Phase transitions are normally associated with changes of temperature but a new type of transition – caused by quantum fluctuations near absolute zero – is possible, and can tell us more about the properties of a wide range of systems in condensed-matter physics

Quantum phase transitions

Subir Sachdev

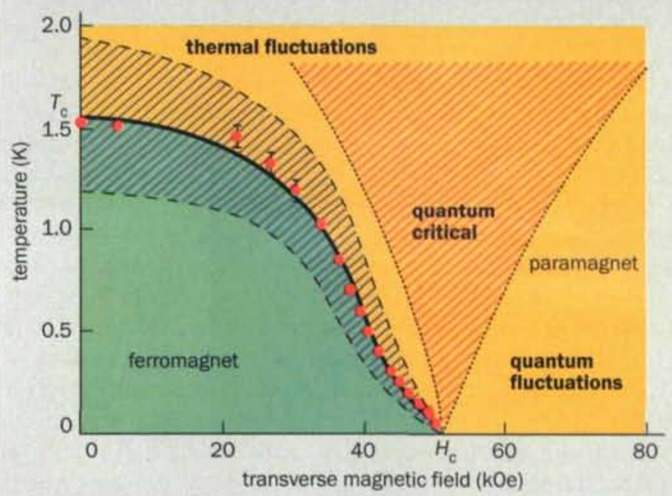
NATURE abounds with phase transitions. The boiling and freezing of water are everyday examples of phase transitions, as are more exotic processes such as superconductivity and superfluidity. The universe itself is thought to have passed through several phase transitions as the high-temperature plasma formed by the big bang cooled to form the world as we know it today.

Phase transitions are traditionally classified as first or second order. In first-order transitions the two phases co-exist at the transition temperature – e.g. ice and water at 0 °C, or water and steam at 100 °C. In second-order transitions the two phases do not co-exist. Two familiar and historically important examples of second-order phase transitions are the Curie point of a ferromagnet and the critical end-point of carbon dioxide. The Curie point is the temperature above which the magnetic moment of a material vanishes. The phase transition occurs at a point where thermal fluctuations destroy the regular ordering of magnetic moments. The critical end-point of carbon dioxide – the end-point of the line that separates the liquid and gaseous states in the pressure–temperature plane – can also be interpreted in terms of competition between order and thermal fluctuations. The study of phase transitions, and such second-order phase transitions in particular, has been one of the most fertile branches of theoretical physics in the 20th century.

At first glance it might appear that the study of such special points in the phase diagram is an abstruse problem of interest only to specialists. However, developments in the last three decades have clearly established the contrary. Insights gained from studies of second-order phase transitions have influenced wide areas of physics, such as the field theories that underpin elementary particle physics.

In the last decade, attention has focused on phase transitions that are qualitatively different from the examples noted above: these are quantum phase transitions and they occur only at the absolute zero of temperature. The transition takes place at the "quantum critical" value of some other parameter such as pressure, composition or magnetic field strength. A quantum phase transition takes place when co-operative ordering of the system disappears, but this loss of order is driven solely by the quantum fluctuations demanded by Heisenberg's uncertainty principle.

1 Quantum criticality in a ferromagnet



The temperature–magnetic-field phase diagram of $LiHoF_4$ as measured by Bitko and co-workers. The full black line is a line of phase transitions separating the ferromagnet (green) and the paramagnet (yellow). A classical phase transition takes place at $H=0$, $T=T_c$ and a quantum phase transition occurs at the point $H=H_c$, $T=0$. An effective classical theory applies within the blue hatched region, while the influence of the quantum critical point is predominant in the red hatched region. Bitko and co-workers used measurements of magnetic susceptibility to determine the different Curie points (red circles).

