Quantum criticality in metals

Andrew Schofield

From
Physics World
August 2003

ISSN: 0953-8585

IoP
Institute of Physics Publishing
Bristol and Philadelphia

FEATURE

Phase transitions that occur at absolute zero could provide the key to understanding many of the puzzling properties displayed by novel materials

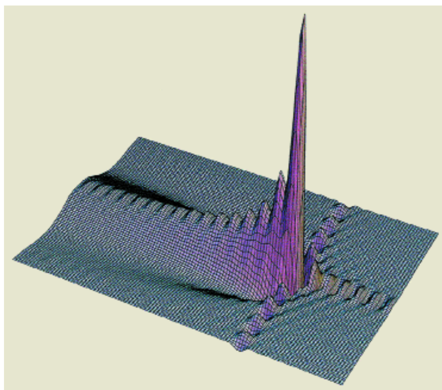
Quantum criticality in metals

Andrew Schofield

In THE mid-1970s, when the study of phase transitions was undergoing a renaissance, a young assistant professor at the University of Chicago asked a question that seemed purely academic: "What would happen to a metal if there was a phase transition at absolute zero?" The answer that John Hertz arrived at involved quantum theory, extra dimensions and a singularity that could undermine our view of the metallic state.

However, a theory about what might happen at the inaccessible temperature of absolute zero did not seem relevant to any conceivable experiment. Hertz promptly switched fields, although his 1976 paper was retained in the collective conscience of condensed-matter physicists, appearing as the final reprint in Philip Anderson's influential book *Basic Notions of Condensed Matter Physics*.

Now all this is changing. Recent advances in materials science and experimental physics mean that Hertz's work is but the opening chapter of what is a highly active research field. Zero-temperature phase transitions may prove to be the unifying theme that we need to understand many puzzles in novel materials.



GILBERT LONZARICH

Critical point – an electron quasiparticle in a metal that is near a quantum phase transition produces a magnetic shock wave, which can cause a breakdown of the conventional metallic state and lead to unusual forms of superconductivity.

Beyond the microscopics

In order to understand the vast collection of electrons and ions in a typical material, new ways of thinking are required. To begin with the Schrödinger equation – which captures the microscopic laws for an individual atom – is like trying to understand the layout of a city by examining it one brick at a time. Instead, we need to discover the principles that govern complex assemblies of atoms. Statistical mechanics does exactly that, and has given us new principles such as the laws of thermodynamics. These laws do not depend on underlying microscopic details, and this is seen most starkly near a phase transition.

When matter is cooled it will make transitions from one state (or phase) to another in a quest to minimize its free energy. This means that disorder wins at high temperatures and states of a random, gaseous nature are favoured. At low temperatures more ordered states are preferred. The transitions between states come in two varieties. The freezing of water into ice at 273 K, for example, occurs abruptly – the water molecules suddenly switch from being free to being fixed rigidly in position. This is called a first-order phase transition. In contrast, when iron is cooled from high temperatures it starts to magnetize at 1043 K, and the degree of magnetization rises smoothly as the temperature is lowered further. This is called a second-order phase transition.

A key feature of second-order phase transitions is the existence of a critical temperature. This is the point at which the transition begins, when the material is in maximum confusion as it equivocates between the two competing phases. In the vicinity of this "critical point", physical properties such as the specific heat capacity can be characterized by a handful of numbers called critical exponents. For example, the magnetization of iron grows as $(1043-T)^{0.36}$ as the temperature, T, is lowered below 1043 K.

Crucially, the same sets of critical exponents appear in transitions that have entirely different microscopic origins, such as the ordering of alloys and the transition between a liquid and a gas. The essential point is that the details of the atoms and molecules in the material are no longer important in specifying its physical properties. This is called universality (see box on page 24).

Quantum phase transitions

So what is different about phase transitions that occur at absolute zero? Well for one thing, if the temperature is fixed at absolute zero then any change of state must result from changes in a different control parameter, such as pressure, magnetic field or even chemical composition. Furthermore, the nature of the phases themselves is quite different at 0 K. Take a metal, in which the electrons are mobile just like the atoms in a liquid. Absolute zero is the temperature at which thermal motion ceases, so how can electrons in a metal remain liquid down to 0 K?

