Melting at the absolute zero of temperature:
Quantum phase transitions in condensed matter

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We dedicate this article to our father, Prof. Dr. Günter Vojta, on the occasion of his 80th birthday. While colleagues appreciate him as an excellent scientist with widespread interests and an amazingly broad knowledge, we see in him the father, teacher, friend, and much more. Thanks to him, we grew up immersed in the fascinating world of physics from an early age. His example and encouragement have crucially influenced our own lives.

I. INTRODUCTION

Phase transitions play an essential role in nature. Everyday transitions include the boiling of water or the melting of ice, while the transition of a metal into the superconducting state upon lowering the temperature provides a more complicated example. 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 occur upon variation of an external control parameter; their common characteristics is a qualitative change in the system properties. The phase transitions mentioned so far occur at finite temperature; here macroscopic order (e.g. the crystal structure in the case of melting) is destroyed by thermal fluctuations. During recent years, a different class of phase transitions, the so-called quantum phase transitions, has attracted the attention of physicists. These transitions constitute the subject of this article.

Quantum phase transitions (QPTs) [1, 2] occur, in a strict sense, at exactly zero temperature only. A non-thermal control parameter, such as pressure, magnetic field, or chemical composition, is varied to access the transition point. In contrast to phase transitions at finite temperature, quantum effects play a decisive role, as the fluctuations driving the transition follow quantum in-
stead of classical statistical mechanics. While QPTs were originally considered mere curiosities of theoretical physics, the impact of this field on modern research is now emerging with increasing pace.

Violent quantum fluctuations caused by Heisenberg’s uncertainty principle are at the heart of quantum critical points (QCPs) which control continuous QPTs. They affect the finite-temperature behavior of condensed matter as well, with a multiplicity of new and unexpected phenomena. For instance, the standard model of metals with electronic interactions, the Landau Fermi-liquid picture, may break down in the vicinity of a QPT. In addition, since QPTs occur between nearly degenerate phases whose characteristic energy scales are driven to zero, small perturbations may become important which otherwise would be masked by the primary energy scales. This has the fascinating prospect of inducing novel states of matter around QCPs.

II. EXAMPLE: TlCuCl₃ - A COUPLED DIMER MAGNET

To set the stage, we consider as a paradigmatic example the crystalline material TlCuCl₃ which displays several magnetic quantum phase transitions. TlCuCl₃ is a so-called Mott insulator, i.e., it is electrically insulating despite the presence of a partially filled conduction band. This insulating behavior arises from the strong Coulomb repulsion which tends to localize electrons in the 3d shells of copper. As a result, each copper atom in TlCuCl₃ carries one unpaired electron with a spin-1/2 magnetic moment – these moments are responsible for the variety of magnetic phenomena in TlCuCl₃.

Fig. 1 shows magnetic phase diagrams of TlCuCl₃ as function of pressure or applied magnetic field and temperature. While the material is a paramagnet at ambient pressure and low field, it can be driven – at the lowest temperatures – into antiferromagnetic phases either by pressure or field. Conversely, these antiferromagnetic states at $T = 0$ can be destroyed either by increasing temperature – leading to a conventional thermal phase transition – or by lowering pressure or field, thereby crossing a quantum phase transition point.

The underlying reason for this interesting behavior is the presence of pairs of spin-1/2 moments, dubbed dimers, which form a three-dimensional network, as shown in Fig. 2.

Before giving a more detailed description, we collect the basic concepts of phase transitions and critical behavior [6, 7] which are necessary for the later discussions.
FIG. 1: Magnetic phase diagrams of TlCuCl$_3$. a) Pressure–temperature phase diagram at zero magnetic field. The antiferromagnetic phase breaks the SU(2) spin symmetry of the underlying Heisenberg model and is bounded by a line of finite-temperature phase transitions. This line terminates in the quantum critical point (QCP), where antiferromagnetic order can be tuned by pressure at $T = 0$. (Data from Ref. [3].) b) Field–temperature phase diagram at ambient pressure. Here, the antiferromagnetic phase breaks a U(1) symmetry and can be interpreted as Bose-Einstein condensate of magnons. (Data from Ref. [4].)