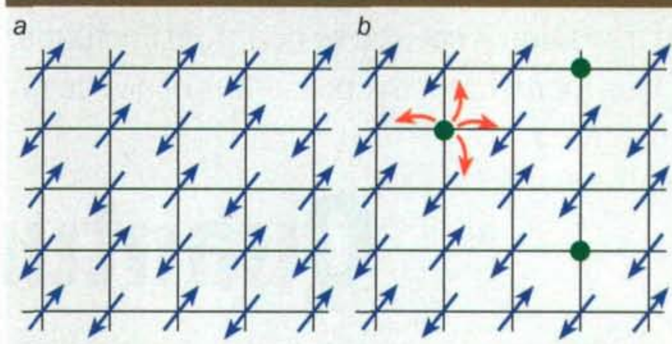
The physical properties of these quantum fluctuations are quite distinct from those of the thermal fluctuations responsible for traditional, finite-temperature phase transitions. In particular, the quantum system is described by a complex-valued wavefunction, and the dynamics of its phase near the quantum critical point requires novel theories that have no analogue in the traditional framework of phase transitions.

Again, it might appear that the study of quantum phase transitions is only of academic interest, as such transitions occur at only one value of a special parameter at the experimentally impossible temperature of absolute zero. However, recent experimental and theoretical developments have made it clear that the presence of such a zero-temperature quantum critical point holds the key to understanding a wide range of behaviour in many condensed-matter systems. Examples include rare-earth magnetic insulators, high-temperature superconductors and two-dimensional electron gases.

As we shall see below, the magnetic properties of all these materials cannot be described in terms of any simple independent-electron picture: rather the electrons behave cooperatively, and the study of such "correlated electron systems" is currently a rapidly developing branch of theoretical physics. Similar behaviour has been observed in magnetic transitions in transition metal and rare-earth alloys, the superconductor–insulator transition in thin films and junction arrays, and the metal–insulator transition in alloys and amorphous systems.

Once the quantum critical point of such a system has been identified, it can be used as a point of departure to investigate the entire phase diagram. Indeed, quantum critical behaviour can be observed at temperatures well in excess of room temperature. This new approach is succeeding where more familiar approaches have failed.

2 Magnetic interactions in a superconductor



(a) The lattice of copper ions in a single layer of La_2CuO_4. Each ion has a free spin, and at zero temperature these spins exhibit antiferromagnetic order. (b) The addition of strontium ions has the effect of introducing "holes" (green) into the spin lattice. These holes can hop between lattice sites, as indicated by the red arrows.

Quantum Ising systems

Magnetism, and magnetic interactions and ordering will be common features of all the systems we describe. The spin of the electron is the key to magnetism, and the magnetic properties of most materials are determined by the distribution of unpaired electron spins within the material. A common approach to studying quantum phase transitions is to determine the lowest energy or "ground" state of the material at zero temperature and a fixed value of some variable such as pressure. It is also important to know the low-energy excitations of the system. It is often found that the ground state will be qualitatively different at different values of the relevant variable. This means that the material must undergo a phase transition as this variable is changed – even at absolute zero. This is the quantum phase transition.

Consider the ionic crystal lithium holmium fluoride ($LiHoF_4$). The magnetic properties of this rare-earth insulator are well understood on the microscopic scale. Most notably, at temperatures below about 2 K the only important magnetic excitations reside on the holmium ions (which have a charge of +3) and the spins on neighbouring holmium ions are coupled by the magnetic dipolar interaction. These spins prefer to point either "up" or "down" relative to a certain crystalline axis. Such an arrangement of spins on a lattice is known as an Ising model. The geometry of the crystal is such that, in the absence of an external magnetic field, the lowest energy state is very simple: either all the spins point up or they all point down. In other words, $LiHoF_4$ is a fully polarized ferromagnet.

In 1996 David Bitko and Thomas Rosenbaum of the University of Chicago, and Gabriel Aeppli of NEC Research in Princeton investigated the behaviour of $LiHoF_4$ as a function of temperature, T, and an external magnetic field. First, consider the case of no magnetic field (the y-axis in figure 1). Initially thermal fluctuations caused a small number of spins to flip: that is if the crystal was in a ground state with all the spins pointing up, thermal fluctuations caused some spins to point down, and vice versa. As the temperature was increased, the number of minority spins also increased until there was an equal number of up and down spins. This was a conventional, second-order phase transition, driven entirely by thermal fluctuations, from a ferromagnetic state to a paramagnetic state at the Curie temperature.

