϕ to a n-component one by introducing replicas of the order parameter ϕ_a with replica index a $= 1, ..., n$. This process along with a disorder averaging help us in writing $\overline{Z^n}$ as a functional integral in terms of replica fields and the resulting action to leading order becomes,

$$\begin{aligned}\mathcal{A} &= \frac{1}{2\beta}\sum_{m,\mathbf{q},a,b}(\omega_m^2 + \mathbf{q}^2 + r)|\phi_a(\omega_m,\mathbf{q})|^2\delta_{ab} + \frac{u}{4!}\int d\mathbf{x}\int_0^\beta d\tau\phi_a^4(\mathbf{x},\tau)\delta_{ab} \\ &- \frac{g}{4}\int d\mathbf{x}\int_0^\beta d\tau\int_0^\beta d\tau^{'}\phi_a^2(\mathbf{x},\tau)\phi_b^2(\mathbf{x},\tau^{'}).\end{aligned} \tag{5.5}$$

Here a, b are the replica indices which take positive integer values up-to some integer n and the last term is a interactions between fields with different replica indices is a consequence of the disorder averaging. It is to be noted that this interaction is between the fields is non-local in time. This is due to the quenched nature of the disorder and has important consequences in case of a quantum phase transition and is absent in classical phase transition where dynamics of the system play no role. Now we use the action (5.5) to study the dielectric behavior of this disordered system. To begin with, first we consider a *replica symmetric case*. We define replica symmetric solution as replica independent field configurations, i.e.

$$\phi_a(\mathbf{x},\tau) = \phi(\mathbf{x},\tau) \text{ for all } a \tag{5.6}$$

and their replica diagonal two point correlation functions, i.e.

$$\chi_{ab} = \chi_{aa}\delta_{ab} = \chi_0\delta_{ab} \text{ for all } a,\ b. \tag{5.7}$$

We consider a paraelectric phase i.e. the thermodynamic average of the dipolar field $< \phi >= 0$ and make a self-consistent quasi-harmonic approximation to decouple the quartic term as done in the chapter 2. In this scheme a quartic term such as $\int dxd\tau\phi^4(x,\tau)$ is decomposed as $\lambda_0 \int dxd\tau\phi^2(x,\tau)$. Where $\lambda_0 = \int dx^{'}d\tau' < \phi^2(x^{'},\tau') >$. Thus susceptibility of the disordered paraelectric can be written as,

$$\chi_0(\omega_m, \mathbf{q}) = \frac{1}{(\omega_m^2 + q^2 + r + \lambda_0)}. \tag{5.8}$$

In the above equation λ_0 describes the fluctuation corrections to ferroelectric gap and is defined by the following self-consistent equation

$$\begin{aligned} \lambda_0 &= \sum_{m,\mathbf{q}} (u\chi_0(\omega_m, q) - g\chi_0(0, q)) \\ &= u\int d^3q \frac{1}{\Omega_q} \coth \beta\Omega_q - g\int d^3q \frac{1}{\Omega_q^2}. \end{aligned} \tag{5.9}$$

Here the fluctuation renormalized natural frequency Ω_q is dependent on λ_0 and is defined as,

$$\Omega_q^2 = q^2 + r + \lambda_0. \tag{5.10}$$

It is to be noted that the second term in the equation(5.9) is a zero frequency contribution. The reason is that we consider quenched disorder which has no dynamics and thus strongly correlated in time. However above two equations can be obtained by integrating $\delta r(x)$ without introducing replica trick and need to be solved self-consistently. It is clear form the expression for λ_0 that the second integral in the equation (5.9) gives a shift in gap and depending on its strength controls quantum fluctuations. In this scheme the solution of the equation $r - g\int d^3q\Omega_q^{-2} = 0$ for r gives the quantum critical point. Experimentally one is interested in low but finite temperature behavior of a system at that point and expects power law dependencies in temperature for various physical quantities. We have seen in our previous discussions in chapter 2 that the static dielectric susceptibility $\sim T^{-2}$ at a ferrolelectric quantum critical point.