III. CONCEPTS OF CLASSICAL AND QUANTUM PHASE TRANSITIONS

Phase transitions are traditionally classified into first-order and continuous transitions. At first-order transitions the two phases co-exist at the transition point, examples include ice and water at $0^\circ$ C, or water and steam at $100^\circ$ C. In contrast, at continuous transitions, also called critical points, the two phases do not co-exist, instead they become indistinguishable at the transition point. An important example is the ferromagnetic transition of iron at $770^\circ$ C, above which the magnetic moment vanishes. This phase transition occurs at a point where thermal fluctuations destroy the regular ordering of magnetic moments – it happens continuously in the sense that the magnetization vanishes continuously when approaching the transition from below.

In the following we concentrate on systems near a continuous phase transition. Such a transition can usually be characterized by an order parameter; this is a thermodynamic quantity that is zero in one phase (the disordered) and non-zero and generally non-unique in the other (the ordered) phase. Very often the choice of an order parameter for a particular transition is obvious as, e.g.,
FIG. 2: Schematic crystal structure of TlCuCl$_3$. It consists of planar dimers of Cu$_2$Cl$_6$ which are stacked on top of each other to form infinite double chains along the crystallographic a axis. Magnetic copper atoms are shown in red. (Reprinted with permission from Ref. [5]. Copyright 2002 by the American Physical Society.)

for the ferromagnetic transition where the total magnetization is an order parameter. However, in some cases finding an appropriate order parameter is complicated and still a matter of debate, e.g., for the interaction-driven metal–insulator transition in electronic systems (the Mott transition [8]).

While the thermodynamic average of the order parameter is zero in the disordered phase, its fluctuations are non-zero. When the critical point is approached, the spatial correlations of the order parameter fluctuations become long-ranged. Close to the critical point, their typical length scale, the correlation length $\xi$, diverges as

$$\xi \propto |r|^{-\nu}$$

(1)

where $\nu$ is the correlation length critical exponent and $r$ is some dimensionless measure of the distance from the critical point. If the transition occurs at a non-zero temperature $T_c$, it can be defined as $r = |T - T_c|/T_c$. In addition to the long-range correlations in space there are analogous long-range correlations of the order parameter fluctuations in time. The typical time scale for a decay of the fluctuations is the correlation (or equilibration) time $\tau_c$. As the critical point is approached the correlation time diverges as

$$\tau_c \propto \xi^z \propto |r|^{-\nu z}$$

(2)
where \( z \) is the dynamical critical exponent. Close to the critical point, there is no characteristic length scale other than \( \xi \) and no characteristic time scale other than \( \tau_c \). (Note that a microscopic cutoff scale must be present to explain non-trivial critical behavior, for details see, e.g., Goldenfeld [7]. In a solid such a scale is, e.g., the lattice spacing.)

The power-law singularities (1) and (2) are responsible for the so-called critical phenomena. At the phase transition point, correlation length and time are infinite, fluctuations occur on all length and time scales, and the system is said to be scale-invariant. As a consequence, all observables depend via power laws on the external parameters. The set of corresponding exponents – called critical exponents – completely characterizes the critical behavior near a particular phase transition.

One of the most remarkable features of continuous phase transitions is universality, i.e., the fact that the critical exponents are the same for entire classes of phase transitions which may occur in very different physical systems. These universality classes are determined only by the symmetries of the order parameter and by the space dimensionality of the system. This implies that the critical exponents of a phase transition occurring in nature can be determined exactly (at least in principle) by investigating any simple model system belonging to the same universality class. The mechanism behind universality is again the divergence of the correlation length. Close to the critical point the system effectively averages over large volumes rendering the microscopic details of the Hamiltonian unimportant.

