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# Bose-Einstein condensation in $BaCuSi_2O_6$

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**Abstract.** $BaCuSi_2O_6$ is a model spin dimer system in which a BEC QCP is realised. Universal BEC power law scaling is experimentally observed, with $3d$ critical behaviour above 0.5 K, but a crossover to $2d$ BEC critical scaling down to 30 mK. Here we briefly review and expand on the results presented in the recent Nature paper [8].

## 1. Spin dimer systems

Spin dimer materials have attracted considerable attention recently due to the realisation of Bose-Einstein condensate (BEC) physics in these systems [1, 2, 3, 4, 5, 6, 7, 8, 9]. The ground state of these systems comprises singlets with gapped triplet excitations. In an applied magnetic field, Zeeman splitting of the triplet levels lowers the energy of the lowest triplet, such that above critical field $H_{c1}$, a finite concentration of triplets is populated in the ground state. The ground state above $H_{c1}$ is an ordered $XY$ antiferromagnet, separated from the disordered quantum paramagnet below $H_{c1}$ by a BEC quantum critical point (QCP) at 0 K. The applied magnetic field plays the role of the chemical potential in tuning the system from disorder to order, enabling experimental access to the vicinity of the QCP.

### *1.1. $BaCuSi_2O_6$: the model spin dimer system for Bose Einstein condensation*

$BaCuSi_2O_6$ is a particularly elegant realisation of a spin dimer system. A well separated hierarchy of exchange couplings places the system in the 'strong coupling' limit, such that the ordering phase boundary extends as high as $\sim$ 3.6K in temperature [1, 2]. The significant extent of the ordering phase boundary means that a large window of low temperatures exhibits universal behaviour driven by the QCP at absolute zero, making the system ideal for experimentation. In addition, the relatively high crystal symmetry of $BaCuSi_2O_6$ (tetragonal with a weak orthorhombic distortion [10]) means that any anisotropic terms that would break the rotational symmetry occur at a very low energy scale. The energy scale over which BEC power law scaling can be observed, therefore is well separated from any rotational symmetry-breaking terms that may exist [11].

The extended universal region of the phase boundary which can be experimentally accessed very close to the QCP facilitates the measurement of two universal BEC regimes in $BaCuSi_2O_6$.

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The critical exponent belongs to the $3d$ BEC universality class down to $\approx$ 0.5 K, but the mechanism of geometrical frustration results in layer decoupling at lower temperatures. Down to $\approx 30$ mK, the critical exponent belongs to the $2d$ BEC universality class, corresponding to the $2d$ BEC QCP in $BaCuSi_2O_6$. *The system thus exhibits the novel phenomenon of dimensional reduction at a QCP, which is of fundamental importance to our understanding of emergent quantum phenomenon in real materials.*

## 2. Ordering phase boundary in $BaCuSi_2O_6$

The crystal structure of $BaCuSi_2O_6$ consists of vertical $Cu^{2+}$ dimers arranged on a square bilayered lattice staggered between bilayers [12]. The intra-dimer ($J$), inter-dimer ($J'$) and inter-bilayer ($J''$) exchange couplings have a distinct hierarchy $J'' \ll J' \ll J$. Representing each dimer by a pseudospin with two possible states (lowest energy states $s^z = 0$ singlet and $s^z = 1$ triplet), and using appropriate transformations, the effective Hamiltonian describes a $3d$ gas of hardcore bosons with in-plane nearest neighbor hopping and repulsive interactions [1]. At low temperatures, the system undergoes a second order phase transition to an ordered state at a critical value of applied magnetic field $H_{c1} \approx 23.5T$. The magnetic phase transition in $BaCuSi_2O_6$ was experimentally measured on flux grown single crystals [11] by a series of torque, heat capacity, magnetisation, and magnetocaloric effect experiments at low temperatures and high magnetic fields ($H > H_{c1}$) to map out the ordering phase boundary [1, 2, 8]. Features in these thermodynamic quantities characterise the classical $3d$ XY phase transition into the ordered state at finite temperatures.

