Quantum Criticality and Unconventional Order in Magnetic and Dielectric Material

S E Rowley¹, R Smith¹, M L Sutherland¹, P Alireza¹, S S Saxena¹² and G G Lonzarich¹

¹Physics Department, University of Cambridge, Cavendish Laboratory, J J Thomson Avenue, Cambridge CB3 0HE, United Kingdom
²Centre for Materials and Microsystems, Fondazione Bruno Kessler, via Aommarive 18, 38123 Trento, Italy

Email: GGL1@phy.cam.ac.uk

Abstract. We present an overview of unconventional phenomena arising close to ferromagnetic and ferroelectric quantum phase transitions. The applicability and potential breakdown of traditional field theories of quantum criticality and the emergence of a multiplicity of critical fields in particular will be discussed.

1. Introduction

Critical phenomena have traditionally been described in terms of Ginzburg-Landau-Wilson field models in which spontaneous fluctuations of the order parameter fields are treated in the classical approximation. Quantum extensions of this description have been developed in more recent times in attempts to predict the behaviour of materials on the border of phase transitions that occur in the limit of absolute zero as a function of control parameters such as chemical doping, hydrostatic pressure, uniaxial stress and external applied magnetic or electric fields.[1] Here we consider briefly some of the successes and limitations of the quantum Ginzburg-Landau-Wilson model in describing in particular ferromagnetic metals and ferroelectric materials with Curie temperatures near to absolute zero.

Our discussion will be based on the phase diagram illustrated schematically in figure 1, which has provided a helpful guide to thinking about a number of nearly ferromagnetic and ferroelectric materials. The phase diagram is consistent with a quantum Ginzburg-Landau-Wilson model in which the mode-mode or two-mode coupling of the order parameter field \( \phi \) is attractive at absolute zero, i.e., a model in which the \( T = 0 \) K coefficients of the \( \phi' \) and \( \phi'' \) terms are negative and positive, respectively. When account is taken of the effect of spontaneous order parameter fluctuations one finds that a second order transition line bifurcates into two symmetry related first order sheets near to the quantum phase transition on the border of ferromagnetism or ferroelectricity.

This model with an attractive two-mode coupling provides only a partial view of the phase diagrams actually observed. In particular, the critical region depicted in figure 1 is expected to be generally unstable to the formation of other phases. Examples of unusual forms of order observed near to the critical regime have been noted in figure 1 and will be defined briefly below.
2. Quantum critical paraelectrics

We begin by considering the nearly ferroelectric compound SrTiO$_3$ and its weakly ferroelectric relative obtained by replacing $^{16}\text{O}$ by $^{18}\text{O}$. These systems are believed to lie very close to the critical regime in figure 1 in which, however, the tricritical point is at very low temperatures. This is expected to lead to a temperature dependence of the dielectric function of the form $\varepsilon \sim 1/T^2$, a prediction that depends only on the dimensionality of space, $d = 3$, and on the dynamical exponent, $z = 1$ (defined via the frequency-wavevector dispersion relation, $\omega \sim q^z$, of quantum critical fluctuations of the order parameter field). Interestingly, in this case the relevant quantum field theory is effectively in three plus one dimension, as is the case of more general quantum field theories of the physical vacuum.

The predicted phase diagram in the quantum critical regime for SrTiO$_3$ is shown in figure 2. The control parameter in this case is the square of the relevant correlation wavevector, which vanishes at the quantum critical point where the Curie temperature tends to absolute zero. As shown in figure 3 the temperature dependence of the dielectric function is well described by the prediction for a quantum critical paraelectric, apart from the weak anomalous upturn at very low temperatures that can be understood quantitatively in terms of the coupling of the order parameter field with acoustic phonons.

3. Non-local marginal Fermi liquid

In a ferromagnetic metal dissipative critical spin fluctuations with $z = 3$ replace the critical transverse optical phonons that characterize the displacive ferroelectric discussed in the last section. This leads to the temperature-pressure phase diagram shown in figure 4 for the case of the nearly critical metallic ferromagnet ZrZn$_2$ for which $d$ and $z$ are both taken to be equal to three. In the critical regime the magnetic susceptibility is in this case predicted to diverge as $\chi \sim 1/T^{4/3}$, a behaviour that has not been unambiguously observed.

Interestingly, the scattering of charge carriers from nearly critical spin fluctuations is expected to give rise to a $T^{5/3}$ electrical resistivity along with a $T$-linear thermal resistivity. As shown in figure 5, this behaviour has indeed been seen in ZrZn$_2$ in the weakly ferromagnetic state well below the Curie temperature of 28 K and above a Fermi liquid crossover temperature of the order of 1 K. This is the temperature dependence expected for a non-local marginal Fermi liquid state, which may be a precursor to unexplained non-Fermi liquid states observed near to ferromagnetic, antiferromagnetic and other quantum phase transitions in metals.