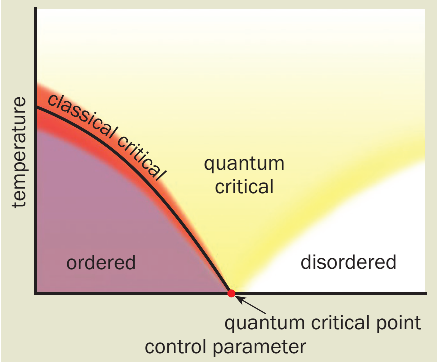
The answer lies in the uncertainty principle, which forbids both the position and momentum of a particle from being fixed. This means that all particles are in a state of quantum agitation at absolute zero, and this "zero point motion" is too violent to allow the electrons in a metal to crystallize. It therefore remains a quantum fluid. But the third law of thermodynamics holds that entropy, which is often a measure of disorder, should vanish at absolute zero. So the zero-temperature quantum fluid is just as unique as a state in which the electrons are absolutely stationary – both have zero entropy. As we will see, a quantum phase transition takes place when there is a change in the nature of the lowest energy quantum state.

The key new feature of a quantum phase transition – which Hertz showed – is that quantum mechanics effectively increases the number of dimensions. In statistical mechanics our description of nature comes from averaging all possible configurations of the particles in space, weighted by the Boltzmann factor, $\exp(-E/k_B T)$, where E is energy and k_B is Boltzmann's constant. This gives us classical thermodynamics. Quantum theory is similarly probabilistic, and quantum probabilities come from averaging all the ways in which a particle moves in time weighted by a Schrödinger factor. Developing a quantum approach to statistical mechanics to understand quantum phase transitions involves combining these two uncertainties.

Curiously, the Boltzmann factor and the Schrödinger factor are very similar in form, which means that a single combined description is possible. The time variable in the Schrödinger factor is replaced by $\hbar/k_B T$ in the Boltzmann factor – where $\hbar$ is Planck's constant divided by 2π – and the two factors also differ by the imaginary number i, the square root of -1. A quantum description of phase transitions is therefore like a classical one except that the configurations vary in space and additionally in "imaginary time".

This unification of quantum theory and statistical mechanics has important implications for critical points because one of the few factors determining the universality class is the number of dimensions. Imaginary time now appears as an extra dimension and its effect on the universality class is to change the critical exponents.

1 Quantum criticality in theory



Understanding a quantum critical point involves quantum theory, extra dimensions and singularities. By changing a control parameter, such as pressure or chemical composition, the temperature of a transition to an ordered state (black line) can ideally be suppressed to 0 K. This defines a quantum critical point that separates a classically ordered state (purple region) from a quantum "disordered" state (white). In between (yellow) the material can be in a state of quantum criticality at temperatures above absolute zero.

Given our belief that quantum mechanics describes the underlying microscopic behaviour of nature, this begs the question of why the appearance of a new dimension in the quantum description has not invalidated previous classical thermodynamic results? The answer is that at any temperature above absolute zero, the new time dimension is unimportant near the phase transition. When the correlation length of fluctuations in the imaginary time direction becomes greater than $1/T$, quantum mechanics becomes just like another microscopic detail – which is unimportant to universality – and we enter a classical critical region.

This is reminiscent of the extra spatial dimensions that are proposed in some theories of quantum gravity, and which "curl up" so that they are not apparent on everyday length scales. However, for a phase transition at $T = 0$ K the curled-up length is infinite, so the quantum dimension does not disappear. Some 10 years ago Andrew Millis, while at Bell Labs, calculated the effect that the quantum fluctuation correlation length would have just before it reached the curling-up scale. He found that it would lead to a region of "quantum criticality" even at finite temperatures (figure 1). It turns out that these effects can not only be seen, they also have a devastating effect on the nature of the metal.