An analysis with a replica symmetric ansatz is not capable of including spatial inhomogeneity created by disorder and thus the effects of disorder considered here are identical to the effects of hydrostatic pressure. However in a doped quantum paraelectric we find the so called relaxor behavior which can not be captured in a theory without considering the existence of macroscopic number of local minima configurations. Next question is why such local configurations occur and how to include them in a consistent theory. In a simple minded picture one can argue the existence of such local configurations as follows. Since $\delta r(x)$ is a random variable, there are certain regions in the sample where $r - \delta r(x) < 0$. Those regions become ferroelectric even above $r = 0$, the mean field quantum critical point of the pure system. Those ferroelectric islands have non-zero polarization $\sim \sqrt{|\delta r(x) - r|}$ and are often dubbed as *droplet*. Since such solutions have finite spatial extension, one must consider a large number (exponential in its volume) such solutions to get appreciable effects in the thermodynamic limit. Ideally finding such a huge number of local solutions, doing a sum over their contributions to the free energy followed by a disorder averaging will complete the task. Clearly it is an impossible. An alternate way to include the effects of spatial inhomogeneity created by disorder is to use replica trick with replica symmetry broken at the vector level[64]. By the word 'vector' we mean that ϕ_a is the ath component of a n-component field in the 'replica space' and its symmetry breaking means different components assume different values. In this scheme the field configurations are assumed as

$$\begin{aligned} \phi_a(\mathbf{x},\tau) &= \phi_k(\mathbf{x},\tau) + \psi(\mathbf{x},\tau) \,\text{for}\, a = 1,..,k \\ \phi_a(\mathbf{x},\tau) &= \psi(\mathbf{x},\tau) \,\text{for}\, a = k+1,.....,n. \end{aligned} \tag{5.11}$$

Without loss of generality, we assume the correlation functions to be block-diagonal, i.e.,

$$\begin{aligned} \chi_{ab}(\mathbf{x},\tau) &= \chi_1(\mathbf{x},\tau) + \chi_2(\mathbf{x},\tau)\delta_{ab} \,\text{for}\, a,b = 1,..,k \\ &= \chi_2(\mathbf{x},\tau)\delta_{ab} \,\text{for}\, a = k+1,.....,n. \end{aligned} \tag{5.12}$$

Here $k \geq 1$ is an integer that determines the degree of the symmetry breaking process. In principle one should break replica symmetry and hence should chose k in all possible ways. If we use the ansatz (5.11) and write an equation of motion

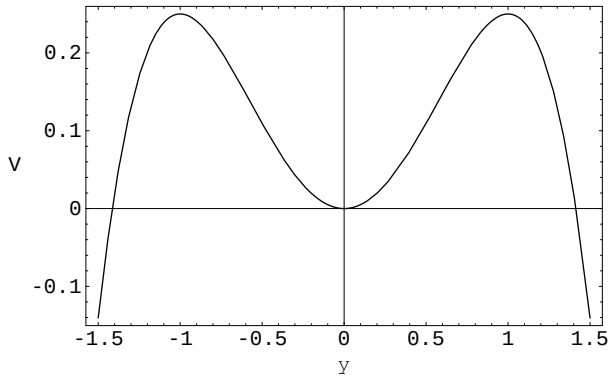

Figure 5.1: A sketch of an inverted double well potential.

corresponding to the action (5.5). It lead to a non-linear Schrödinger equation for a particle in an *inverted double well* potential as follows

$$-\nabla^2\phi_k(\mathbf{x}) + r\phi_k(\mathbf{x}) - (gk - u)\phi_k^3(\mathbf{x}) = 0. \tag{5.13}$$

A sketch of an inverted double well is given in the figure 5.1. We should keep in mind that this replicated action is not the actual free energy which can be obtained only after taking the limit $n \to 0$. In replica formalism the solution of the equation (5.13) corresponding to the maxima are the physical minima. Hence a solution corresponding to the minima of the inverted double-well potential is opted. It is to be noted that the existence of a local solution depends on the disorder strength g and our choice of k. One has to consider all possible choice of k among n replicas and has to make sure that a summation over contributions from all possible choices of k survives at the limit of $n \to 0$. A replica symmetric ansatz corresponds to $k = n$ which can be chosen in a single way and have a vanishing contribution in the limit $n \to 0$. All those details are not relevant for further discussions. For more detailed discussions we refer to [64, 65]. However in a classical treatment for $gk > u$, ϕ_k corresponds to localized solution which can be written in a scaled form for isotropic case as,

$$\phi(\mathbf{x}) = \sqrt{\frac{r}{(gk - u)}}\psi(\sqrt{r}x). \tag{5.14}$$

So that $\psi(z)$ obeys a scale independent equation

$$-\nabla_z^2\psi(z) + \psi(z) - \psi^3(z) = 0. \tag{5.15}$$

The proper boundary conditions are $\psi(0) =$ constant and $\psi(\pm\infty) = 0$. Equation (5.15) has exponential decaying solutions for $x \gg \sqrt{r}$ and is smooth for $x < \sqrt{r}$. The size of the droplet R is determined by the dipolar correlation length and $R \sim \frac{1}{\sqrt{r}}$. At very low temperature the dynamics of the droplets become important. In a simplest approximation spatial and the time dependent parts of the polarization field can be decoupled completely.

