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Spatially resolved magnetization in the Bose-Einstein condensed state of BaCuSi$_2$O$_6$: Evidence for imperfect frustration

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In order to understand the nature of the two-dimensional Bose-Einstein condensed (BEC) phase in BaCuSi$_2$O$_6$, we performed detailed ^{63}Cu and ^{29}Si NMR above the critical magnetic field, $H_{c1} = 23.4$ T. The two different alternating layers present in the system have very different local magnetizations close to H_{c1}; one is very weak, and its size and field dependence are highly sensitive to the nature of inter-layer coupling. Its precise value could only be determined by "on-site" ^{63}Cu NMR, and the data are fully reproduced by a model of interacting hard-core bosons in which the perfect frustration associated to tetragonal symmetry is slightly lifted, leading to the conclusion that the population of the less populated layers is not fully incoherent but must be partially condensed.

Weakly coupled antiferromagnetic (AF) spin-1/2 dimers are currently of great interest since they offer an ideal playground to study the physics of hard-core bosons on a lattice.[1] All these systems have in common a collective singlet ground state, separated by an energy gap from a band of triplet excitations. For magnetic fields H larger than the critical field H_{c1} closing this gap, the dimers acquire a triplet, i.e., a hard-core boson density controlled by the bare chemical potential, that is the reduced magnetic field $h = H - H_{c1}$. Depending on the geometry of the interdimer couplings, a rich variety of ground states can occur: canted XY AF order, which can be described as Bose-Einstein condensation (BEC),[2,3] observed in TlCuCl$_3$,[3,4] BaCuSi$_2$O$_6$,[5] NiCl$_2$-4SC(NH$_2$)$_2$ (DTN),[6] and CuBr$_4$(C$_5$H$_{12}$N)$_2$ (BPCB);[7] magnetization plateaus described as Mott insulators, observed in SrCu$_2$(BO$_3$)$_2$;[8] Bose glasses observed in doped DTN[9] and other systems;[10] and even supersolids[11] for which a physical realization has not been found yet. The hallmark of a BEC system is the field dependence of the temperature T_{BEC} of transition into the ordered phase of canted XY AF type, $T_{BEC} \propto h^\phi$, where $\phi = 2/d = 2/3$ in the standard three-dimensional (3D) case. In two dimensions, the BEC can only occur at $T = 0$. However, depending on the strength and the nature of the interlayer coupling which stabilizes the 3D order, the T_{BEC} vs h dependence of quasi-two-dimensional (quasi-2D) systems can be dominated by the properties of the quantum critical point at $T = 0 = h$ and thus can exhibit a 2D exponent $\phi = 1$.

During the last few years BaCuSi$_2$O$_6$, also known as Han purple, has emerged as an outstanding candidate for studying this quasi-2D case. Its T_{BEC} was found to vary linearly with h, i.e., $\phi = 1$,[12,13] pointing to a quasi-2D BEC. This was originally attributed to frustration between the Bose-Einstein condensates of equivalent adjacent layers because the Cu dimers form a body-centered tetragonal (bct) lattice[12,14] (inset of Fig. 1). In that scenario, the field dependence of T_{BEC} is exotic, linear instead of $h^{2/3}$, but the low-temperature phase is a standard 3D condensate. Whether the frustration is perfect

or not will not affect the nature of the low-temperature phase but will simply induce a crossover to the regular $h^{2/3}$ behavior at very low temperature.[15]

However, BaCuSi$_2$O$_6$ undergoes around 90 K a first-order structural transition[16] which introduces some structural incommensurability and, more importantly, two types of alternating layers, A and B, along the c axis. The B layers have a bigger intradimer exchange coupling and hence bigger local gap than the A layers, as shown by inelastic neutron scattering[17] and by NMR,[13] leading to an about 3 T higher nominal (local) critical magnetic field. In that case, whether frustration is perfect or not makes a qualitative difference regarding the low-temperature phase: For perfect frustration the B layers are only populated by a frustrated proximity effect from the A layers, and they remain uncondensed up to a second critical field,[15,18] with, however, algebraic correlations at zero temperature.[19] By contrast, for imperfect frustration the B layers are also populated by a regular proximity effect, and they contain a condensate which, if large enough, dominates their population. It is thus of crucial importance to determine whether the structural distortion leads to a negligible or to a significant departure from perfect frustration in order to unambiguously identify the nature of the low-temperature phase of BaCuSi$_2$O$_6$ above the first critical field.