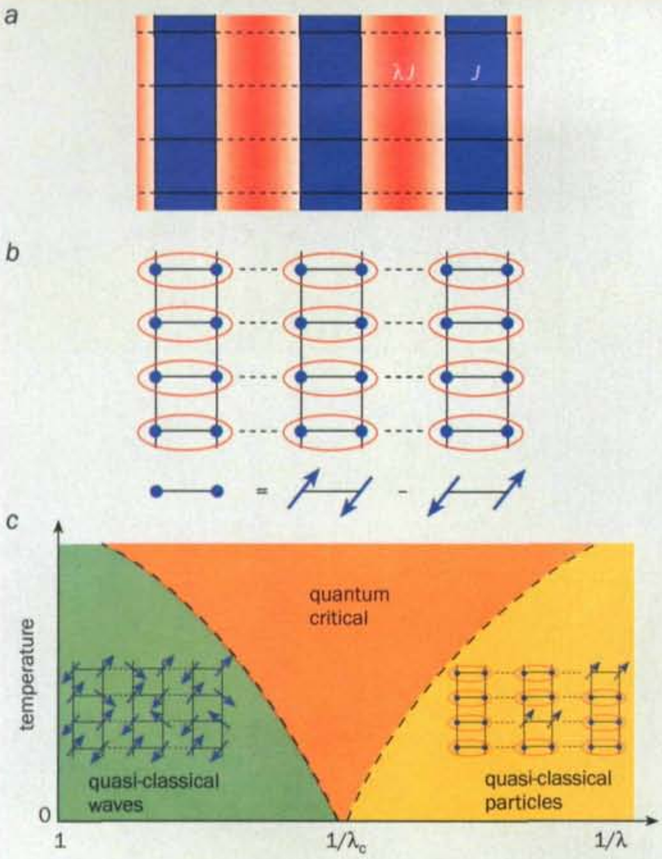
The novel property of $LiHoF_4$ is that it is possible to destroy the ferromagnetic order by turning an entirely different knob, even at absolute zero. An external magnetic field, H, applied at right angles to the Ising spin orientation, allows quantum tunnelling to take place between the up and down spin states. If this field is larger than a critical field, H_c, these tunnelling events are frequent enough that the ground state becomes a paramagnet, even at zero temperature. In other words, a phase transition driven entirely by quantum fluctuations can occur.

However, unlike the earlier thermal transition, it is now quite misleading to think of the paramagnetic state as having spins that are fluctuating between the up and down states in real time; rather there is a unique phase-coherent wavefunction for the ground state that is a quantum superposition of the up and down states. The properties of this quantum paramagnet are well understood.

So what happens in the remainder of the phase diagram, when both H and T are non-zero? The classical phase transition at zero field and the quantum phase transition at zero temperature are connected by a line of second-order phase transitions that separates the ferromagnet from the paramagnet. The dynamic properties of the thermal paramagnet are quite distinct from those of the quantum paramagnet, but these two regions of the phase diagram are nevertheless continuously connected without any intervening thermodynamic singularity. Instead there is a smooth crossover through an intermediate "quantum critical" region (figure 1). In a sense, quantum and thermal fluctuations are equally important in this region, and conventional models do not apply here. Developing the correct theoretical framework to explain the behaviour observed in this regime has been an important focus of recent research. In particular, as we shall see, many macroscopic properties are determined by just the temperature and the fundamental physical constants, like the charge and magnetic moment of the electron. Put another way, the macroscopic properties of the quantum critical regime are often independent of microscopic details.