### *2.1. $3d$ Bose gas description*

The experimentally measured phase boundary of ordering transitions separating the ordered magnetic phase from the disordered phase at finite temperatures is shown in Fig. 1 down to $\approx 0.6$ K. The data is fit well by the Monte Carlo simulation of an equivalent $3d$ lattice of hard core bosons, considering values of exchange interaction $J = 4.45$meV, $J' = 0.51$ meV, $J'' = 0.168$ meV (Fig. 1). The phase boundary can therefore be interpreted as the ordering transition of a $3d$ interacting Bose gas into an ordered state, which is a Bose Einstein condensate. In other words, the system magnetically orders as an XY antiferromagnet, which may equivalently be interpreted as a BEC of triplets.

## 3. Critical behaviour describing BEC QCP

The $BaCuSi_2O_6$ spin dimer system enables unique field-tuned experimental access to the BEC QCP separating a quantum paramagnet from a BEC of triplets. The magnetic field acts as a chemical potential, which provides a convenient means of changing the particle number, thereby tuning the BEC to criticality at the QCP. Critical exponents for the $d \geq 2$ BEC universality class are mean field, and the power law relating the ordering temperature ($T_c$) to the proximity to the critical magnetic field is given by [13, 14, 15]

$$T_c \propto (H - H_{c1})^{2/d} \tag{1}$$

Figures 2c shows the entire phase boundary with additional measurements of magnetic torque in a dilution refrigerator down to $\approx 30$ mK. Since data are available in close proximity to the QCP, a two-parameter fit is performed with $\nu$, $H_{c1}$ varying. A power law is fit to data in sliding windows of different width (as indicated) centred at $T_{win}$. Figure 2a,b show the evolution in fitted $\nu$ and $H_{c1}$ values as the QCP is approached. As the temperature is lowered, the system enters the region of universal behaviour. The value of $\nu$ tends to $2/3$ in this region as $T$ tends to $\approx 0.5$ K from above ($T_{win} \approx 0.9$ K), characteristic of the $3d$-BEC universality class. However, below these temperatures there is an unexpected crossover to a value of $\nu = 1$ for the fit range

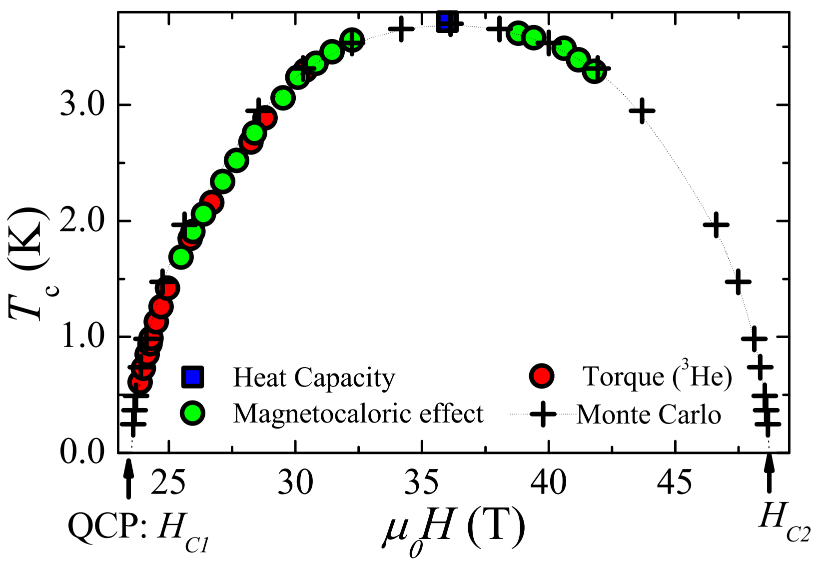


**Figure 1.** $BaCuSi_2O_6$: Experimentally determined phase boundary down to $\approx 0.6$ K. Points on the phase boundary are determined from magnetic torque (red circles), magnetocaloric effect (green circles), specific heat (blue square) measurements and Monte Carlo simulation (+ symbols, dotted line is a guide to the eye).

$T < 1$ K ($T_{\rm win} < 0.65$ K). From Eqn. 1, it is clear that the value of $\nu$ is characteristic of the $2d$-BEC universality class [8].