A direct way to answer that question is to measure very precisely the *local* magnetization of the *less* polarized B layers. Indeed, as shown in Refs. 15 and 18, the magnetization, i.e., the boson density, of the less polarized layers just above H_{c1} depends crucially on the level of frustration: it is expected to grow quadratically with h if the frustration is perfect[18] and linearly otherwise.[15] In that respect, the ^{29}Si NMR data of Ref. 13, which have clearly demonstrated the existence of two types of layers, are not sufficiently precise, mostly because the lines of the two types of layers overlap.

In this Rapid Communication, we show that this difficulty can be overcome by turning to technically much more difficult ^{63}Cu NMR measurements in the vicinity of the quantum critical field $H_{c1} = 23.4$ T of BaCuSi$_2$O$_6$.[13] Unlike the ^{29}Si
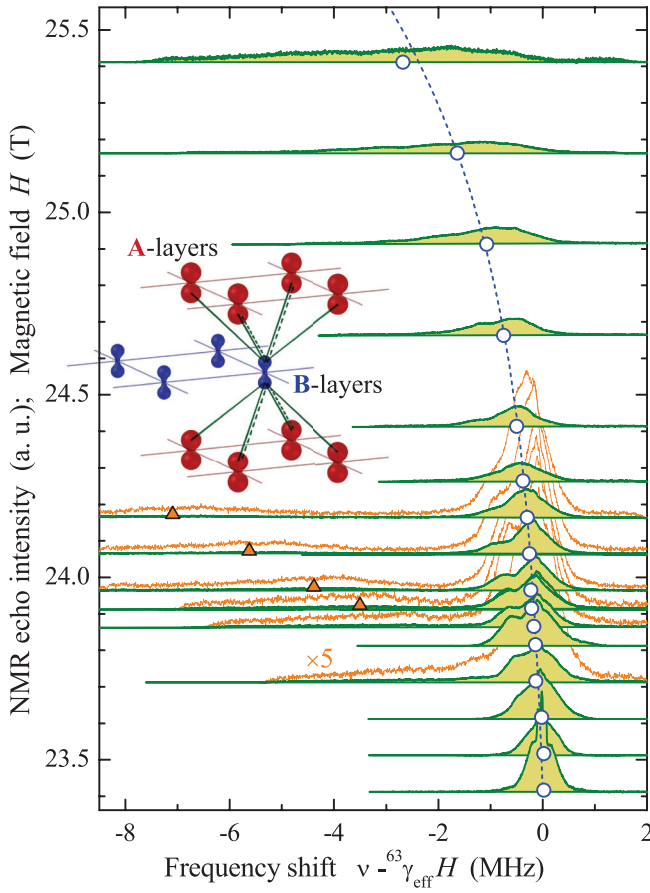
FIG. 1. (Color online) Magnetic field dependence of the low-temperature (0.05–0.08 K) ^{63}Cu NMR spectra on entering the BEC phase of BaCuSi$_2$O$_6$ (green lines). The vertical offset of the spectra defines the corresponding field value. The spectra are almost exclusively due to B layers, and their first moment (open circles) measures the corresponding local magnetization. The dotted line is the theoretical prediction; see the text and Fig. 2. The 5× zoom of selected spectra (orange lines) reveals a weak, broad, and strongly shifted contribution of A layers, and the orange triangles denote a rough estimate of its first moment. The inset shows the schematic structure of the spin-1/2 dimers: green lines present perfectly frustrated, symmetric interlayer exchange couplings, and dotted lines denote weak breaking of this symmetry.

NMR spectra,[13] the ^{63}Cu NMR signal from the B layers is clearly separated from the one from the A layers and provides an accurate determination of boson densities in the B layers as a function of h (Fig. 1). As predicted in Ref. 18, a quadratic h dependence has been observed; however, its size is much stronger than the predicted one, and there is also a non-negligible linear component. We attribute this latter to a deviation from the perfect frustration due to the incommensurate (IC) lattice modulation[16] and a weak breaking of the tetragonal symmetry,[20] both of which characterize the structural distortion at 90 K.