$$\phi(\mathbf{x}, \tau) = \phi_k(\mathbf{x})\mathcal{T}(\tau). \tag{5.16}$$

This choice assumes that the droplet tunnels as a whole. Substitution of equation (5.16) in the replicated action (5.5) followed by a summation of all possible choices of k among n replicas (which can be done in C_k^n ways) in the limit $n \to 0$ leads to

$$Z[\mathcal{T}] \approx \int D\mathcal{T}(\tau)\ e^{-r^{2-d/2}E_2\mathcal{F}[\mathcal{T}]/u}. \tag{5.17}$$

Where the dynamical part of the action is given as[66],

$$\mathcal{F}[\mathcal{T}] \;=\; \frac{1}{\beta}\int_0^\beta d\tau \left\{ \frac{M}{2}\left(\frac{d\mathcal{T}}{d\tau}\right)^2 + \frac{\mathcal{T}^2(\tau)}{2} - \frac{1}{4\beta}\mathcal{T}^2(\tau)\int_0^\beta d\tau'\mathcal{T}^2(\tau') \right\}. \tag{5.18}$$

This is clearly the dynamical part of the action for undamped Bosonic system. The coefficient M in equation [5.18] is given as,

$$M \;=\; \frac{E_1}{E_2}\, r^{-1}. \tag{5.19}$$

The parameter M can be associated with the *mass* of a *quantum particle* in a double well potential. It is an undamped tunneling problem where the tunneling splitting between two configurations $X = \pm 1$ is given by,

$$r_L \approx 2e^{-r_0/r} \tag{5.20}$$

with $r_0 \sim E_1/E_2$ a constant, where $E_N = \int dz\phi(z)^{2N}$. If we consider Gaussian fluctuations around the droplet solutions, an effective action for those fluctuations $\psi(x,\tau)$ can be written as,

$$\mathcal{S}[\psi] = \frac{1}{2\beta}\sum_{m,\mathbf{q},a,b}((\omega_m^2+\mathbf{q}^2)\delta_{ab}+\mathcal{M}_{ab})\psi_a\psi_b. \tag{5.21}$$

The presence of droplets and its tunneling introduces a "gap-matrix" $\{\mathcal{M}_{ab}\}$ which contains $k\times k$ block with elements,

$$\mathcal{M}_{ab} = r(1-\frac{gk-3u}{gk-u}\lambda_L)\delta_{ab} - \frac{2gkr}{gk-u}\lambda_L \tag{5.22}$$

and diagonal elements for the remaining $n-k$ replicas

$$\mathcal{M}_{ab} = r(1-\frac{gk}{gk-u}\lambda_L)\delta_{ab}. \tag{5.23}$$

Here λ_L encodes the contributions from the localized solutions along with their dynamics and is given as

$$\begin{aligned}\lambda_L &= \sum_\omega \int dz < \psi(z)\mathcal{T}(\omega)\psi(z)\mathcal{T}(\omega) > \\ &= \int dz\psi^2(z)\sum_\omega < \mathcal{T}(\omega)\mathcal{T}(\omega) > \\ &\sim \frac{1}{\omega_-} \text{ at } T=0\end{aligned} \tag{5.24}$$

Here $\omega_\pm = 2\pm r_L$. It is to be noted that the vector breaking of replica symmetry not only introduces inhomogeneous solutions but also glassy effects through off-diagonal elements in the gap-matrix. Putting $\lambda_L = 0$ identically, we get back the behavior of a pure system. However in this scheme, inverse replica correlators for disordered paraelectric is given by

$$\chi_{ab}^{-1}(\omega_m,\mathbf{q}) = ((\omega_m^2+\mathbf{q}^2)\delta_{ab}+\mathcal{M}_{ab}). \tag{5.25}$$

We look for replica diagonal correlations in equations (5.22) and (5.23). Diagonal-

ization of the gap-matrix are given as,

$$\hat{\mathcal{M}}_{aa} = \begin{cases} r(1 - \frac{gk-3u}{gk-u}\lambda_L),\ a = 1, ..k-1, \\ r(1 - \frac{3gk-3u}{gk-u}\lambda_L),\ a = k, \\ r(1 - \frac{gk}{gk-u}\lambda_L), a = k+1, .., n \end{cases} \tag{5.26}$$