Some readers will be aware that strong fluctuations in the vicinity of a second-order phase transition lead to a divergence in the magnetic susceptibility, and that this divergence is characterized by a "critical" exponent that is also independent of the material properties. In fact, the critical divergence observed near the classical transition at zero field also applies sufficiently close to the entire phase boundary, with the excep-

3 Stripes in a superconductor



(a) Segregation of holes in a copper-oxide layer into hole-rich stripes (red) and hole-poor regions (blue). Interactions between spins within each hole-poor region (solid line) have an antiferromagnetic exchange energy J. Interactions between spins in neighbouring hole-poor regions (dashed line) have a smaller exchange energy, λJ, where $0<\lambda<1$. λ can be varied by varying the level of doping. The spins in the hole-rich stripes (not shown) are assumed to only mediate the λJ exchange. (b) For small λ, valence bonds (shown in red) form between neighbouring spins. Each bond is a quantum linear superposition of two spin configurations in which the total spin, S, is zero. (c) The phase diagram as a function of temperature and $1/\lambda$. The only singularity is at the quantum critical point, $\lambda=\lambda_c$ and $T=0$. The dashed lines indicate the locations of smooth crossovers. The excitations in the quasi-classical wave regime (green) are spin waves. The excitations in the quasi-classical particle regime (yellow) are broken valence bonds with S = 1. These excitations can hop between sites like particles. No effective classical picture applies in the quantum critical regime (orange).

tion of the quantum critical point itself at absolute zero (figure 1). This means that classical behaviour can always be observed sufficiently close to the phase boundary. Indeed, the quantum critical region is actually farther away from the phase boundary than this classical region, except at the quantum critical point.

High-temperature superconductors

In 1986 Georg Bednorz and Alexander Müller of IBM Research in Zurich discovered that a class of ceramic compounds was able to conduct electricity without resistance – i.e. they were superconducting. These materials consisted of two-dimensional layers of copper oxide with lanthanum and strontium ions in between. The particular compound studied by Bednorz and Müller, $La_{1.85}Sr_{0.15}CuO_4$, had a superconducting transition temperature of 30 K, which was significantly higher than any other superconductor at the time – hence the name high-temperature superconductivity.

Since then, other copper oxides with transition temperatures as high as 130 K have been discovered and a vast number of experimental techniques have been used to probe their physical properties. These include measurements of a quantum phase transition at which the magnetic order vanishes. The insight gained from understanding the magnetic quantum critical point should have significant implications for the superconductivity itself, which is still not fully understood, although that is beyond the scope of this article.

First, let us consider one of the parent compounds of the cuprate superconductors: La_2CuO_4 is an insulator, and its important low-energy excitations consist primarily of spin fluctuations on the copper ions, which have a charge of +2. These ions are stacked in layers, with the copper ions placed on the vertices of a square lattice. The coupling between adjacent layers is quite small, so we can consider the properties of a single two-dimensional layer. Each copper ion has a single unpaired electron, and has total spin $S=½$. The interactions between the electrons are antiferromagnetic: that is neighbouring spins tend to point in opposite directions.

If the spins were classical objects, the ground state of the system would be quite simple: the spins would arrange themselves in a checkerboard pattern containing two sublattices: the spins would point "up" in one sublattice and "down" in the other (figure 2*a*). The quantum mechanical system is considerably more complicated, but it is now well established that its ground state is qualitatively similar to that of the classical model. However, the average magnetic moment on each site has been reduced from $S=½$ due to quantum fluctuations. The low-lying excitations above this ground state consist of "spin waves". These are slow variations in the local direction of the antiferromagnetic order throughout the system. This combination of an $S=½$ antiferromagnetic ground state and spin-wave excitations is found in several systems that undergo quantum phase transitions.