BaCuSi$_2$O$_6$ can be described as Cu^{2+} spin-1/2 dimers located on a bct lattice with Cu-Cu bonds perpendicular to the ab plane (inset of Fig. 1). The real space group at room temperature is indeed $I4_1/acd$.[21] However, below 90 K a first-order structural transition renders adjacent layers

inequivalent[20] and also induces an IC distortion of the lattice along the b axis.[17] This IC distortion converts the typical splitting of each NMR line below T_{BEC} (Ref. 7) into a continuously distributed spectrum.[13] In addition, although the boson densities n_A and n_B in the A and B layers are very different, the ^{29}Si NMR lines originating from the two types of layers overlap due to the small sensitivity of the ^{29}Si NMR to local boson density.[22] Finally, through a significant long-range (dipolar) coupling, the ^{29}Si nuclei from the B layers are also somewhat sensitive to the A layer boson density. Altogether, this renders an accurate determination of the small n_B values by ^{29}Si NMR quite difficult. Although experimentally more complicated, a study by ^{63}Cu NMR presents major advantages. Unlike the distant ^{29}Si nucleus, the ^{63}Cu nucleus is at the site of the electron spin and thus senses only local magnetization through a much larger hyperfine coupling tensor, $^{63}A_{zz} = -16.4$ T/μ_B [Ref. 13; defined later, see Eq. (2)], enhancing the sensitivity of the ^{63}Cu NMR line position to small values and variations of local boson density. One thus expects that above H_{c1} the different n_A and n_B values correspond to separated NMR lines, rendering the determination of n_B much easier.

The ^{63}Cu and ^{29}Si NMR have been performed in the 23–27 T field range, in a resistive magnet at the Laboratoire National des Champs Magnétiques Intenses (LNCMI). The sample was a $2.4 \times 2.4 \times 1.2$ mm^3 single crystal of BaCuSi$_2$O$_6$, grown in a mirror furnace and fully enriched in ^{29}Si to enhance the ^{29}Si NMR signal. It was placed in the mixing chamber of a ^{3}He-^{4}He dilution refrigerator, with its c axis oriented parallel to the external field H, i.e., $c \parallel z$. NMR spectra have been recorded at fixed field by summing up Fourier transforms of the NMR echoes taken at regular frequency steps. The frequency ν for the ^{63}Cu NMR spectra corresponds to the central line of ^{63}Cu and is taken relative to the reference $\nu_0 = ^{63}\gamma_{eff}H$, where $^{63}\gamma_{eff} = ^{63}\gamma(1 + K_{orb})$, with $^{63}\gamma = 11.285$ MHz/T and $K_{orb} = 1.68\%$.[13] The value of the applied field H was calibrated with the metallic ^{27}Al reference placed in the same coil as the sample.

In a Bose-Einstein condensate spins S bear both a longitudinal, $M_z = g_z\mu_B\langle S_z\rangle$, and a transverse, staggered component of the magnetization, $M_\perp = g_\perp\mu_B\langle S_\perp\rangle$, where g_z and $g_\perp$ are the corresponding components of the g tensor. In general, both components define the observed NMR frequency through the corresponding components of the hyperfine coupling tensor A_{zz} and $A_{z\perp}$:

$$\nu_\pm = \nu_0 + \gamma A_{zz}M_z \pm \gamma A_{z\perp}M_\perp. \tag{1}$$

While for the ^{29}Si NMR in BaCuSi$_2$O$_6$ this is indeed the case, the hyperfine tensor of the Cu is dominated by on-site interactions (core polarization and on-site dipolar interaction) and is diagonal with its main component A_{zz} along the c axis. The Cu line shapes are thus insensitive to the staggered magnetization (as long as $H \parallel c$), and the frequency shift of the first moment ν_1 (average frequency) of the line, $\Delta\nu = \nu_1 - \nu_0$, reflects directly the longitudinal magnetization only, that is, the boson density $n_{A(B)}$, by