Using equation(5.26) and (5.24) we find the values of r at which zero temperature diagonal susceptibility ($\sim \frac{1}{\hat{\mathcal{M}}_{aa}}$) diverges. The instability points depends on the disorder strength and the value of k and is given as,

$$r_c = \begin{cases} -r_0/\log(1 - A\frac{(gk-3u)}{(gk-u)}),\ a = 1, .., k \\ -r_0/\log(1 - A\frac{(3gk-3u)}{(gk-u)}),\ a = k \\ -r_0/\log(1 - A\frac{gk}{(gk-u)}),\ a = k+1, .., n \end{cases} . \tag{5.27}$$

and is k dependent. Here A is a system dependent parameter. For a simple minded

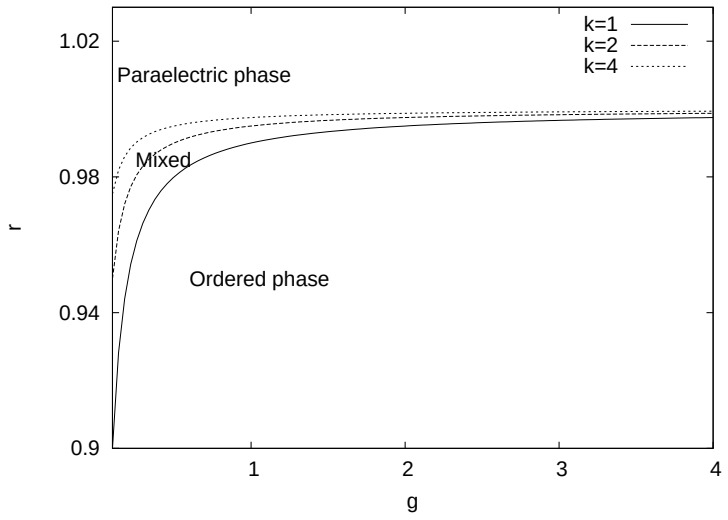

Figure 5.2: Phase diagram at zero temperature in $g - r$ plane for two particular values of k with u taken as unity. It is clear that the phase diagram acquires a region of mixed phase for any non-zero disorder strength.

analysis, let us consider the $a = k+1, .., n$ elements only. For $|\frac{Agk}{(gk-u)}| << 1$, r_c s

can be written in the following form

$$\begin{aligned} r_c(k) &= r_0^{'}\frac{(gk-u)}{gk} \\ &= B - \frac{C}{k}\ (B = r_0^{'},\ C = \frac{r_0^{'}u}{g}). \end{aligned} \tag{5.28}$$

Apart from the physical parameters like disorder strength, anharmonic strength etc., r_c depends of the choice of k. For each choice of k, we get a curve in the $g - r$ plane which separates an ordered phase from a paraelectric phase. When such curves are plotted for more than one value of k, the region surrounded by the upper most and the lower most curves represents a region of mixed phase as shown in figure 5.2. In this regime a disordered system is characterized by a set of clusters of ordered phase, off-critical para phase and critical-para phase. Since the choice of k is random, depending on its distribution at the limit $n \to 0$, we can estimate a distribution, hence width of r_c. In a replicated action with n replicas, k can be chosen in C_k^n ways. Thus we can define a normalized distribution of $\mathcal{P}(k)$ as follows

$$\mathcal{P}(k) = \frac{C_k^n}{\sum_{k=1}^{n} C_k^n} = \frac{1}{2^n - 1}\frac{\Gamma(n)}{\Gamma(k)\Gamma(n-k)}. \tag{5.29}$$

Since the gamma function with negative argument is infinity, the limit of k can be extended to infinity. In the limit $n \to 0$, using the asymptotic form of gamma functions, $\mathcal{P}(k)$ can be approximated as[64]

$$\mathcal{P}(k) \approx \frac{1}{\log 2}\frac{(-1)^{k-1}}{k} \approx \frac{1}{\log 2}\frac{\cos\pi\cos\pi k}{k}. \tag{5.30}$$