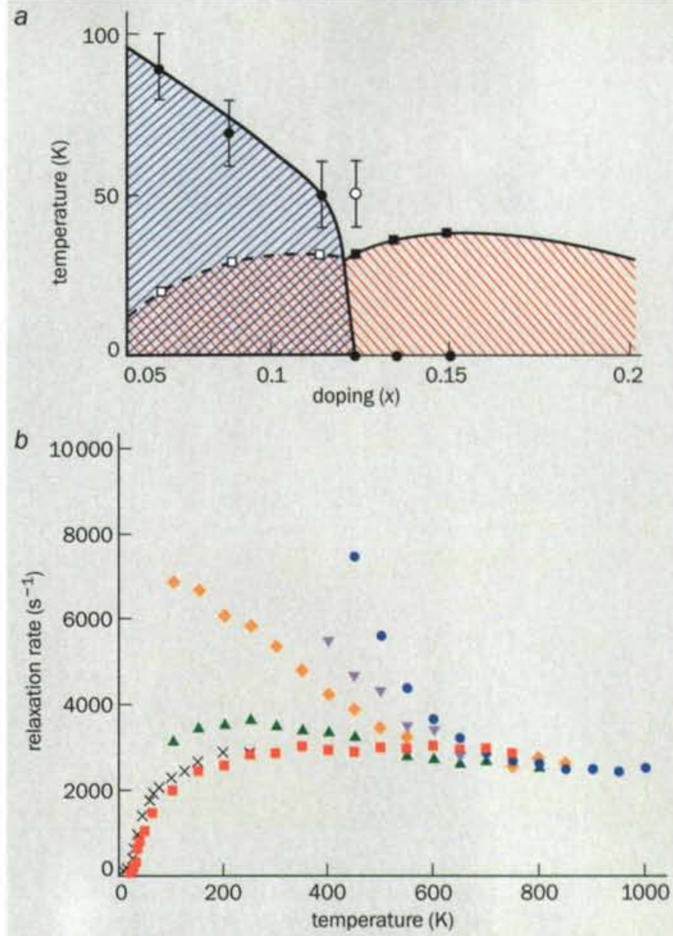
At any non-zero temperature, a large number of these low-energy spin waves are excited. The resulting thermal state has two crucial properties: (a) it allows the system to be described in terms of quasi-classical waves; (b) the nonlinear interactions between the spin waves are particularly strong in two dimensions, leading to large fluctuations in the local orientation of the antiferromagnetic order and hence to the destruction of any long-range order.

So far we have only considered thermal fluctuations. To access a quantum critical point we must remain at $T=0$ and vary a material parameter. The most common choice is to replace the lanthanum ions with another metallic ion such as strontium to form $La_{2-x}Sr_xCuO_4$, where x is the amount of doping. Superconductivity appears at $x\approx5\%$, but we are interested in the quantum phase transition in the magnetic order that takes place at $x\approx12\%$.

The strontium ion has one less electron to donate to the copper oxide layers than lanthanum, so the effect of doping is to introduce "holes" into the square-lattice antiferromagnet. These holes can move through the lattice by a quantum hopping process (figure 2*b*). The physics of such a "diluted" quantum antiferromagnet is incredibly rich.

A number of recent neutron-scattering experiments, especially those performed by John Tranquada and co-workers at the Brookhaven National Laboratory in the US, have provided crucial new understanding of the properties of diluted antiferromagnets. Experiments have shown that the holes like to concentrate in "stripes" in the material that are a few lattice spacings wide and about the same distance apart. Moreover, the holes tend to move only along these stripes (figure 3*a*). Interspersed between these "hole-rich" stripes are regions of undoped, insulating antiferromagnet. Aspects of this stripe formation were in fact predicted theoretically in 1989 by Jan Zaanen of the Lorentz Institute in Leiden and Olle Gunnarsson of the Max Planck Institute in Stuttgart, and also by the late Heinz Schulz of the University of Paris-Sud in Orsay.