Quantum criticality observed

Finding a phase transition in nature that occurs at precisely 0 K would appear to require an enormous amount of luck. But since the mid-1990s advances in materials and techniques have made it possible to tune phase transitions down to absolute zero. One of the richest sources of tuned quantum phase transitions comes from metallic magnets that have low transition temperatures.

Universality

Second-order phase transitions have universal properties that do not depend on the microscopic details of the material. The origin of this "universality" lies in the gradual nature of the transitions. Each phase of matter describes the most probable arrangement of its constituent atoms, but the atoms can fluctuate around this configuration. As the transition temperature is approached from the high-temperature side, say, regions of the low-temperature phase will spontaneously appear and disappear, driven by the ambient temperature.

Far from the critical temperature, these fluctuations are small islands in a sea of the high-temperature phase. But as the temperature approaches the critical temperature, these islands grow with a characteristic size – called a correlation length – that increases towards infinity at the critical temperature itself. At this point it is no longer possible to identify the typical size of an island, or indeed which of the two phases is the "island" and which is the "sea". These are the critical fluctuations that dominate measurable quantities.

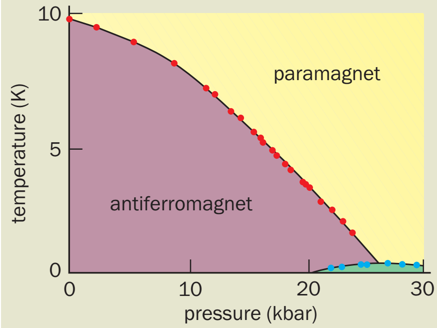
When the correlation length is much larger than the atomic size, the properties of a material are determined by general features of the phase transition, rather than physical laws that govern the atomic scale. Symmetry is one such feature. A more ordered low-temperature phase has less symmetry than a disordered high-temperature phase. When iron magnetizes below 1043 K, for example, its atoms develop a preferred orientation with respect to the Earth's magnetic field. Above 1043 K there is no preference and all directions look the same. Symmetry changes can be classified into types. In the case of iron, the ordered state chooses one direction from all possible orientations.

The other factor influencing the phase transition is dimensionality. Very thin films of a material, for example, can effectively behave as if they are 2D when the correlation length is longer than their thickness. Despite having very different microscopic properties, second-order phase transitions that exhibit the same type of symmetry change and the same number of spatial dimensions all possess the same critical exponents. In other words, they fall into the same universality class. Temperature also appears to play a dual role in phase transitions, both driving the fluctuations and locating the critical point.

Metals that are on the cusp of magnetism were studied long before Hertz's paper. In particular, Tôru Moriya at the Science University of Tokyo anticipated the results of Hertz and Millis by studying the effect that random fluctuations of the electron spins have on a metal. In the early 1970s he found that the fluctuations become increasingly influential near a phase transition, when the spins start to become more ordered.

One of the first examples of tuned quantum criticality was seen in the cerium–palladium–silicon compound $CePd_2Si_2$ by Gilbert Lonzarich, Stephen Julian and co-workers at Cambridge University in 1996. In this material the cerium ions behave as small magnets that arrange themselves in an alternating antiferromagnetic pattern at temperatures below about 10 K. By applying very high pressures the researchers were able to progressively reduce the ordering temperature, and effectively squeeze the magnetism out of the metal (figure 2).

2 Tuning in to absolute zero



Quantum criticality has been observed in the metal $CePd_2Si_2$. The ordered state (purple region) is antiferromagnetic, which is suppressed to zero by high pressure, while the disordered state is paramagnetic (yellow). The quantum critical point occurs at 28 kbar, which has a dramatic effect on the metal and influences a wide region of the phase diagram. For example, in an attempt to avoid the singularity of the critical point itself, this metal becomes a superconductor (green region, temperature enhanced by a factor of 10).

3 Scattering singularities

Just as planes leave hazardous vortices in their trail that can scatter other aircraft, quasiparticles in metals that are on the border of magnetism can leave behind a disturbance that can scatter other quasiparticles. At a quantum critical point these vortices become infinitely long-lived and result in a scattering singularity.

