Quantum behaviour in an ancient Chinese pigment

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The physical behaviour of all matter in the universe is determined by the statistics of its constituent particles. Each microscopic particle can be one of two types, 'fermionic' or 'bosonic': particles with half-integer spins are fermions, while particles with integer-spins are bosons. At very low temperatures where quantum properties of matter are revealed, the radical difference in the underlying nature of these two families of particles becomes apparent. Fermions are easily accessible for experimentation, and their quantum properties well understood - for example, the ubiquitous electron, which is a fermion determines the physics of each element in the periodic table. However, much less is understood about the quantum behaviour of bosons, with very few accessible experimental systems available. My research involves the study of magnetic compounds in which the pairing of half-integer spins leads to bosonic particles, making these experimentally accessible model systems to investigate the low temperature quantum behaviour of bosons.

Quantum mechanics dictates that only one fermion can occupy each quantum state, whereas an unlimited number of bosons can share the same quantum state. As absolute zero temperature (-273°C $\equiv$ 0 K) is approached, bosons all condense into the lowest available quantum state, in which the individual particles lose their identities, and become components of one giant collective entity known as a 'Bose Einstein condensate' (BEC). This entire body moves as a collective whole without any interparticle collisions, leading to the spectacular phenomenon of superfluidity, which is motion characterized by a complete lack of viscosity (i.e. the ability to flow around obstacles with absolutely no dissipation.) The first BEC observed was liquid ^{4}He cooled to 2K in 1938 [1]. Creating a BEC is so experimentally challenging, that the next experiment to successfully create a BEC was not until the Nobel Prize winning effort in 1995, in which a gas of Rb atoms was laser cooled close to absolute zero temperature [2]. In the hunt for a new BEC, we exploit the fact that collective forms of particle behaviour such as superfluidity are insensitive to the material-specific properties of individual particles (i.e.) they are 'emergent' properties that emerge from a large collection of particles behaving as a whole.

The experimental system which I have studied as part of my research is a transparent purple copper oxide material - $BaCuSi_2O_6$. This material is by no means a new invention and quite to the contrary, has been known to man since 2000 years ago. The first use of $BaCuSi_2O_6$ was as a purple dye made into rods for paint production, used as pigment layers in the Terracotta army, or applied to stained glass during either the Warring States period (479 - 221BC) or the Q'in and Han dynasties (221BC - 220AD) in China [3, 4]. Indeed, it remains an archaeological curiosity as to how the ancient Chinese possessed the technological sophistication to prepare this vivid purple pigment (known as Han purple) over 2000 years ago. $BaCuSi_2O_6$ is a layered compound comprising planes of spin (s) = $\frac{1}{2}$ Copper ions. Spins on adjacent layers form pairs, leading to coupled layers of 'spin dimers'. A necessary feature to create bosons in a spin-dimer compound is the application of a high external magnetic field: $s = \frac{1}{2}$ particles can point either up or down, and in order to form an $s = 1$ pair (i.e. a particle with integer spin that obeys Boson statistics), both $s = \frac{1}{2}$ members must point in the same direction. This is achieved by a very strong external magnetic field of the order of 30 Tesla (800,000 times the size of the Earth's magnetic field!). We then proceed to experimentally look for BEC in this compound by cooling it to very low temperatures in a high applied external magnetic field.

The observation of a BEC signature in a magnetic system is not as simple as measuring the same quantities one would in an atomic BEC. For instance, in a ^{4}He superfluid, atoms of ^{4}He flow without friction. What it would look like for magnetic spins to flow 'without friction' is not as yet a question with a well defined answer! Instead, we exploit the physics of 'phase transitions' for an experimental signature. When matter transforms from one 'collective' state to another: such as ice melting into water, or spins aligning themselves into a ferromagnet, characteristics of the phase transition from one state to another are independent of specific systems, and depend only on the collective behaviour in each ‘phase’. In systems driven by their quantum properties, the relevant phase transitions are at zero temperature (where quantum effects dominate), known as quantum phase transitions (QPTs) [5]. The point at which a QPT occurs is known as a quantum critical point (QCP). Hence the QCP at which bosons condense into a BEC (shown in Fig. 1) has characteristic behaviour, irrespective of the specific properties of the constituent particles. I have experimentally studied the properties of a BEC QCP in the magnetic system $BaCuSi_2O_6$ as part of my research.