IV. QUANTUM PHASE TRANSITIONS AND THE ROLE OF QUANTUM MECHANICS

The question of to what extent quantum mechanics is important for understanding a continuous phase transition has at least two aspects. On the one hand, quantum mechanics can be essential for understanding the ordered phase, (e.g., superconductivity) – this depends on the particular transition considered. On the other hand, one may ask whether quantum mechanics influences the asymptotic critical behavior. For this discussion we have to compare two energy scales, namely \( \hbar \omega_c \), which is the typical energy of order parameter fluctuations, and the thermal energy \( k_B T \). We have seen in the preceding section that the typical time scale \( \tau_c \) of the fluctuations diverges as a continuous transition is approached. Correspondingly, the typical frequency scale \( \omega_c \) goes to zero and with it the typical energy scale

\[
\hbar \omega_c \propto |r|^{\nu z}.
\] (3)
Quantum mechanics will be important as long as this typical energy scale is larger than the thermal
energy $k_B T$; on the other hand, for $\hbar \omega_c \ll k_B T$ a purely classical description can be applied to the
order parameter fluctuations. This implies that the character of the order parameter fluctuations
crosses over from quantum to classical when $\hbar \omega_c$ falls below $k_B T$.

Now, for any transition occurring at some finite temperature $T_c$, quantum mechanics will be-
come unimportant for $|r| \lesssim T_c^{1/\nu z}$, in other words, the critical behavior sufficiently close to the
transition is entirely classical. This justifies calling all finite-temperature phase transitions “clas-
sical”. Quantum mechanics can still be important on microscopic scales, but classical thermal
fluctuations dominate on the macroscopic scales that control the critical behavior. In contrast, if
the transition occurs at zero temperature as a function of a non-thermal parameter $r$, the order
parameter fluctuations always obey quantum statistical mechanics. Consequently, transitions at
zero temperature are called “quantum” phase transitions. The characteristic scale $\hbar \omega_c \propto r^{\nu z}$ often
represents the energy gap to excitations above the quantum mechanical ground state.

The interplay of classical and quantum fluctuations leads to an interesting phase diagram in the
vicinity of the quantum critical point. Two cases need to be distinguished, depending on whether
or not long-range order can exist at finite temperatures.

Fig. 3a describes the situation where order only exists at $T = 0$, this is the case, e.g., in two-
dimensional magnets with SU(2) symmetry where order at finite $T$ is forbidden by the Mermin-
Wagner theorem. In this case there will be no true phase transition in any experiment carried out at
finite temperature. However, the finite-$T$ behavior is characterized by three very different regimes,
separated by crossovers. For low $T$ and $r > 0$ thermal effects are negligible ($T \ll r^{\nu z}$), and the
critical singularity is cutoff by the deviation of the control parameter $r$ from criticality. This regime
is dubbed “quantum disordered” and characterized by well-defined quasiparticle excitations; for
a magnetic transition in a metallic system this will be the usual Fermi-liquid regime. For $r < 0$
and $T > 0$, we are in the “thermally disordered” regime; here the order is destroyed by thermal
fluctuations of the ordered state (yet quasiparticles are still well defined on intermediate scales).
A completely different regime is the high-temperature regime above the QCP. In this “quantum
critical” regime [9], bounded by crossover lines $T \sim |r|^{\nu z}$, the critical singularity is cutoff by
the finite temperature. The properties are determined by the unconventional excitation spectrum
of the quantum critical ground state, whose main characteristics is the absence of conventional
quasiparticle-like excitations, which are replaced by a critical continuum of excitations. In the
quantum critical regime, this continuum is thermally excited, resulting in unconventional power-
FIG. 3: Schematic phase diagrams in the vicinity of a quantum critical point (QCP). The horizontal axis represents the control parameter $r$ used to tune the system through the quantum phase transition, the vertical axis is the temperature $T$. a) Order is only present at zero temperature. The dashed lines indicate the boundaries of the quantum critical region where the leading critical singularities can be observed; these crossover lines are given by $k_B T \propto |r - r_c|^{\nu z}$. b) Order can also exist at finite temperature. The solid line marks the finite-temperature boundary between the ordered and disordered phases. Close to this line, the critical behavior is classical.

