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Bose–Einstein condensation in magnetic materials

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Abstract

Bose–Einstein condensation denotes the formation of a collective quantum ground state of identical particles with integer spin or intrinsic angular momentum. In magnetic insulators, the magnetic properties are due to the unpaired shell electrons that have half-integer spin. However, in some magnetic compounds (e.g., $TlCuCl_3$) two Cu^{2+} ions are antiferromagnetically coupled to form a spin singlet with total spin $S=0$, separated by an energy gap from the excited triplet states with total spin $S=1$. In such dimer compounds, Bose–Einstein condensation becomes possible when the energy of one of the triplet components intersects the ground-state singlet, resulting in long-range magnetic order; this transition represents a quantum critical point at which Bose–Einstein condensation occurs. Here we summarize recent neutron scattering investigations of the excitation spectrum in $TlCuCl_3$, for which the quantum critical point was realized by the application of both an external magnetic field and hydrostatic pressure. The theoretically predicted gapless Goldstone mode characteristic of the Bose–Einstein condensation of the triplet states was verified.

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1. Introduction

The basic idea of Bose–Einstein condensation (BEC) dates back to 1925 when Einstein [1], on the basis of a paper by Bose [2], devoted to the statistical description of the quanta of light, predicted the occurrence of a phase transition in a gas of noninteracting atoms. This phase transition is associated with the condensation of atoms in the state of lowest energy and is the consequence of quantum statistical effects. Little notice was taken of this curious possibility until 1938 when London [3], immediately after the discovery of superfluidity in liquid helium [4,5], had the intuition that superfluidity could be a manifestation of BEC. When helium is cooled to a critical temperature of 2.17 K, a remarkable discontinuity in heat capacity occurs, the liquid density drops, and a fraction of the liquid becomes a zero viscosity superfluid. A condensation effect is also credited with producing superconductivity. In the Bardeen–Cooper–Schrieffer theory [6], pairs of electrons are coupled by lattice interactions, and the pairs (called Cooper pairs) act like bosons and can condense into a state of zero electrical resistance. In copper-oxide high-temperature superconductors the Cooper pairs are formed by holes. The case of excitons, i.e., quasi-particles, as candidates for BEC below a critical temperature has also been postulated based on the observation of a sharp emission line which has been attributed to the presence of a laser-induced Bose–Einstein condensate of excitonic molecules in CuCl [7]. More recently, it was discovered that even atoms can be forced to undergo BEC and thereby to form a novel state of matter. A magnetically trapped, spin polarised gas of rubidium [8] and sodium [9] atoms was cooled down to nanokelvin temperatures where a fraction of the individual atoms condenses into a "superatom" behaving as a single entity. BEC has also been widely applied to fundamental problems in nuclear and elementary particle physics (see e.g. Ref. [10]).

BEC was first described for an ideal gas of free bosons with mass m and density N. BEC occurs below a critical

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temperature T_c where the chemical potential μ vanishes. The chemical potential μ is obtained from the conditions:

$$n_k = \left[\exp\{\beta(\varepsilon_k - \mu)\} - 1\right]^{-1}, \quad \sum_k n_k = N, \tag{1}$$

where $\varepsilon_k = \hbar^2 k^2/2m$ denotes the spectrum of the noninteracting particles. In $d>2$ dimensions, the critical temperature T_c results from the condition

$$\int_0^\infty d\varepsilon g(\varepsilon)\left[\exp\{\beta_c \varepsilon\} - 1\right]^{-1} = N, \tag{2}$$

where $g(\varepsilon)$ is the single-particle density-of-states. Below T_c a macroscopic number of particles accumulates in the lowest state, the BEC state. The integral in Eq. (2) diverges for $d \leqslant 2$, and BEC does not occur (i.e., $T_c = 0$). In reality we usually deal with a gas of interacting particles. The interaction leads to a depletion of the condensate ($N_0 < N$ at $T = 0$), i.e., the condensate fraction depends on the interaction energy [10].