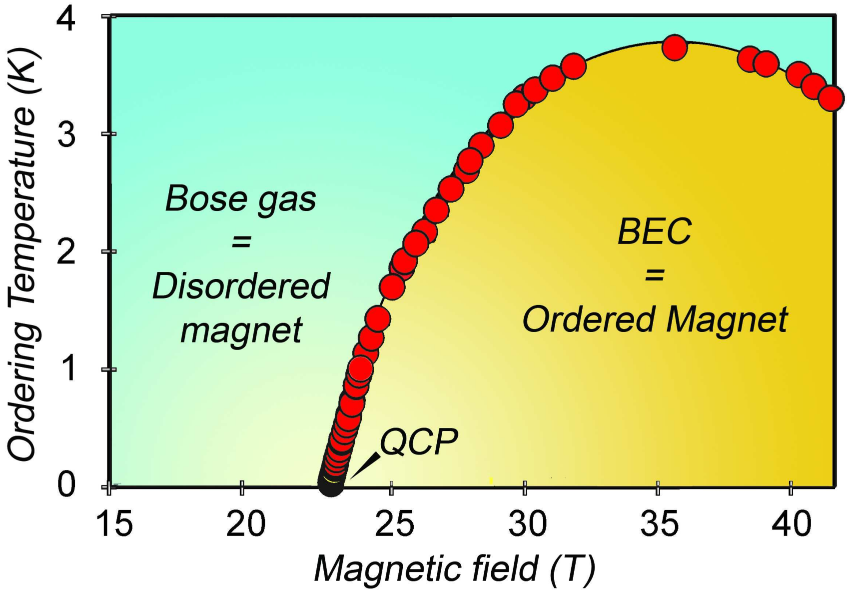

Figure 1. Quantum properties drive collective behaviour in the vicinity of the QCP (lighter shading). The collective state of spins in a magnetically ordered system is analogous to condensed bosons in a BEC. The shape of the phase boundary determines the characteristic power law describing collective behaviour. The solid circles represent results of my experiments on $BaCuSi_2O_6$

As the conglomerate of bosons (known as a Bose gas) is cooled, the particles condense into a BEC at a characteristic ‘ordering’ temperature. The ordering temperature is related to the total number of bosons raised to a characteristic power ν. It is this characteristic power law that is ‘universal’ for all BECs irrespective of the nature of the individual bosons, with a theoretically predicted value of $\frac{2}{3}$ for a BEC in three dimensions, and a theoretically predicted value of 1 for a BEC in two dimensions [6, 7].

In a magnetic system, the number of bosons correspond to the number of spins aligned in the same direction. The alignment of spins is controlled by the strength of the external magnetic field, and hence the total number of bosons can be experimentally tuned by using the external magnetic field as a control mechanism. Further, the alignment of spins determines the extent of ‘magnetisation’ of the system, which is an experimentally measurable quantity. Hence the exact number of condensed bosons is determined by experimentally measuring the magnetisation [8]. The tuneability of the number of bosons is a significant experimental advantage of the magnetic spin system. Conventional atomic BECs cannot be tuned in such a fashion, and hence there is no experimental evidence in any BEC that measures this theoretically predicted characteristic power law (the power law has been studied in experiments conducted on differing fractions of ^{4}He adsorbed into a spongy glass [9]. However, in this case, the effects due to the glass background cannot be neglected, leading to additional complications.)

My research involved experiments on single crystals of $BaCuSi_2O_6$ which I grew out of a solvent using the flux growth technique [10]. High quality single crystals are crucial for extracting critical exponents, since impurities will lead to effects such as disorder, preventing approach to the QCP. Experiments were performed at low temperatures down to 0.03K (where quantum behaviour is most apparent) in a large external magnetic field of up to 36Tesla. These extreme conditions are available at the National High Magnetic Field Laboratories at Tallahassee and Los Alamos. I performed magnetic torque, magnetocaloric effect, and specific heat experiments at different temperatures and applied magnetic fields. Each of these measured quantities exhibits a sharp feature at the ordering transition. Thus the ordering temperature is obtained at different magnetic fields (particle concentrations) and a phase boundary mapped out separating the Bose gas from a BEC (experimentally obtained results shown in Fig. 1) [11]. The shape of this phase boundary near the QCP measures the characteristic power law. Results of my experiments measure a characteristic power law of $\frac{2}{3}$ (corresponding to a three dimensional BEC) at temperatures down to 0.5K, which abruptly rises to a power law of 1 (corresponding to a two dimensional BEC) at temperatures down to 0.03K (shown in Fig. 2). The result is novel for more than one reason. While it has been theoretically well-established that there is a direct analogy between magnetic spin-dimer systems and BECs, this is the first experimental result that measures a characteristic power law equal to the theoretically predicted value for a BEC [10]. However, the more surprising experimental finding is the crossover from three dimensional to two dimensional BEC behaviour at low temperatures [11]!