$$n_{A(B)} \equiv 2\langle S_z^{A(B)}\rangle = 2\Delta\nu_{A(B)}\big/(\gamma g_{cc}\mu_B{}^{63}A_{zz}), \tag{2}$$

where $g_{cc} = 2.30$.[23] This formula implies that $n_{A(B)} = 1$ when there is one boson per dimer in the A (B) layer. With

this convention, one has $n_A + n_B = 2$ at the saturation field $H_{c2} \approx 50$ T. Figure 1 presents the field dependence of the ^{63}Cu NMR line in the BEC phase. The fact that the boson density and its fluctuations are much bigger in the A layer makes their contribution to the spectra nearly unobservable for two reasons. First, the intensity of the A line is much smaller due to a much shorter spin-spin relaxation time T_2 reflecting the fluctuations. This mechanism is particularly effective close to the phase transition. Second, the width of the line, being related to boson density, is expected to be much broader for the A line and thus of proportionally smaller intensity (as the total signal is given by the integral over the line). Indeed, in the enhanced scale spectra in Fig. 1, the A line can be observed as a very weak broad signal to the left of the main B line only above 23.9 T, where the two lines become fully separated. Note that the main line corresponding to the B layer is obviously shifting much more slowly than the one corresponding to the A layer, reflecting a much lower boson density n_B.

Using Eq. (2), the first moments of the lines shown in Fig. 1 are converted to n_B, and their field dependence is plotted in Fig. 2. As n_A determined from ^{63}Cu NMR suffers from huge error bars due to a very poor signal-to-noise ratio of the A lines, we have instead plotted the total boson density $n_A + n_B$, deduced from the first moment of the ^{29}Si line shapes recorded on the same sample.[24] As already mentioned, the contributions of the A and B layers overlap in the ^{29}Si spectrum, and the line shape is sensitive to both the transverse magnetization and IC distortion.[13] Fortunately, both latter effects average to zero when calculating the first moment ν_1. Therefore, $^{29}\nu_1$ of the total Si spectrum, including the A and B layer contributions, gives an accurate determination of $n_A + n_B$ through the formula $n_A + n_B = 4(^{29}\nu_1 - {}^{29}\nu_0)/(^{29}\gamma\, g_{cc}\mu_B\, {}^{29}A_{zz})$, which is the same formula as Eq. (2) corrected for a factor of 2 for proper normalization. A possible source of error is the difference in the $^{29}A_{zz}$ values for the A and B layers; here we have used the
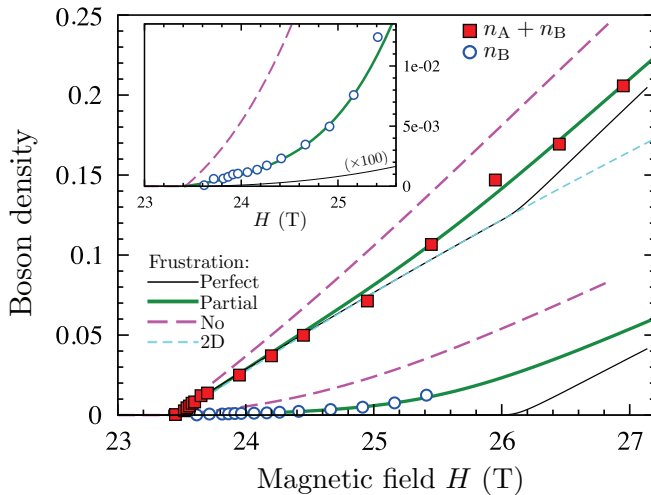

FIG. 2. (Color online) Magnetic field dependence of boson densities. The n_B from ^{63}Cu NMR (open blue circles) and $n_A + n_B$ from ^{29}Si NMR (red squares) are compared to three different theoretical models (see the text) having perfect, partial, and no frustration (thin black, thick green, and dashed magenta lines, respectively). The thin cyan dashed line shows the boson density of identical, decoupled layers (2D). The inset shows a zoom on n_B in the vicinity of H_{c1}.

former one (0.30 T/μ_B) to ensure that the error is negligible in the relevant field range where $n_B \ll n_A$.[24]