Negative values of $\mathcal{P}(k)$ for some values of k may turn out to be counter intuitive to the usual notion of a distribution function. But such distributions are allowed in replica scheme. There are several possible broken replica symmetric cases, each characterized by the number k which follows a distribution $\mathcal{P}(k)$. For a fixed disorder strength δ, each k results a different instability point r_c. In stead of k, if we characterize various possible broken replica symmetric cases by r_c, a distribution of r_c can be estimated as

$$\mathcal{P}(r_c) = \mathcal{P}(k)|\frac{\Delta k}{\Delta r_c}| \sim \frac{1}{B - r_c} \times \cos(\pi k). \tag{5.31}$$

This is a broad power-law distribution of r_c around a system dependent parameter B with a cosine factor. The probability distribution can be assumed to be smooth around $k =$ any positive integer, excluding zero. The expansion around $k = 0$ is excluded as it corresponds to small u/g limit where the action (5.5) becomes unstable even in a replica symmetric ansatz. In that limit the system will undergo a first order transition in a replica symmetric analysis, the stability of the system needs a ϕ^6 term in the action (5.5) which will lead to more complicated localized solutions in a broken replica symmetry picture. However we focus on those u/g values where the above possibilities are not present and the distribution function is smooth. It is to be noted that the power law nature of $\mathcal{P}(r_c)$ arises because of the dynamics of the locally ordered regimes and also depends on the distribution of k used. Neglecting cosine factor within some range of r_c say $(B+R, B-R)$, average susceptibility of the disordered quantum paraelectric can be estimated as,

$$\begin{aligned} \overline{\chi(r,T)} &\sim \int_{B-R}^{B+R} dr_c \frac{1}{B-r_c} \times \frac{1}{r-r_c+T^2} \\ &= \frac{1}{r-B+T^2} \log \frac{r-B-R+T^2}{r-B+R+T^2}. \end{aligned} \tag{5.32}$$

It is evident that inclusion of fluctuations due to locally ordered regime introduce a parameter $R \sim \mathcal{O}(u/g)$ and changes the usual quantum critical behavior of a paraelectric. In the limit $r \to B$ the temperature dependence of the static dielectric constant of a disordered quantum paraelectric can be predicted as,

$$\overline{\chi(r,T)} \sim \begin{cases} \text{constant}, \; T << R \\ 1/T^4, \; T >> R. \end{cases} \tag{5.33}$$

This is a deviation from the standard quantum critical behavior which predicts $\chi(T) \sim T^{-2}$ in a mean field theory. In an infinite disorder limit, i.e. for $u/g \to 0$, $R \to 0$ and $\chi(T)$ shows a power law behavior ($\sim 1/T^4$) with non-universal exponent. Such situation is often dubbed as quantum Griffiths phenomena in the quantum phase transition literature.

5.3 Discussions

In this chapter, the low temperature dielectric behavior of a quantum paraelectric in presence of quenched disorder is addressed. A suitable action for these materials, with random T_c type disorder have been studied using a replica trick. The effects of disorder induced locally ordered regimes and their tunneling in the low temperature are captured in this formalism. We derive an expression for the distribution of instability points for a fixed value of disorder strength and demonstrate the possibility of a mixed phase at non-zero disorder strength. This analysis predicts a broad power law distribution around a system dependent parameter with a cosine correction for the instability points. Using the distribution it is possible to show analytically how the temperature dependence of static dielectric susceptibility of a disordered quantum critical paraelectric deviates from its pure counterpart. Our analysis is a completely new attempt in context of the effects of disorder in ferroelectrics near a quantum critical point. In a qualitative manner it predicts certain new features such as occurrence of a phase with mixture of critical and non-critical regimes with a distributions of transition points which are missing in earlier works in similar issues in context of itinerant magnets. Moreover the whole analysis is interesting in context of the use of replica trick to incorporate disorder induced inhomogeneities or locally ordered regime in the studies of quantum phase transition and may turn out to be useful in explaining certain experimental results on disordered ferroelectrics near a quantum critical point.

6

Summary

This thesis contains some theoretical studies on the various fluctuation effects on the low temperature dielectric properties of certain incipient ferroelectrics in the vicinity a of a quantum phase transition. Studies are base on some experimental findings on the low temperature dielectric behavior of some incipient ferroelectrics such as $SrTiO_3$, $KTaO_3$, $EuTiO_3$ etc. under various external perturbations. We have studied a minimal model in each case of pure quantum paraelectrics, its coupling with anti-ferromagnetic fluctuations, strain and disorder. Studies based on self-consistent mean-field approximations and scaling arguments, are capable of explaining many experimental findings and making various interesting predictions about the dielectric behavior of these materials. These systems are of displacive type i.e. phase transition in these systems is associated with a softening of a transverse optic mode. More microscopic scenario could be, a set of dipoles sitting at the center of each unit cells of these perovskite materials are interacting via long range dipolar interactions. As a result of the long range nature of the dipolar interaction the fluctuations along the longitudinal direction with respect to the wave vector are gaped out and the transverse mode fluctuations become the most relevant to describe the low temperature dielectric properties of these materials.