## 4. Dimensional reduction at the QCP due to geometrical frustration

We show that this experimental observation of dimensionally reduced $2d$-quantum criticality in the $3d$ spin system $BaCuSi_2O_6$ is by the mechanism of geometrical frustration. The $CuO_4$ square planes in $BaCuSi_2O_6$ are staggered in the vertical direction, such that a dimer in a single layer is at the centre of the square configuration of dimers in the next layer, but vertically offset [8]. Thus a situation arises where the spins on each dimer cannot simultaneously align in antiparallel fashion with all nearest neighbouring spins on the next vertical layer (i.e. the system configuration is geometrically frustrated.) Approaching the problem of geometrical frustration from the single particle limit, the single particle dispersion relation is given by

$$\epsilon(\mathbf{k}) = J + J'[\cos k_x + \cos k_y] + 2J_f[\cos\frac{k_x}{2}\cos\frac{k_y}{2}\cos k_z]. \quad (2)$$

The intra-layer and inter-layer components of the dispersion are shown in Fig. 3a. $\epsilon(\mathbf{k})$ is minimised by $\mathbf{q} = (\pi, \pi, k_z)$ and boson condensation occurs with momenta around this minimum. The form of the dispersion is quadratic at the quantum phase transition, which also has the consequence that the velocity of triplon excitations at the QCP tends to zero. Expanding in deviations from the minimum in momentum $\mathbf{k} = (\pi + \delta k_x, \pi + \delta k_y, k_z)$, the energy takes the form

$$\epsilon(\mathbf{q} + \delta\mathbf{k}) = J - J'[\cos\delta k_x + \cos\delta k_x] + 2J_f[\sin\frac{\delta k_x}{2}\sin\frac{\delta k_y}{2}\cos k_z]. \quad (3)$$