4 Relaxation times and quantum criticality



(*a*) The phase diagram of $La_{2-x}Sr_xCuO_4$ as measured by magnetic resonance experiments at MIT (Hunt *et al.*). There is quasi-static charge stripe order over the blue hatched region. Superconductivity is present in the red hatched regions. The measurements indicate quasi-classical wave (particle) behaviour at low (high) doping. (*b*) Measurements of the nuclear relaxation rate as a function of temperature for zero doping (blue circles), $x = 0.02$ (purple triangles), $x = 0.04$ (orange diamonds), $x = 0.075$ (green triangles), $x = 0.115$ (black crosses) and $x = 0.15$ (red squares). The relaxation time is a measure of the density of states for the low-energy spin excitations. The three types of behaviour shown in figure 3c can be observed in these measurements, as discussed in the text.

Spins and stripes

We can construct a simple "toy" model for the destruction of the magnetic order as a function of doping in the presence of these charge-ordered stripes. Imagine that the spins in the hole-poor regions interact with their nearest neighbours by an antiferromagnetic exchange of energy J, and that the spins at the boundaries of two neighbouring hole-poor regions are coupled by a weaker exchange of magnitude λJ, where λ has a value between zero and one. We ignore the spins in the hole-rich stripes and assume that their only role is to mediate the coupling λJ. We have now defined an $S = ½$ antiferromagnet on an anisotropic two-dimensional lattice. In this simple picture the value of λ can be changed by altering the amount of strontium in the superconductor. The ground state of this system will evolve as a function of λ and, as we shall see, undergo a quantum phase transition at a critical value, $\lambda = \lambda_c$.

At $\lambda = 1$ this model reverts to the ordinary square lattice $S = ½$ antiferromagnet shown in figure 2*a* and its ground state exhibits long-range magnetic order. Now consider the ground state for $\lambda \ll 1$. Let us assume that each hole-poor region consists of a pair of coupled one-dimensional spin chains. A reasonable trial wavefunction for the ground state is to assume that neighbouring pairs of spins form "valence" bonds in which their orientation is a linear superposition of "up" and "down" such that there is no net magnetic moment on each site (figure 3*b*). While this trial wavefunction is not quantitatively correct, it has all the correct qualitative properties, which is currently sufficient. The total spin, S, of this ground state is zero and it is known as a quantum paramagnet. Finally, a finite amount of energy (of order J) is required to create an excitation above such a ground state: a valence bond must be broken and the spins will then point in the same direction to form an $S = 1$ triplet state.

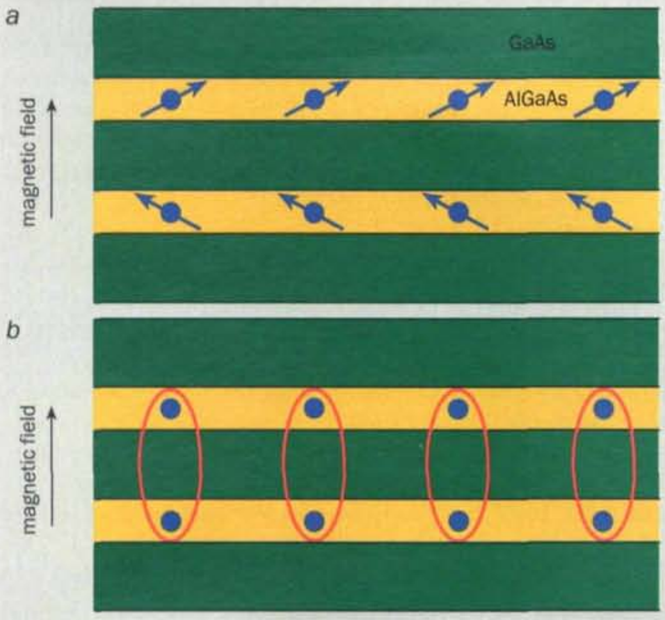
Since this system has two qualitatively different ground states at small λ and $\lambda \approx 1$, there must be a quantum phase transition at some critical value of λ or the doping. The properties of this transition have been examined by many theorists – notably Sudip Chakravarty of the University of California at Los Angeles, Andrey Chubukov of the University of Wisconsin, Bertrand Halperin and David Nelson of Harvard University, Matthias Troyer of ETH Zurich, and the author – and now appear to be quite well understood. The phase diagram contains only one thermodynamic singularity, the quantum critical point at $\lambda = \lambda_c$ (figure 3*b*). Unlike $LiHoF_4$ there are no phase transitions at temperatures above absolute zero, although different regions of the phase diagram exhibit distinctly different physical properties.