When the pressures reach almost 30 000 times that of the atmosphere, the ordering temperature is suppressed to absolute zero and the metal is tuned to a quantum critical point. At this point the metal is pushed to the limits of our understanding of the conventional quantum-liquid state. The Cambridge group found a metal that had a resistivity with an unusual temperature dependence up to 40 K – which is well away from the quantum critical point – and became a superconductor at the very lowest temperatures (see Mathur *et al.* in further reading and *Physics World* October 1998 pp22–23).

Observations of quantum criticality are no longer limited to magnetic phase transitions. For example, a new type of quantum phase transition has recently been seen in an organic material. Sachio Horiuchi and co-workers at the National Institute of Advanced Industrial Science and Technology in Japan found that the nature of the chemical bonds holding this material together changes at its quantum critical point. By changing either the pressure or chemical composition they have transformed the Van der Waals attraction into ionic bonding. The electric charge undergoes quantum critical fluctuations, and is rearranged at the transition point.

Furthermore, quantum criticality is not restricted to exotic materials. Last year a team led by Gabriel Aeppli at the NEC Research Institute in New Jersey and Thomas Rosenbaum at the University of Chicago found a quantum phase transition in a surprisingly simple metal – chromium (see Yeh *et al.* in further reading and *Physics World* January 2003 pp20–21). Their findings emphasize that whenever a quantum critical point occurs in a metal, it seems to profoundly affect the metallic quantum fluid.

Our conventional picture of the metallic quantum fluid derives from Lev Landau's Fermi-liquid theory, which was developed in the 1950s. The quantum ground state defines a "Fermi surface" that separates the quantum states that are occupied by quantum particles and those that are empty. The quantum states are standing-wave configurations, while the quantum particles are electron-like fermions that are usually called quasiparticles. (Fermions are particles that satisfy the Pauli exclusion principle, which forbids more than one fermion from occupying a given quantum state.) The Landau Fermi liquid is the starting point for almost all that we know about metals, including the development of magnetism and superconductivity.

Two conditions must be met for a quasiparticle to scatter in such a way that it disrupts the Fermi surface. The state that it scatters into must be unoccupied, and the quasiparticle that it scatters from must also find an empty state to scatter into. These two constraints lead to the scattering rate being proportional to the square of the temperature. As a result, the scattering rate vanishes sufficiently quickly to preserve the Fermi surface as the temperature is reduced.

Bob Laughlin of Stanford University and David Pines of the Los Alamos National Laboratory have coined the term "quantum protectorate" to describe such cases where the properties of materials are not sensitive to scattering and disorder. However, in the case of the Cambridge study of $CePd_2Si_2$, a resistivity of $T^{1.2}$ was being seen near the quantum critical point. This suggests that even Fermi-liquid theory is being pushed to its limits.

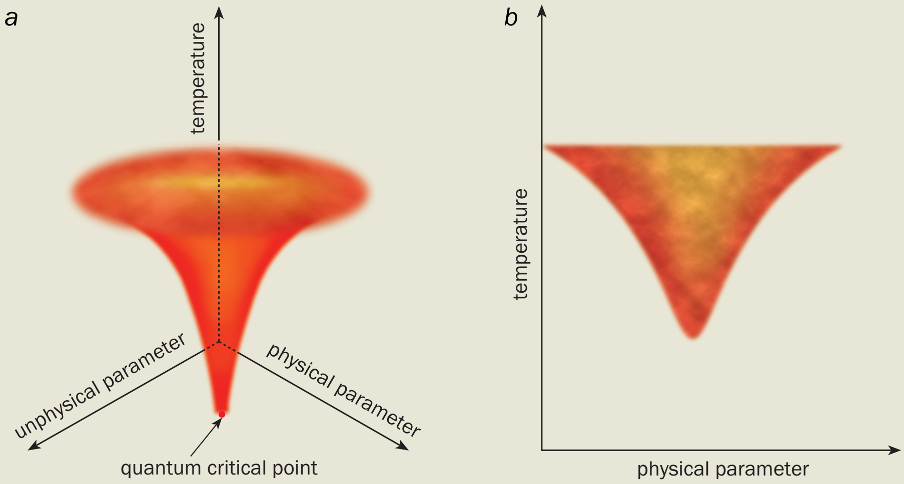
It is the quantum fluctuations themselves that are threatening the quantum protectorate at the critical point. As a quasiparticle moves through the quantum critical metal, it leaves a trail in the fluctuating magnetic background – rather like the vortex trail left by a large aircraft as it comes into land (figure 3). Air-traffic controllers space out plane landings to allow these vortices to decay, thus preventing them from "scattering" the next plane. However, near a quantum critical point the quasiparticle vortices take an increasingly long time to decay, which increases the scattering probability.