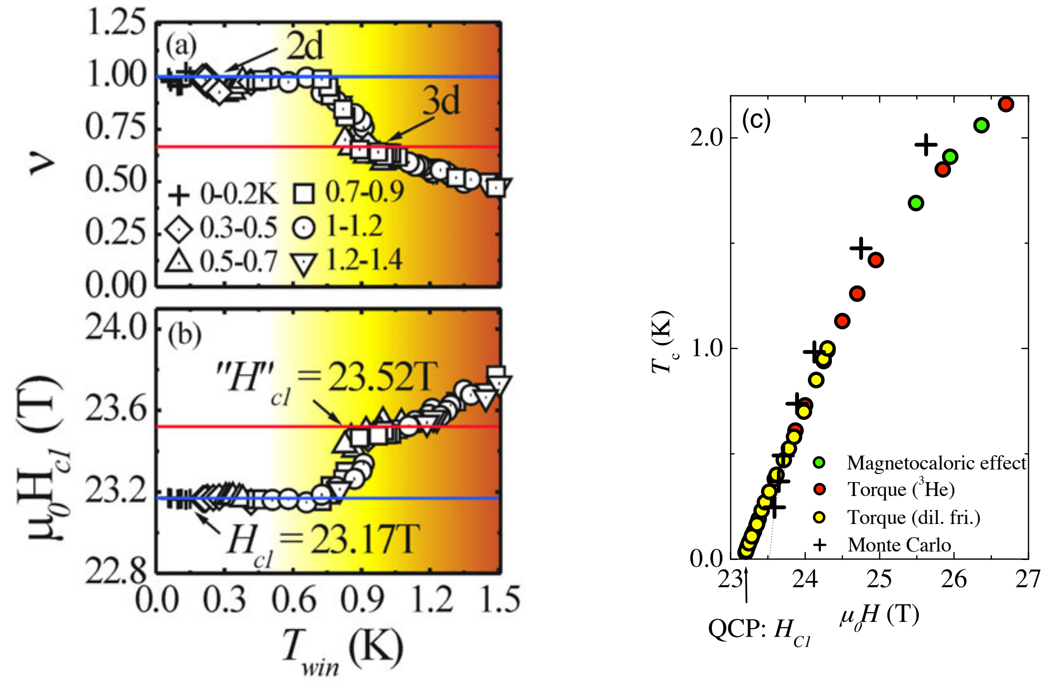


**Figure 2.** Experimental measurements of $\nu$ and $H_{\mathrm{c1}}$ down to 30 mK, showing a crossover from $3d$ to $2d$ behaviour. (a) A two-parameter fit with $\nu$ and $H_{\mathrm{c1}}$ varying is used to fit the data in a sliding window centred at $T_{\mathrm{win}}$ (K). The window-size varies from 0.05 to 1.4 K, as indicated by different symbols. Solid horizontal lines show the theoretical values of $\nu = 2/3$ and 1 for $2d$- and $3d$-BEC respectively. (b) Estimates of $H_{\mathrm{c1}}$ obtained along with $\nu$ during the fit. $H_{\mathrm{c1}}$ approaches 23.52 T in the intermediate regime, and crosses over to 23.17 T before the QCP in approached. (c) Deviation of the experimental phase boundary near the QCP from the Monte Carlo simulation of a $3d$ BEC.

Thus the dispersion can be written as

$$\epsilon(\mathbf{q}+\delta\mathbf{k}) = J - 2J' + \frac{J'}{2}(\delta k_x^2 + \delta k_y^2) + \frac{J_f}{2}\delta k_x \delta k_y (1 + \frac{k_z^2}{2}), \qquad (4)$$

so that there is no $k_z$ dispersion to leading order. This implies that a boson in the condensate that occupies any given layer does not hop to another layer. The vanishing leading order contribution of $k_z$ in the dispersion is a direct consequence of the frustrated form of the inter-layer exchange. The colour shading in figure 3a represents the inter-layer hopping. While an unfrustrated system would disperse with a uniform bandwidth in the $z$-direction, frustration leads to a vanishing bandwidth along $(\pi, k_y, k_z)$ and $(k_x, \pi, k_z)$ directions. Specifically, at the band minimum $\mathbf{q} = (\pi, \pi, k_z)$ where bosons condense, the inter-layer hopping vanishes.

### *4.1. 'Order from disorder' defeated*

A competing effect known as 'order from disorder' arises from zero-point phase fluctuations, which generate an effective inter-layer coupling ($K$) and restore phase-coherence along the $c$-axis. This result was recently obtained by Maltseva and Coleman [16], using a spin-wave approach on the same spin lattice to derive an unfrustrated effective coupling $K$ between adjacent layers. $K$ is biquadratic in the local order parameter, implying a quadratic density dependence.

In $BaCuSi_2O_6$, since the QCP is in the BEC universality class and hence driven by amplitude fluctuations, the particle density grows vanishingly small as the QCP is approached. The '$3d$-

ness' of the system (given by the size of the effective coupling $K$ relative to the temperature scale $k_B T_c$) is therefore arbitrarily small close enough to the QCP, and inter-layer tunnelling is too small for the system to be observably $3d$. By field-tuning toward $H_{c1}$, we experimentally access this region, and hence observe critical power law behaviour consistent with the $2d$-BEC universality class.