For $\lambda > \lambda_c$ there is a regime of quasi-classical wave dynamics that we discussed earlier: the ground state displays antiferromagnetic ordering and the predominant excitations are spin waves. For $\lambda < \lambda_c$ there is a complementary regime of quasi-classical particle dynamics: in the ground state the spins are paired in singlet ($S = 0$) valence bonds and the excitations are a dilute concentration of triplet ($S = 1$) states. These excitations can hop from site to site, much like particles, and their de Broglie wavelength is much smaller than the inter-particle spacing, which allows an effective classical description.

In between these two quasi-classical regimes is the fascinating quantum critical regime. The crossover from the quasi-classical particle regime to quantum criticality occurs when the spacing between the particles becomes of the order of their de Broglie wavelength. Similarly, the crossover from the quasi-classical wave regime to the quantum critical regime happens when short-wavelength spin waves, which cannot be treated classically, proliferate. Neither quasi-classical picture is applicable in the quantum critical regime and more sophisticated theoretical tools are necessary to describe the dynamics.

A key property of the quantum critical regime is that quantum interference effects operate up to a characteristic time, the phase-coherence time, which is given by hC/k_BT, where h

5 Magnetic ordering in 2-D electron gases



Electrons are confined within two two-dimensional quantum wells in a structure consisting of alternating layers of gallium arsenide (green) and aluminium gallium arsenide (yellow). The two possible magnetic phases shown have been observed in recent experiments. These phases are separated by a quantum phase transition. The phase diagram can be explored by varying the spacing between the layers, the direction of the magnetic field (not shown) through the sample and various other parameters.

is the Planck constant, k_B is the Boltzmann constant and C is a universal number that is independent of microscopic details. This remarkable dependence of the phase-coherence time on the fundamental constants of nature and nothing else also applies to a number of relaxation rates and transport coefficients that can be measured in neutron-scattering or nuclear magnetic resonance experiments.

Experiments on superconductors

Various experimental techniques can be used to test the theoretical picture outlined above. Initial evidence for the localization of holes, possibly in stripes, appeared in measurements by Gregory Boebinger of the Los Alamos National Laboratory and collaborators in 1996. More details emerged in ingenious experiments by Takashi Imai's group at the Massachusetts Institute of Technology (MIT). Imai and co-workers used measurements of nuclear quadrupole resonance in copper-63 to determine which parts of the T–x phase diagram exhibited charge stripes (figure 4*a*). Within the striped region, they were able to follow the spin dynamics by measuring a nuclear magnetic resonance relaxation rate, and observed three distinct types of behaviour that neatly correspond to the three regions of the theoretical phase diagram (figure 4*b*).

For small values of the doping, x, the relaxation rate increases rapidly as the temperature is lowered: this is precisely the behaviour expected in the quasi-classical wave regime. At higher temperatures, the relaxation rate is independent of both temperature and the amount of doping, as expected in the quantum critical region. Notice also that for certain intermediate values of x the relaxation rate remains independent of temperature down to rather low temperatures, as we would expect for $\lambda \approx \lambda_c$. Subsequent neutron-scattering experiments by Aeppli, Thomas Mason of the Oak Ridge National Laboratory and others have confirmed this picture by observing spin correlations characteristic of the quantum critical region. Quantum critical behaviour is seen for a range of temperatures between 50 K and 350 K.