Up to the present, little attention has been paid to BEC in magnetic materials, because the magnetic properties are due to the unpaired electrons which are fermions constrained by the Pauli exclusion principle, but only integer spin bosons (governed by Bose–Einstein statistics) can condense in unlimited numbers into a single ground state. Nevertheless, in the past few years new materials were synthesized in which the magnetic ions are pairwise arranged to form a crystalline network of dimers. Of particular interest are the compounds $BaCuSi_2O_6$ [11,12], $Sr_2Cu(BO_3)_2$ [13] and $ACuCl_3$ (A = K, Tl, NH_4) [14–17] in which the two Cu^{2+} ions forming the dimer are antiferromagnetically coupled according to the spin Hamiltonian $H = -J\mathbf{S_1}\cdot\mathbf{S_2}$. The dimer ground state is then a singlet

$$|s\rangle = \left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle\right)/\sqrt{2} \tag{3a}$$

with total spin $S = 0$, separated by an energy gap $\Delta = -J$ ($J<0$) from the excited triplet states with total spin $S = 1$. The energy gap Δ can be modified by external magnetic fields. For instance, a field H_z splits the excited triplet according to the Zeeman energy $g\mu_B H_z$ into three components with $S_z = +1, 0, -1$ corresponding to

$$\begin{aligned} |t_{+1}\rangle &= |\uparrow\uparrow\rangle, \\ |t_0\rangle &= \left(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle\right)/\sqrt{2}, \\ |t_{-1}\rangle &= |\downarrow\downarrow\rangle. \end{aligned} \tag{3b}$$

At a critical magnetic field H_c the energy of the triplet component $|t_{+1}\rangle$ intersects the ground-state singlet, i.e., the chemical potential $\mu = J - g\mu_B H_c$ vanishes, thus H_c is a quantum critical point separating a gapped spin-liquid state ($H<H_c$) from a field-induced magnetically ordered state ($H>H_c$). The triplet components $|t_{+1}\rangle$ can be regarded as bosons with hard-core repulsion, thus BEC occurs in these dimer compounds at the quantum critical point H_c, i.e., the gas of triplet bosons undergoes a phase transition into a novel condensate state with macroscopic occupation of the single-particle ground-state which can be described by the coherent superposition

$$|\psi\rangle = a_s|s\rangle + a_t e^{i\Phi}|t_{+1}\rangle, \tag{4}$$

where $a_s \approx 1$ and $a_t << 1$ are the singlet and triplet amplitudes, respectively, and Φ denotes the phase factor. a_t and Φ are determined by the spin expectation values of the two Cu^{2+} ions of the dimer which in the limit $a_s \approx 1$ are given by

$$\begin{aligned} \langle S_{x1}\rangle &= -\langle S_{x2}\rangle \propto a_t \cos\Phi, \\ \langle S_{y1}\rangle &= -\langle S_{y2}\rangle \propto a_t \sin\Phi, \\ \langle S_{z1}\rangle &= \langle S_{z2}\rangle \propto a_t^2. \end{aligned} \tag{5}$$

What is the experimental proof that a system is Bose–Einstein condensed? The relevant parameter is certainly the occupation number N_0 of the particles in the BEC state which is proportional to the magnetization along the field direction, i.e., $N_0 \propto a_t^2$. The critical exponents of the ordered phase provide further evidence. An alternative is offered by the properties of the magnetic excitation spectrum in the BEC state which has been theoretically predicted to be a gapless Goldstone mode associated with the breaking of rotational symmetry by the staggered magnetic order, thus the presence of a spin-wave-like mode with a linear dispersion is a convincing signal for the existence of the Bose–Einstein condensate [18]. This theoretical prediction was observed for the first time by inelastic neutron scattering in the dimer spin compound $TlCuCl_3$ [19].

The singlet-triplet gap of $TlCuCl_3$ can also be closed by the application of hydrostatic pressure which occurs at a critical pressure $p_c \approx 1$ kbar [20]. In contrast to the field-induced case, all the triplet components can condense into the singlet ground-state at the quantum critical point p_c. The magnetic excitation spectrum has again the nature of a gapless Goldstone mode [21] which was recently observed in the pressure-induced ordered phase [22].

The present work summarizes the results obtained for $TlCuCl_3$, and some implications to other magnetic materials are discussed.

2. The dimer spin compound $TlCuCl_3$

Monoclinic $TlCuCl_3$ crystallizes in the space group $P2_1/c$ with four formula units per unit cell [23]. The magnetic properties are determined by the $S = \frac{1}{2}$ moments of the Cu^{2+} ions, which are arranged in centrosymmetric pairs. Static measurements reveal that the ground state is a nonmagnetic singlet separated from the excited triplet by the energy gap Δ which is estimated from high-field magnetization data as $\Delta/k_B \approx 7.5$ K [24]. The spin interactions are almost isotropic, with coinciding static behavior when scaled by the gyromagnetic factor g [25].

Comprehensive inelastic neutron scattering measurements were performed to establish the magnetic excitations corresponding to the singlet–triplet transition as shown in