Law temperature dependencies of physical observables. Universal behavior is only observable in the vicinity of the quantum critical point, i.e., when the correlation length is much larger than microscopic length scales. Quantum critical behavior is thus cut off at high temperatures when $k_B T$ exceeds characteristic microscopic energy scales of the problem – in magnets this cutoff is, e.g., set by the typical exchange energy.

If order also exists at finite temperatures, Fig. 3b, the phase diagram is even richer. Here, a real phase transition is encountered upon variation of $r$ at low $T$; the quantum critical point can be viewed as the endpoint of a line of finite-temperature transitions. As discussed above, classical fluctuations will dominate in the vicinity of the finite-$T$ phase boundary, but this region becomes narrower with decreasing temperature, such that it might even be unobservable in a low-$T$ experiment. The fascinating quantum critical region is again at finite temperatures above the quantum critical point, and the thermally disordered regime is restricted to temperatures $T > T_c$.

It is instructive to discuss QPTs in terms of behavior of the low-energy many-particle states of the quantum system upon a change of $r$, starting with finite system size and then taking the thermodynamic limit. A first-order quantum phase transition corresponds to a ground-state level
crossing at $r = r_c$, which remains sharp in the thermodynamic limit. In contrast, a continuous quantum phase transition manifests itself in an avoided level crossing, which becomes sharper upon increasing the system size. Here, a large number of low-lying excited state approach the ground state, reflecting the fact that the characteristic scale $\omega_c$ vanishes at $r = r_c$. In the thermodynamic limit, the avoided crossing becomes sharp, but only the second derivative of the ground state energy $E(r)$ becomes singular.

V. BACK TO THE EXAMPLE: COUPLED DIMER MAGNETS

Let us return to coupled dimer magnets, with TlCuCl$_3$ being a prime example. The copper atoms carry spin 1/2, with mutual antiferromagnetic exchange interactions of Heisenberg type. The monoclinic crystal structure contains two copper atoms per unit cell, hence the spins are naturally paired into dimers, and the structure can be understood as three-dimensional arrangement of coupled dimers.

A minimal microscopic model for the relevant magnetic degrees of freedom is an antiferromagnetic quantum Heisenberg with spatially modulated coupling, with the Hamiltonian

$$H = \sum_{\langle ij \rangle} J_{ij} \vec{S}_i \cdot \vec{S}_j - \vec{h} \cdot \sum_i \vec{S}_i .$$  \hspace{1cm} (4)

Here $\vec{S}_i$ is a vector spin operator for the copper spin at lattice site $i$, $\vec{h}$ is an external magnetic field. The couplings $J_{ij}$ obey

$$J_{ij} = \begin{cases} J & \text{intra – dimer} \\ J' & \text{nearest inter – dimer} \end{cases};$$  \hspace{1cm} (5)

for simplicity we have reduced the inter-dimer couplings to a single coupling constant.

In the absence of a field, the ground state is determined by the ratio $J'/J$. For $J' \ll J$, quantum mechanical singlets are formed on each dimer, and the ground state is a quantum paramagnet. Its elementary magnetic excitations are so-called triplons, i.e. dispersive spin-1 excitations with a minimum excitation energy $\Delta$. In the limit of $J' \rightarrow 0$ the ground state is simply given by a product state of dimer singlets, and the gap $\Delta = J$. If, in contrast, $J' \sim J$, the system is a three-dimensional antiferromagnet displaying magnetic long-range order. (We are assuming that the magnetic frustration inherent in the lattice structure is small.) Thus, tuning the ratio $J'/J$ will lead to a magnetic order–disorder transition in the ground state. This is exactly what happens upon applying pressure to TlCuCl$_3$, Fig. 1a. Increasing the pressure $p$ changes the lattice constants such
that the ratio $J'/J$ increases, and the system is driven into an antiferromagnetic state, accompanied by a spontaneous breaking of the SU(2) symmetry of the Heisenberg model. The gap of the paramagnet vanishes upon approaching the critical pressure $p_c$ as $\Delta \propto (p_c - p)^{\nu z}$ with mean-field critical exponents $\nu = 1/2$ and $z = 1$ (with logarithmic corrections).