To explore the possible consequences of a quantum fluctuations in the low temperature dielectric behavior of these materials a semi-phenomenological Landau-Ginzburg theory is used. We restrict ourselves to an one component model to make our analysis simpler. Moreover anisotropy induced by the dipolar interaction in the transverse optic modes is neglected and a justification for the same is given in chapter 2. With this minimal model we are able to describe the effects of quantum

fluctuations in case of pure $SrTiO_3$, a prototype quantum paraelectric material. The same analysis is extended to predict its dielectric behavior when it is tuned to a quantum critical point. A prediction about the $1/T^2$ behavior in contrast to the usual Curie-Weiss behavior is made and verified by a recent experiment[25]. A schematic phase diagram is proposed to to classify various dielectric materials in a quantum phase transition point of view.

Our first chapter was devoted to set up a basic theoretical ground for discussing low temperature properties of quantum paraelectrics along with some predictions about the quantum critical behavior of them. With this background we focus on understanding more detail experimental observations on various quantum paraelectrics. In this context some interesting behavior of quantum critical $SrTiO_3$ is revealed in a recent spectroscopic experiment which signals a weak first order nature of the quantum phase transition in $SrTiO_3$. We assume such a behavior is a result of the coupling of the paraelectric fluctuations to strain fluctuations. Strain fluctuations are integrated out and it results a long range interaction among paraelectric fluctuations. In a pure mean field scenario, a weak first order transition occurs when the effective quartic coupling of the paraelectric action is negative and close to zero. In this case one can add a higher order term with positive coefficient in the paraelectric action and make some mean field prediction about the transition. We emphasis that in such a case one should consider fluctuation effects in the quartic coupling, namely four point vertices and show that fluctuation effects can stabilize the system without invoking higher order terms. The crucial role played by long range interaction mediated by the strain fluctuations in this process is also explained. A self consistent parquet approximation is used to take care of leading order fluctuation effects. The fact that the presence of the finite temperature restores the second-order nature of the transition near a quantum phase transition is also captured in this theory.

Next, we extend our theory to an incipient ferroelectric $EuTiO_3$ where ferroelectric fluctuations are coupled to anti-ferromagnetic fluctuations. We write an action where paraelectric fluctuations are coupled to anti-ferromagnetic fluctuations in a bipartite lattice and and in presence of non-zero magnetic field. We consider the case of coupled quantum criticality and its effect on the dielectric behavior of this system. A new power law behavior of the static dielectric constant, namely $T^{-\frac{3}{2}}$ variation, in presence of small non-zero magnetic field is predicted. It

is in contrast of the $1/T^2$ behavior of the quantum critical paraelectric and already got attentions of the experimental community[67].

Next section is an account of the effects of quenched disorder in quantum critical paraelectrics using a replica formalism. In this case the coupling between random T_c type disorder with energy density is considered. Near quantum criticality in these systems, a bare power counting scheme predicts such disorder effects to be marginally relevant. However a classical replica formalism with broken replica symmetry at the vector level predicts inhomogeneous solutions in these system. Gaussian fluctuations around such solutions in case of classical phase transitions were studied earlier. In their static limit the renormalization of the coefficient of the Gaussian fluctuations due to such inhomogeneous solutions are found to independent of their sizes and a single instability was predicted. We consider the tunneling of such solutions in the quantum limit and consider a quantum phase transition in terms of the instability of Gaussian fluctuations around them. A broad power law distribution of the quantum critical points is predicted. Its consequences of the static dielectric behavior at finite temperature is also emphasized.

In conclusion, in this work the physics of ferroelectrics is put in a broad perspective. The effects of quantum critical points on finite temperature properties of certain dielectric systems are studied. Possible exponents of the power law behavior of static dielectric constant at finite temperature are predicted. The effects of disorder induced inhomogeneity and their dynamics at low temperature is described in a replica formalism. Some results are in accord with experiments. Many aspects of these works are quite general in context of quantum phase transitions and deserve further experimental and theoretical studies.

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