4. Breakdown of non-local marginal Fermi liquid behaviour in ferromagnetic metals

An example of an unexplained non-Fermi liquid property is the $T^{3/2}$ resistivity observed over wide ranges in pressure in ZrZn$_2$ and MnSi above the critical pressure where the Curie temperature vanishes (figures 6 and 7). A number of models have been proposed that naturally lead to an attractive two-mode coupling and to the phase diagram in figure 1 in which a first order transition is accompanied by a textured magnetic state (figure 8). The precise nature of the magnetic texture and how it leads to anomalous transport properties remains, however, incompletely understood (figure 9).

5. Superconductivity on the border of ferromagnetism

Virtual spin fluctuations can in principle lead to electron pairing instabilities on the border of magnetic order at low temperatures, and to forms of superconductivity that can be very sensitive to details of the underlying lattice and electronic structure. In the model calculations presented in figure 10, for example, the superconducting transition temperature can be much higher in a nearly two-dimensional structure compared to a three-dimensional structure, other factors being equal. This prediction is borne out in particular in studies of some closely related cubic and tetragonal compounds.

The same theoretical model also predicts that superconductivity on the border of ferromagnetism is generally not favoured except in reduced dimensionality and in particular in the presence of strong magnetic anisotropy. The physical ideas behind these predictions have stimulated the search for new superconductors and have led in particular to the discovery of superconductivity on the border of
ferromagnetism in uniaxial ferromagnets (see, e.g., figures 11 and 12) and in a quasi-two-dimensional ferromagnet (figure 13).

Interestingly, the combination of uniaxial magnetic anisotropy and two-dimensionality is predicted to produce a maximum transition temperature comparable to that of a nearly antiferromagnetic metal in quasi-two dimensions, other factors again being equal. The latter requirement, however, is in practice difficult to achieve and experience suggests that effects not included, such as orbital degeneracies and more, could in practice lead to predictions that are very different from those in figure 10.

6. Concluding comments

The border of ferroelectric quantum phase transitions in displacive ferroelectrics such as SrTiO$_3$ can be understood in terms of a quantum generalization of the classical Ginzburg-Landau-Wilson model of critical phenomena. Properties on the ferromagnetic side of the phase diagram in figure 1 in metals such as ZrZn$_2$ can also be understood in an analogous and appropriately extended framework. However, the non-ferromagnetic side of the phase diagram remains largely unexplained and characterized by magnetic texture not fully anticipated by the quantum Ginzburg-Landau-Wilson model. Moreover, the critical region of the phase diagram is unstable to the formation of potentially novel phases and thus to the emergence of a multiplicity of coupled order parameter fields, for which the electron-pair field is only one example.

Figure 1. Phase diagram on the border of metallic ferromagneticm.[2,3] Qualitative form of the phase diagram predicted in a quantum Ginzburg-Landau-Wilson model with an attractive mode-mode coupling term (attractive $\phi^4$ term) in the low temperature limit. With increasing density, or applied pressure, a second order ferromagnetic transition line bifurcates at a tricritical point into two sheets of first order metamagnetic transitions that depend on the applied magnetic field. Selected examples of phenomena observed on the border of metallic ferromagnetism (FM) and metamagnetism (MM) are indicated: NL-MFL – non-local marginal Fermi liquid in ZrZn$_2$; FM-SC = ferromagnetism and superconductivity in UGe$_2$; QTC = quantum tri-criticality in Ni$_3$Ga; ST = spin texture (skyrmions) in MnSi; ST-SC = spin-triplet superconductivity on the border of ferromagnetism in M$_2$SrO$_4$ (M stands for Sr or Ca) and NP = nematic phase in Sr$_3$Ru$_2$O$_7$. 
Figure 2. Temperature-pressure phase diagram predicted by the quantum Ginzburg-Landau-Wilson model for a displacive ferroelectric, SrTiO₃, for which \(d = 3\) and \(z = 1\).[4] FE stands for ferroelectric, QPE for quantum paraelectric and QCPE for quantum critical paraelectric. The tuning parameter is measured in terms of the relevant correlation wavevector. The chemical formulae for three displacive ferroelectrics or paraelectrics are located approximately above the corresponding ambient pressure values of the tuning parameters.

Figure 3. Temperature dependence of the inverse dielectric function \(\varepsilon\) in SrTiO₃.[4] Measured inverse dielectric function vs the square of the temperature up to 50 K. The lower inset is an expanded view at low temperature, which exhibits an upturn below 4 K. The upper inset illustrates the unit cell of SrTiO₃ in its cubic perovskite structure. A quantitative description of the \(T^2\) temperature dependence and of the upturn of \(1/\varepsilon\) at low temperatures is presented in reference [5].
Figure 4. Temperature-pressure phase diagram predicted by the quantum Ginzburg-Landau-Wilson model or self-consistent renormalization model [6] for a weakly ferromagnetic metal, ZrZn$_2$, for which $d = z = 3$. [7] FL stands for Fermi liquid and MFL stands for non-local marginal Fermi liquid. In the latter regime (excluding a narrow region near the Curie temperature), the model predicts a $T^{5/3}$ electrical resistivity and $T$-linear thermal resistivity.