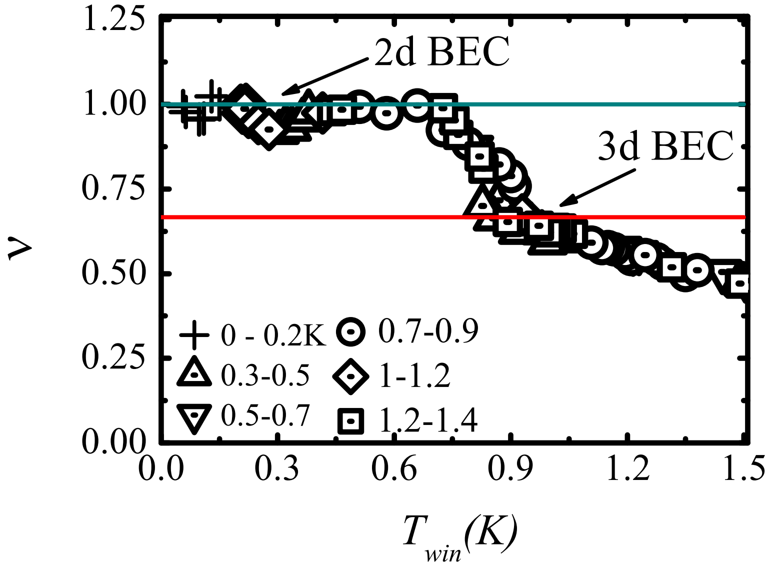

Figure 2. Values of the critical exponent ν obtained from fitting experimental points on the phase boundary in a sliding windown centred at T_{win} (K). The window-size varies from 0.05 to 1.4 K, as indicated by different symbols. Solid horizontal lines show the theoretical values of $\nu = \frac{2}{3}$ and 1 for 2d- and 3d-BEC respectively. The data approaches $\nu = \frac{2}{3}$ in the intermediate regime, and there is a distinct crossover toward $\nu = 1$ before the QCP is reached.

On analysing the magnetic structure of BaCuSi$_2$O$_6$, we find an explanation for this unexpected dimensional reduction. In most systems, magnetic interactions between the coupled layers of spins lead to three-dimensionality. However, in $BaCuSi_2O_6$, we find a situation where different sets of magnetic interactions between layers compete with each other, resulting in their net cancellation (shown in Fig. 3). This net cancellation is complete only exactly at the QCP (i.e. at zero temperature), with an unobservably small net interlayer coupling present near the QCP, which becomes larger as increasing temperature and magnetic field tune the system further away from the QCP, until finally the system becomes observably three-dimensional. This truly unique result reflects the special nature of collective quantum behaviour. We have seen how BEC is a collective quantum phenomenon or 'emergent' behaviour observed only near zero temperature. The experimental observation from $BaCuSi_2O_6$ provides the first example of dimensionality itself being an 'emergent' phenomenon (i.e.) away from the QCP, the individual particles belong to a three dimensional system. However, when the particles collectively condense into a BEC at the QCP, the BEC behaves as a two dimensional entity!

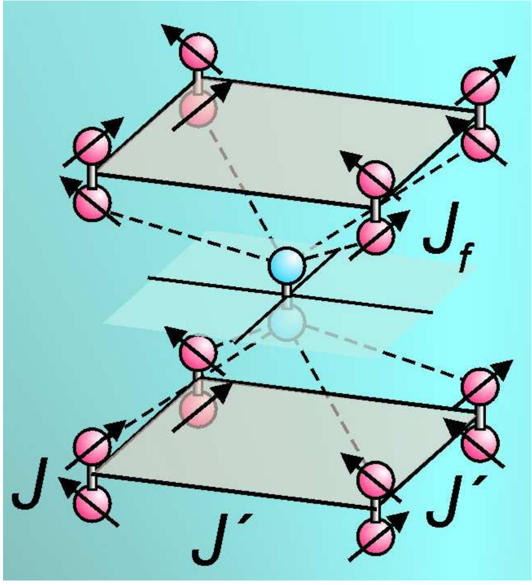

Figure 3. Geometrically frustrated body-centred tetragonal $BaCuSi_2O_6$ structure [12]. All interactions between spins require antiparallel alignment of the spins. As an example, the spins comprising the dimer in the middle layer are considered: each spin cannot simultaneously align in antiparallel fashion with all four of its nearest neighbour sites on the next vertical layer. Hence, there is no resultant interaction between vertically separated layers. Such a configuration with conflicting interactions is termed geometrically frustrated, and results in decoupling of layers in the vertical direction.

The implications of this finding extend beyond magnetic or BEC systems. Purely two dimensional quantum behaviour has only been a theoretical construct thus far, and has never been experimentally observed in a three dimensional material. These experimental results on $BaCuSi_2O_6$ demonstrate that it is a new model system to study not only BEC behaviour, but an entire gamut of two dimensional quantum effects as well. The two dimensional quantum behaviour observed in this spin system may provide us with a tool to better understand the origin of the mysterious phenomenon of 'high temperature superconductivity'(the dissipationless flow of current, known as superconductivity - which has been found to occur at temperatures as high as 150K) [13], that has eluded explanation by the physics community for decades. Several theories (for example the proposed theory of 'locally critical quantum critical behaviour') have related the mechanism of electron pairing in high temperature superconductors to collective two dimensional quantum behaviour [14, 15]. While the $BaCuSi_2O_6$ system is not a current carrier that would enable us to observe superconductivity, promising future experiments include introducing electrons which carry current into the system, and looking for novel collective behaviour such as superconductivity near the QCP.

A further analogy can be drawn between collective behaviour in the $BaCuSi_2O_6$ system and its superconducting analogue (where charge flows without dissipation), or its superfluid analogue (where atoms flow without dissipation). In $BaCuSi_2O_6$, the equivalent quantity is a flow of spins. While this 'spin current' is expected to dissipate in a finite time interval, it is capable of transporting far more information than a conventional 'charge current', and may prove extremely useful in future applications such as quantum computation [16]. Promising experiments include probing the dynamics of the 'spin current' in $BaCuSi_2O_6$.

References and Notes

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