We now focus on the variation of n_B as a function of h. In earlier models[14] all the layers were taken to be identical, which is not realized in BaCuSi$_2$O$_6$. When the finite energy barrier, which is the difference between the two local gap values, $\Delta = \Delta_B - \Delta_A \approx 0.4$ meV (Refs. 13 and 17) is considered, perfect frustration along the c axis leads to a classical decoupling of the layers and to the *absence* of bosons in B layers close to H_{c1},[15] a result in contradiction to our previous[13] and new ^{29}Si NMR data,[24] the latter ones being more accurate since they were recorded on a ^{29}Si-enriched sample. However, as shown in Refs. 18 and 19, quantum fluctuations (spin-wave corrections) above such a classical ground state induce above H_{c1} a small boson density $n_B \propto h^2$. As a corollary, a fully three-dimensional coherence is restored in this case through an effective interlayer hopping of bosons, $t_{3D} \propto h$, leading to a modified exponent $T_{BEC} \propto h^{\phi=1}$, in agreement with experiments.[12,13]

In the inset of Fig. 2 we plot in detail $n_B(h)$, which is an order of magnitude smaller than n_A and whose h dependence obviously exhibits a quadratic contribution. However, the measured density n_B turns out to be much larger than earlier theoretical predictions based on a perfectly frustrated scenario.[18,19] Moreover, a linear component is also present in the observed $n_B(h)$ dependence. As already mentioned, the key to explaining these deviations is an imperfect frustration between the A and B layers, which has two origins. According to the recent low-temperature average structure of BaCuSi$_2$O$_6$,[20] the tetragonal symmetry is broken, which introduces two slightly different Cu$_A$-Cu$_B$ distances, corresponding to the two types of Cu$_A$ sites having opposite staggered magnetization. This provides a direct and site-independent lifting of the perfect frustration. Another source of imperfect frustration is the IC modulation in the b direction, which is along the diagonal connecting the next-nearest-neighbor dimers.[16] While both effects are very difficult to estimate, the NMR data provide, with the help of the following theoretical analysis, a reliable estimate of their total "effective" size.

As discussed previously in Refs. 5,14,18, and 19, one can map the system of coupled spin-1/2 dimers in BaCuSi$_2$O$_6$ onto a lattice model of hard-core bosons whose quantum dynamics is governed in each layer $\ell = A, B$ by the Hamiltonian

$$\mathcal{H}_{\parallel}^{\ell} = t_{\parallel} \sum_{\square} [b_{\ell}^{\dagger}(\mathbf{r})b_{\ell}(\mathbf{r}') + \text{H.c.} + n_{\ell}(\mathbf{r})n_{\ell}(\mathbf{r}')]$$
$$- \mu_{\ell} \sum_{\mathbf{r}} n_{\ell}(\mathbf{r}), \tag{3}$$

where the sum runs over the nearest neighbors on a square lattice. It is important to notice the positive (antiferromagnetic) hopping term $t_{\parallel}$ and the distinct chemical potentials on different layers: $\mu_A = \mu_B + \Delta$. The interlayer (or "transverse") tunneling term, which mixes bosons from layers A and B, reads

$$\mathcal{H}_{\perp} = t_{\perp} \sum_{\bowtie} [b_A^{\dagger}(\mathbf{r})b_B(\mathbf{r}') + \text{H.c.} + n_A(\mathbf{r})n_B(\mathbf{r}')]$$
$$+ t_{\perp}' \sum_{\bowtie} [b_A^{\dagger}(\mathbf{r})b_B(\mathbf{r}') + \text{H.c.} + n_A(\mathbf{r})n_B(\mathbf{r}')], \tag{4}$$

