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Field dependence of the quantum ground state in the Shastry-Sutherland system $SrCu_2(BO_3)_2$

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Abstract – We present magnetic torque measurements on the Shastry-Sutherland quantum spin system $SrCu_2(BO_3)_2$ in fields up to 31 T and temperatures down to 50 mK. A new quantum phase is observed in a 1 T field range above the 1/8 plateau, in agreement with recent NMR results. Since the presence of the DM coupling precludes the existence of a true Bose-Einstein condensation and the formation of a supersolid phase in $SrCu_2(BO_3)_2$, the exact nature of the new phase in the vicinity of the plateau remains to be explained. Comparison between magnetization and torque data reveals a huge contribution of the Dzyaloshinskii-Moriya interaction to the torque response. Finally, our measurements demonstrate the existence of a supercooling due to adiabatic magnetocaloric effects in pulsed field experiments.

Quantum antiferromagnetic spin systems with singlet ground states exhibit a variety of magnetic-field–induced quantum phase transitions. Crystalline arrays of $S=1/2$ spin dimers, for instance, can present two contrasting behaviors [1]. When the magnetic field exceeds a critical value at which the lowest energy levels cross each other, the triplet excitations, which can be treated as hard core bosons on a lattice, typically undergo a Bose-Einstein condensation (BEC) [2–4]. Another possibility, however, is the occurrence of magnetization plateaus at fractional values of the saturated magnetization. Such plateaus correspond to the formation of a superlattice of triplets ("magnetic crystal") and may occur when the kinetic energy of the triplets is strongly reduced by frustration, so that the repulsive interactions become dominant. The best known example for the formation of such plateaus is $SrCu_2(BO_3)_2$ with its two-dimensional network of orthogonal dimers of $S=1/2\,Cu^{2+}$ ions [5]. This material shows an excitation gap $\Delta_0=35\,K$ and plateaus at 1/8, 1/4, and 1/3 of the saturated magnetization [6,7]. A magnetic superlattice at the 1/8-plateau has actually been observed in NMR experiments [8]. It has been argued on theoretical basis that some analog of a supersolid phase [9–12], consisting of the superposition of the magnetic crystal and a Bose-Einstein condensate of the interstitial triplets, could occur in the vicinity of the plateau phases. The presence of such an exotic phase, the magnetic analog of the highly debated supersolid phase in ^4He, is however precluded in $SrCu_2(BO_3)_2$ because of the presence of an intradimer Dzyaloshinskii-Moriya (DM) interaction [13,14] which breaks the $U(1)$ symmetry. However, NMR measurements have recently revealed the existence of new magnetic phases above the 1/8 plateau. This prompted us to re-examine the field-temperature (H-T) phase diagram of $SrCu_2(BO_3)_2$ up to 31 T using torque measurements. Our experiments indeed confirm the existence of a new phase adjacent to the 1/8 plateau, in which the magnetization is only slowly increasing. In addition, we report for the first time the field dependence of the pure longitudinal magnetization up to 31 T, measured at the temperature of 60 mK. The results strongly differ from those obtained by torque measurements, as expected in the presence of DM interaction within the dimers [15]. In particular, the magnetization jump before the 1/8 plateau is much larger than previously reported [6,16], in excellent agreement with NMR data.

The torque measurements were performed at the Grenoble High Magnetic Field Laboratory in a 20 MW resistive magnet equipped with a dilution refrigerator. The sample ($\sim 1\times1\times0.5\,mm^3$ size) was mounted on a 25 μm thick