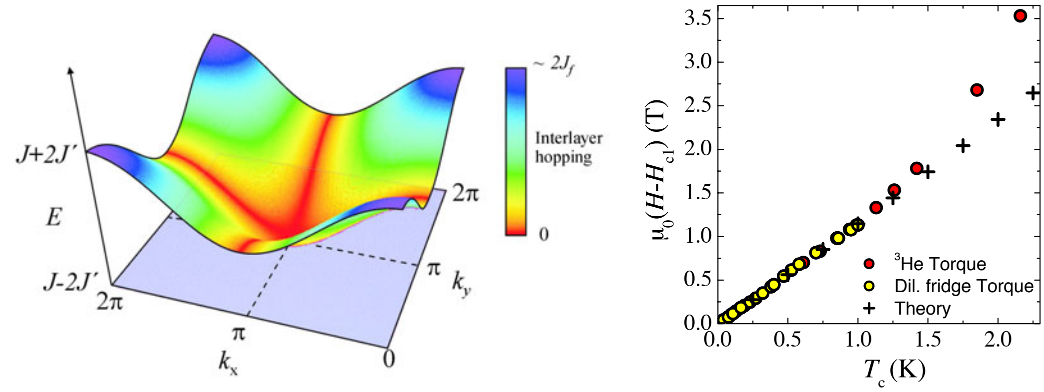


**Figure 3.** (a) Dispersing $s_z = 1$ triplon band in an applied magnetic field of $H_{c1}$, with inter-layer exchange shown on a colour scale. The shape of the dispersion quadratic for all magnetic fields $H \leq H_{c1}$, but the gap to the singlet state decreases with increasing $H$. At $H = H_{c1}$, the gap to the singlet state is closed at wavevector $k_x = k_y = \pi$, resulting in Bose Einstein condensation. The colour shading represents the inter-layer hopping. At the wavevector where BEC occurs, there is no inter-layer hopping. (b) Comparison between the low temperature experimental phase boundary and theoretical prediction for an interacting dilute Bose gas with a $3d$ XY finite temperature transition, but a $2d$ QCP.

## 5. Quantitative comparison with $2d$ interacting Bose gas

To obtain a quantitative measure of whether the system is in fact close to a $2d$ QCP at measured temperatures, we compare the theoretical estimate of the critical temperature $T_c$ with the experimentally measured value. A $2d$ interacting dilute Bose gas undergoes a Kosterlitz-Thouless transition at critical temperature $T_c^{2d}$, the magnitude of which has been estimated in [17] to be:

$$T_c^{2d} \sim \mu \frac{\ln(\frac{\varepsilon_0}{\mu})}{\ln\ln(\frac{\varepsilon_0}{\mu})} \tag{5}$$

where $\varepsilon_0 = \frac{\hbar^2}{2m^* a^2}$, $m^*$ is the effective mass, $a$ is the range of the potential, and $\mu$ is the chemical potential. In the case of $BaCuSi_2O_6$ however, the existence of a small but finite inter-layer coupling away from the QCP leads to a $3d$ XY phase transition at a finite temperature $T_c$ instead of a $2d$ Kosterlitz Thouless transition. The system is in a unique situation where although the classical phase transition is $3d$, the quantum critical point is $2d$, in which case theoretical predictions [18] indicate that

$$\mu \sim H - H_{c1} \simeq \frac{T_c}{\alpha \ln(\frac{\varepsilon_0}{T_c}) + \beta} \tag{6}$$

The value of $\varepsilon_0$ for the experimental system $BaCuSi_2O_6$ is determined by comparing the expression for the energy $\epsilon = \frac{\hbar k^2}{2m^* a^2}$ with the term $J'(\cos k_x + \cos k_y) \simeq J'\left(1 - \frac{k_x^2}{2} - \frac{k_y^2}{2}\right)$ in the dispersion relation (Eqn. 2). Substituting $\varepsilon_0 = \frac{J'}{2} = 3.4$T in Eqn. 6 and using $\alpha$ and $\beta$ as fitting parameters, an excellent fit is obtained to the experimental data as shown in Fig. 3b for $\alpha = 0.03(1)$ and $\beta = 0.83(1)$. We thus confirm that the $BaCuSi_2O_6$ system exhibits a 2*d* QCP, although the finite temperature classical phase transition is of the 3*d* XY type.

## 6. Conclusion

The observation of dimensional reduction at a QCP constitutes a proof of principle that is of fundamental importance to understanding quantum critical phenomenon in anisotropic materials. Geometrical frustration has been established as a potential route to achieve a 2*d* QCP in a bulk material, with possible implications for other 3*d* materials with anisotropic characteristics similar to $BaCuSi_2O_6$. Low dimensionality is recognized as an important factor in high temperature superconductivity and heavy fermion quantum criticality, both of which remain poorly understood. It remains to be investigated whether the anisotropic character typical of several of these systems could result in new types of low dimensional quasiparticle excitations such as those associated with a 2*d* QCP. In the $BaCuSi_2O_6$ system, the interesting possibility arises of a Kosteritz-Thouless-like thermodynamic transition being observed at finite temperatures. Many consider the vanishing quasiparticle velocity at the QCP itself to be a remarkable phenomenon which has even inspired links to cosmology [19].

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