At the critical point itself the scattering probability diverges to infinity and we are left with a kind of scattering maelstrom in the metal – a singularity. In $CePd_2Si_2$ it seems that the metal finds a way of avoiding the singularity by forming a new quantum protectorate, otherwise known as the superconducting state. Here a quasiparticle is attracted by the trail left behind by another quasiparticle, providing the "glue" that binds them together to form superconducting pairs. In other words, quantum criticality can both destroy the conventional quantum liquid and also promote the appearance of new states.

Beyond the Hertz picture

The theoretical ideas of Hertz, Millis and Moriya do not appear to account for all of the observations near a quantum criticality. In 1995 Richard Hlubina and Maurice Rice at ETH Zurich in Switzerland pointed out that the scattering singularity should only scatter those pairs of quasiparticles that have a wavelike interference pattern that matches the wavelength of the underlying magnetic order. This restriction would leave most of the quasiparticles unscathed. Although some of the quasiparticles would be strongly affected by the critical singularity, the vast majority would be insensitive to it and the resistivity should maintain a conventional T^2 form.

4 Criticality without a critical point



It is possible that new materials with unusual properties can display quantum-critical fluctuations, while having no accessible zero-temperature quantum critical point. (*a*) The singularity at the critical point would only be visible if you could tune some unphysical control parameter such as negative pressure. (*b*) In the physical world the critical point would be hidden, but the material would be close enough to a quantum phase transition that its effects would be observable at temperatures above absolute zero.

However, this does not appear to happen experimentally and, moreover, singular scattering seems to persist at temperatures far from the critical point. Achim Rosch at the University of Karlsruhe in Germany has argued that this may be a consequence of the residual disorder left in these materials, even though they are highly purified.

Experiments at another antiferromagnetic quantum critical point suggest a more radical departure from Hertz's picture. The cerium–copper compound $CeCu_6$ can be tuned to an antiferromagnetic quantum critical point when 1.7% of the copper is replaced by gold. This quantum critical point has been studied extensively by Hilbert von Löhneysen and colleagues at Karlsruhe since 1993, who have also found unusual temperature dependencies that suggest the Fermi-liquid picture is being pushed to its limits.

When Almut Schröder of the University of Karlsruhe and co-workers used neutrons to observe the nature of the magnetism near the critical point in 2000, they unexpectedly found critical fluctuations with a wide range of wavelengths. This implies that the critical fluctuations are localized in space, with each part of the material fluctuating independently, and offers the possibility of a more dramatic destruction of the Fermi-liquid state. But it leaves open the question of what could be happening locally?

Part of the answer may lie in our choice of materials for many of these experiments. Compounds containing cerium form metals that are already pushing this Fermi-liquid quantum protectorate to the limits. The outermost electrons in cerium atoms form a magnetic moment that is tightly bound to its ion. When cerium is alloyed with non-magnetic metals these magnetic moments scatter the conducting electrons. However, at low temperatures the conduction electrons have the last laugh.