Finally, for larger x we see that below about 200 K the relaxation rate decreases with temperature: this is a characteristic property of the quasi-classical particle regime. This last interpretation is also consistent with neutron-scattering measurements by the late Jean Rossat-Mignod of the Centre d'Etudes Nucleaires de Grenoble in France, Herb Mook of Oak Ridge, Bernhard Keimer of Princeton University, Philippe Bourges of the Laboratoire Léon Brillouin in Saclay, near Paris, and others. A sharp "resonance" peak believed to be an $S = 1$ particle excitation appears in the scattering cross-section at low temperatures.

The above concordance between theory and experiments suggests that we are close to a reasonable understanding of the spin fluctuations, at least at intermediate energy scales corresponding to temperatures in the range 100–1000 K. At even lower energy and temperatures scales, the coupling between the charge and spin dynamics cannot be neglected, and this is certainly important for the transition into the superconducting state. There is sure to be progress in this important field in the years ahead.

Quantum Hall systems

The study of electrons confined in a two-dimensional layer at the surface of a semiconductor has fascinated physicists for over 30 years. In addition to their fundamental importance, such two-dimensional electron gases have also been exploited in the high-electron-mobility transistors found in mobile phones and satellite dishes.

When a two-dimensional electron gas is placed in a very strong magnetic field, the electrical response displays the remarkable "quantum Hall effect" – the transverse resistivity is quantized at values determined by the fundamental constants of nature. The origin of this effect is ultimately due to the quantization of the electronic kinetic energy into discrete Landau levels by the external magnetic field. This quenching of the electron motion means that other degrees of freedom become important when determining the low-energy excitations of the system. In particular, a considerable amount of energy is needed to excite an electron from one Landau level to the next, so the low-energy magnetic interactions between the electron spins play a key role in the properties of the system. Although the electrons are not actually fixed on a lattice, their magnetic behaviour can be interpreted as if they were (figure 5).

The magnetic quantum phase transition occurs in so-called bilayer quantum Hall systems. In these systems a pair of two-dimensional electron gas layers, each with one Landau level exactly filled, are placed about 20 nm apart. Experiments by Vittorio Pellegrini, Aron Pinczuk and others at Lucent Technologies in the US, and by Anju Sawada and co-workers at Tohuku University in Japan, have probed the magnetic properties of such systems in laser-scattering experiments. They found, under certain conditions, a magnetically ordered phase with a macroscopic ferromagnetic moment within each layer. However, the moments of the layers pointed in different directions (figure 5*a*). This is rather similar to the antiferromagnetic phase found in high-temperature superconductors.

Under different conditions they observed a quantum paramagnet ground state, with particle-like $S = 1$ excitations (figure 5*b*). Again, this is reminiscent of the behaviour observed for small λ in the models of high-temperature superconductors.

This phase diagram can be understood by examining the preferred spin configurations of nearby electrons: two electrons within the same layer prefer to have their spins parallel to each other, while electrons in different layers prefer antiparallel spins. This leads to antiferromagnetic and quantum paramagnet phases observed in the experiments.

Obviously there must be a transition between these two phases, and detailed theoretical predictions have been made for the behaviour of the system near this critical point. The experimental system has a great deal of flexibility in tuning material parameters because, in principle, it is possible to vary the electron density, the layer spacing, the angle between the magnetic field and the electron layer, and other parameters at will. This promises a precise quantitative confrontation between theory and experiment in the near future.

Outlook

For over three decades, physicists have successfully understood the properties of a variety of solid-state materials using what is essentially an independent-electron picture, along with sophisticated variants. This understanding has been exploited in a large number of technological advances that influence our everyday lives. One important frontier in the study of new materials now lies in systems in which such a simple picture fails and strong correlations abound. The electron correlations in the magnetic insulators, high-temperature superconductors and quantum Hall systems described in this article are just a few examples.

A powerful theoretical approach to such systems is to identify the quantum critical points between various zero-temperature phases, and to use these critical points as a vantage point for exploring the rest of the phase diagram. It is clear that we have just scratched the surface of much exciting progress to come.

Further reading

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