Applying a field at ambient pressure does not affect the paramagnetic ground state, but causes a Zeeman splitting of the spin-1 triplon excitations. At a critical field $H_{c1}$, the gap of the lowest triplon branch closes, and the system is again driven into an ordered state, Fig. 1b. This “canted” state has a finite uniform magnetization in the direction of the applied field and a spontaneous antiferromagnetic order perpendicular to the field direction. Further increasing the field leads to a second transition at $H_{c2}$ where the system enters a fully polarized state. (In TlCuCl$_3$, $H_{c2}$ is expected to be around 90 T which has not been reached experimentally.)

We emphasize that the physics of the field-driven case is qualitatively different from the pressure-driven situation. In a field, the spin symmetry of the Hamiltonian is reduced from SU(2) to U(1), and the ordered phase breaks this U(1) symmetry. The lowest triplon branch of the paramagnet can be interpreted as a bosonic excitation on top of the singlet ground state. In the field-ordered phase, the ground state can be written as a superposition of singlet and triplet – a “triplon condensate” – with a complex ratio between singlet and triplet component, determining the orientation of the staggered magnetization perpendicular to the field. Upon raising the temperature, the antiferromagnetic order is destroyed at $T_N$, but the density of triplons turns out to change in a non-singular fashion across $T_N$. Remarkably, the finite-temperature transition at $T_N$ can be understood as Bose-Einstein condensation of triplons (or magnons): These are pre-existing bosons which acquire phase coherence below $T_N$. The phase of the Bose condensate is observable as orientation of the staggered magnetization. In contrast, the triplon density changes singularly upon crossing the quantum phase transition at $H_{c1}$: The density is simply zero below $H_{c1}$ and finite above. Technically, this QPT is in the universality class of the dilute Bose gas.

VI. CONCLUSIONS AND OUTLOOK

This article has illustrated aspects of zero-temperature phase transitions in quantum systems. Conceptually, quantum phase transitions open a field of fascinating physics, as they are connected to the peculiar properties of the quantum critical ground state. Quantum criticality also provides new perspectives in the study of correlated systems, where intermediate-coupling phenomena are
hardly accessible by standard weak- or strong-coupling perturbative approaches. A promising route starts by identifying quantum critical points between stable phases, and then uses these as vantage points for exploring the phase diagram by expanding in the deviation from criticality.

While thermodynamic properties of magnetic quantum phase transitions in insulators, as in our example, are relatively well understood (this also applies to phase transitions with other conventional order parameters), there is a variety of challenges in current research: (i) Quantum phase transitions in metals, often accompanied by non-Fermi liquid behavior, are less understood, due to the coupling between order parameter fluctuations and particle-hole excitations of the metal. (ii) Transitions without a local order parameter are known to exist, but they are a hard task for theory, with examples being the Mott transition and various types of topological transitions. (iii) The influence on quantum phase transitions of quenched disorder, which is unavoidable in solids, is very strong. Even simple models lead to spectacular effects, but little is known for more realistic types of disorder. (iv) Transport, and more generally, non-equilibrium properties, near quantum phase transitions are an exciting and difficult venue of research. Here, beautiful experiments with ultracold atomic gases have triggered a lot of interest. It is clear that we have just scratched the surface of this field, and much exciting progress is expected in the future.

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