In an effect named after Jun Kondo, the conduction electrons "suck" the spins away from their ions to form a Fermi liquid of quasiparticles that contains both the conduction electrons and the cerium magnetism (see *Physics World* January 2001 pp33–38). These quasiparticles are up to 1000 times heavier than an ordinary electron and this makes them much easier to tune with pressure and chemical composition than ordinary metals. But it also means that the magnetic quantum critical point in these compounds is in competition with the Kondo effect.

In 2001 Qimiao Si and colleagues from Rice University in Texas and Kevin Ingersent of the University of Florida showed that in two spatial dimensions the competition between magnetic order and the Kondo effect can give rise to the sort of local criticality that is seen in gold-doped $CeCu_6$. In this material the quantum dimension dominates the fluctuations so that temperature – i.e. the size of that dimension – provides the only energy scale.

Piers Coleman of Rutgers University in the US and Catherine Pépin of CEA-Saclay in France have recently suggested an alternative interpretation. They suggest that the Fermi-liquid side of the critical point is dominated by fermionic quasiparticles, while the excitations on the magnetic side behave like bosons, which do not obey the exclusion principle. This raises the fascinating possibility that the critical point could itself be a point of supersymmetry that links bosons and fermions together.

Unified picture

What researchers in the field of quantum criticality would really like to know without ambiguity is how the Fermi surface itself behaves at the quantum critical point. Is it eaten up by the singularity there? Or does it change smoothly between one quantum liquid state and another? Recent experiments with doped chromium suggest that dramatic changes are taking place, and answering these questions more definitively poses a significant challenge to experimentalists. However, so did obtaining a quantum critical point in the first place.

But what of the materials that have unusual properties yet do not display any obvious phase transitions? Recent work by the author involves a new type of quantum criticality that has recently been seen in a ruthenate-oxide metal, $Sr_3Ru_2O_7$. The tuning parameter in $Sr_3Ru_2O_7$ is a magnetic field and the material displays critical fluctuations without an ordered phase. These fluctuations originate from the critical end-point of a first-order transition that occurs at 0 K with minimal tuning.

In fact, just being close to a quantum phase transition is enough to see quantum critical behaviour. Imagine a material that would have a quantum phase transition if only you could tune it slightly with some impossible tuning parameter, such as negative pressure. When the phase diagram is plotted with the unphysical parameter as one axis and a real parameter such as magnetic field on the other, you would see the cone of quantum-critical behaviour emanating from the critical point (figure 4). However, in an actual experiment you would only observe the intersection of that cone on the physical axis. It would be like seeing the ghost of the quantum criticality without a quantum phase transition.

Oxide metals, such as the ruthenates, have many quantum ground states and often show unusual metallic properties (see *Physics World* April 2002 pp 33–38). This makes them prime candidates for hidden criticality. The high-temperature superconducting cuprates may also fall into this category, and many researchers have remarked on the similarity between their phase diagrams and those of a quantum critical point (see Sachdev in further reading). The temperature scales may differ, but the superconductivity and power-law resistivities are highly suggestive of quantum critical behaviour.

John Hertz modestly remains flattered that anyone is interested in his work. However, it seems that the notion of a quantum critical point may soon be able to offer a unified picture of the breakdown of the metallic quantum fluid.

Further reading

P Coleman and C Pépin 2003 Heavy electron quantum criticality arXiv.org/abs/cond-mat/0302001

J Hertz 1976 Quantum critical phenomena *Phys. Rev.* B **14** 1165–1184

S Horiuchi *et al.* 2003 Quantum phase transition in organic charge-transfer complexes *Science* **299** 229–232

N Mathur *et al.* 1998 Magnetically mediated superconductivity in heavy fermion compounds *Nature* **394** 39–43

S Sachdev 1999 Quantum phase transitions *Physics World* April pp33–38

S Sachdev 2000 Quantum criticality: competing ground states in low dimensions *Science* **288** 475

A Yeh *et al.* 2002 Quantum phase transition in a common metal *Nature* **419** 459–462

Andrew J Schofield is in the School of Physics and Astronomy, University of Birmingham, Edgbaston, Birmingham B15 2TT, UK, e-mail ajs@th.ph.bham.ac.uk