Figure 5. Temperature dependence of transport properties in a weakly ferromagnetic metal, ZrZn$_2$. [7] The temperature dependence of the electrical resistivity $\rho$ and (in the lower inset) the temperature dependence of the difference $\delta$ between the thermal resistivity $\omega$ and $\rho$, where $\omega = L_0T/\kappa$, $\kappa$ is the thermal conductivity and $L_0$ is the Lorentz number. The $T^{5/3}$ electrical resistivity and $T$-linear thermal resistivity are consistent with the predictions of the self-consistent renormalization model for $d = z = 3$. In the pure samples used here with residual resistivities of 0.2 $\mu\Omega$ cm the effect of phonons on $\omega$ is found to be small and ignorable below approximately 15 K.
Figure 6. Temperature-pressure phase diagram of ZrZn$_2$.[7] The magnetic transition becomes weakly first order near the critical pressure $p_c$ of the order of 20 kbar. The resistivity varies as $T^{5/3}$ below $p_c$ and as $T^{3/2}$ above $p_c$ (upper inset). The $T^{3/2}$ resistivity, found to extend from $p_c$ to at least twice $p_c$ at low $T$, is inconsistent with the predictions of the self-consistent renormalization model in its conventional form.

Figure 7. Temperature-pressure phase diagram of MnSi.[2,8] The magnetic transition becomes weakly first order near the critical pressure $p_c$ of below 15 kbar. The resistivity varies as $T^2$ below $p_c$ and as $T^{3/2}$ above $p_c$ (upper inset). The $T^{3/2}$ resistivity, found to extend from $p_c$ to at least twice $p_c$ at low $T$, is again inconsistent with the predictions of the self-consistent renormalization model in its conventional form.
Figure 8. First order transition and texture on the border of metallic ferromagnetism. Effective electron-electron interactions beyond that included in earlier work give rise to attractive two-mode coupling in the quantum Ginzburg-Landau model. This leads to a first order quantum phase transition and magnetic texture. The tuning parameter is the inverse Fermi wavevector (times the lattice constant $a$) in a single electronic band model with a short-range starting interaction.

Figure 9. Spin vortex state on the border of ferromagnetism. Under certain conditions the magnetic textured state on the border of a ferromagnetic quantum phase transition could correspond to a spin vortex or skyrmion liquid or solid. Evidence for the existence of skyrmions has been collected in particular in MnSi.
Figure 10. Superconductivity on the border of long-range magnetic order.[11] Electron pairing via virtual spin fluctuations is highly sensitive to details of the lattice and electronic structure and nature of the magnetic fluctuations. The results shown are based on a system of electrons in a single electron band interacting via the exchange of magnetic fluctuations. The model compares the superconducting transition temperatures calculated for nearly ferromagnetic metals (nFM) and nearly antiferromagnetic metals (nAF) with cubic (3D) or tetragonal (2D) lattices as a function of the square of the magnetic correlation wavevector, which vanishes on the border of long-range magnetic order.

Figure 11. Temperature-pressure phase diagram of the ferromagnetic superconductor UGe$_2$.[12] The upper line corresponds to a ferromagnetic transition and the lower line to a metamagnetic instability with low critical end point. A superconducting dome appears on the border of metamagnetism within the ferromagnetic regime. The ferromagnetic transition becomes strongly first order close to and above about 15 kbar.
Figure 12. Temperature-pressure phase diagram of the ferromagnetic superconductor UCoGe.[13] In contrast to UGe$_2$, the superconducting dome appears to straddle the ferromagnetic quantum phase transition region and thus superconductivity arises both in the ferromagnetic and paramagnetic states.

Figure 13. Temperature-pressure phase diagram of Ca$_2$RuO$_4$.[14] The lattice structure of Ca$_2$RuO$_4$ is based on the layered perovskite structure shown in the inset, but with distorted oxygen octahedral. At ambient pressure the octahedra are compressed, tilted and rotated (space group $S$-$Pbca$) and Ca$_2$RuO$_4$ is described as a spin-one antiferromagnetic insulator. Above ~5 kbar the compression is suppressed (space group $L$-$Pbca$) and Ca$_2$RuO$_4$ transforms gradually into a ferromagnetic state. Above about ~80 kbar the tilt is also lost (space group $Bbcm$) and the ferromagnetic signature ($T_{\text{Curie}}$) in the ac susceptibility appears to be suppressed. Above ~90 kbar superconducting signatures ($T_{\text{SC}}$, magnified by a factor of thirty in the figure) are observed. Signatures of superconductivity are corroborated by two ac susceptibility measurement techniques and by measurements of the electrical resistivity.
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