where the first term accounts for the perfectly frustrated hopping between A and B and the second term mimics a direct (unfrustrated) tunneling between neighboring layers, which is due to the lattice distortion.[16,20] One can immediately rule out the scenario of identical and perfectly decoupled layers ($\Delta = t_\perp = t'_\perp = 0$)[14] for which the boson density is plotted in Fig. 2 by a thin cyan dashed line. Although this gives an approximately correct description of the total measured density, in particular close to H_{c1}, the B layers should simply not exist in this case. A realistic model should include different layers A and B ($\Delta \neq 0$) and an interlayer coupling between them. In order to get the best description of NMR data, we fix the parameters $t_\parallel = 0.3$ meV and $\Delta = 0.35$ meV and allow $t_\perp$ and $t'_\perp$ to vary. A spin-wave analysis of this effective hard-core bosonic Hamiltonian is performed at the linear $1/S$ order.[18,25] We have examined three possible scenarios: (i) perfect frustration with $t_\perp/t_\parallel = 1/8$ and $t'_\perp = 0$, (ii) no frustration with $t_\perp = 0$ and $t'_\perp/t_\parallel = 1/20$, and (iii) partial frustration with $t_\perp/t_\parallel = 1/8$ and $t'_\perp/t_\parallel = 1/50$. In all these cases, the interlayer barrier $\Delta = 0.35$ meV has been chosen in order to give the best description of the densities measured by NMR. This value appears somewhat smaller than the experimental ones,[13,17] but we expect that the IC modulations, particularly strong in B layers, naturally lead to a reduction of this internal barrier. We now examine in detail the three aforementioned scenarios.

(i) While a perfect frustration is particularly interesting since it would induce an exotic boson density in the B layers,[19] it is unfortunately unable to explain the magnitude of the density n_B. This is evidenced in the inset of Fig. 2, where we see that the quadratic contribution coming from quantum fluctuations is extremely small. Moreover, the perfect frustration scenario causes a clear cusp in the field dependence of the total density $n_A + n_B$ at the nominal critical field of the B planes around $H'_{c1} \approx 26$ T, while in the experimental data, one observes only a slight bending.

(ii) The completely unfrustrated case requires a more detailed discussion. The present analysis focuses on the quantum critical regime where the bosons are very diluted, but we have to keep in mind the full experimental phase diagram, and in particular the maximum $T_{BEC} \approx 3.8$ K value occurring at $H \approx 37$ T,[5] which imposes a transverse tunneling

$t'_\perp = t_\parallel/20$. This value of the direct (unfrustrated) hopping leads to theoretical densities n_A and n_B much larger than the experimental data. In particular, n_B is about five times bigger, and the bending of the $n_A + n_B$ curve has completely disappeared with such a scenario, as seen in Fig. 2.

(iii) We are then left with a third scenario of partial frustration, which is able to qualitatively and quantitatively explain all the features observed in NMR experiments: the small but finite density n_B, which has a pronounced convexity, and the total density $n_A + n_B$, which smoothly departs from a 2D behavior above 25 T. The theoretical fit given in Fig. 2 corresponds to a frustrated coupling $t_\perp/t_\parallel = 1/8$, dominantly determined by the total density $n_A + n_B$, and a much smaller direct hopping $t'_\perp/t_\parallel = 1/50$, which controls n_B. This coupling lifts the perfect frustration by $2t'_\perp/(4t_\perp + 2t'_\perp) \approx 7\%$. Regarding the scaling of $T_{BEC}(h)$, we expect the contribution of such a small direct coupling to restore a true 3D critical regime with an exponent $\phi = 2/3$ only below $(t'_\perp)^2/\Delta \approx 10$ mK, which is out of the current experimental range of measured $T_{BEC}(h)$ in $BaCuSi_2O_6$.[12,13]

To summarize, using ^{63}Cu and ^{29}Si NMR, we have shown that the population of the less polarized layers of $BaCuSi_2O_6$ is not purely quadratic in $(H - H_{c1})$ and that it is much larger than predicted assuming perfect frustration. This unambiguously demonstrates the presence of *imperfect* frustration of the interlayer couplings, with an unfrustrated component of about $\approx 7\%$ of the total interlayer coupling strength, and leads to the conclusion that the population of the B layers is not fully incoherent, as it would be if frustration was perfect, but that it must be partially condensed as soon as $H > H_{c1}$. To probe directly this small condensate fraction is a challenging task that is left for future investigation.

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[22]As discussed in our previous study (Ref. [13]), four Si atoms are approximately located in the equatorial plane of each Cu dimer. Their ^{29}Si nucleus senses the local magnetization (boson density) of the nearest dimer via a transferred hyperfine interaction and a direct dipolar interaction. By means of the latter ^{29}Si additionally senses the magnetization of the surrounding dimers belonging to both types of layers.

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