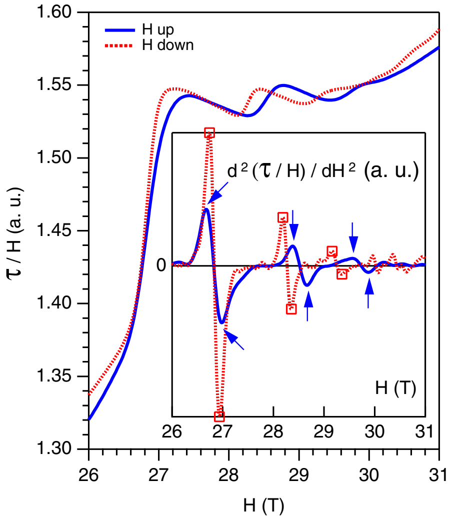

Fig. 1: (Colour on-line) Torque divided by field *vs.* field. The blue continuous line corresponds to a field sweep up at 55 ± 10 mK. The red dashed line corresponds to a field sweep down at 90 ± 10 mK. The inset shows the corresponding second derivatives $\mathrm{d}^2 m_\perp/\mathrm{d}H^2$. Six extrema (three transitions) are found in each sweep and pointed out by arrow for field sweep up, and by squares for field sweep down. For a given transition, the field range between the maximum and the minimum of $\mathrm{d}^2 m_\perp/\mathrm{d}H^2$ corresponds to the range in which both adjacent phases coexist.

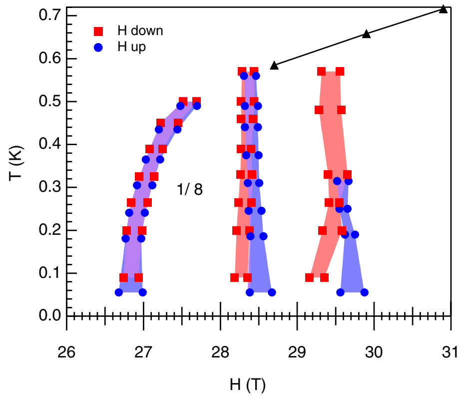

Fig. 2: (Colour on-line) Temperature-field phase diagram. Position of the extrema of the second derivative $\mathrm{d}^2 m_\perp/\mathrm{d}H^2$ made at various constant temperature (see fig. 1). Three transition lines are clearly appearing. For a given transition, the distance between the two corresponding extrema corresponds to the field range within which the two adjacent phases coexist. The blue circles correspond to field sweep up and the red squares to field sweep down. The NMR points, black triangles, delimit the boundary of the magnetic ordering.

CuBe cantilever with its c-axis perpendicular to the surface. *In situ* rotation allowed us to obtain an angle θ between the c-axis and the applied magnetic field of about $0.4°$, a configuration which has been used for most of the experiments. Torque measurements have been performed at various constant temperatures while sweeping field up (at a rate of 100 G/s) and down (at a rate of 200 G/s).

When the sample is placed in a homogeneous field, it is submitted to a torque $\tau = \mathbf{M} \times \mathbf{H}$ in which $\mathbf{M}$ is the magnetization and $\mathbf{H}$ the applied field. This torque is transmitted to the cantilever, which can rotate around a fixed axis perpendicular to $\mathbf{H}$. So the torque is equal to $\tau = m_\perp H$, where $m_\perp$ is the component of the magnetization perpendicular to field and to the rotation axis of the cantilever. One must be careful that the variation of $m_\perp$ as a function of H is not necessarily the same as that of the longitudinal magnetization m_z. However, if the sample is placed in a field gradient, one additionally obtains an access to the magnetization parallel to the applied field, provided the torque component becomes negligible with respect to the force $\mathbf{F} = -\mathbf{M}\mu_0 \vec{\nabla} H$. For this we moved the sample by 1 cm above the magnet center. From here on, we will refer to such measurements as "true" magnetization measurements.

Figure 1 shows the results obtained at the lowest temperature. Three anomalies are clearly visible in the second derivative of the torque divided by field, that is of $m_\perp \propto \tau/H$. The first two correspond to the boundaries of the 1/8 plateau. Above the plateau, there is a second phase which ends up around 29.5 T. In both phases, $m_\perp$ is slightly decreasing with increasing the field. Comparing data acquired in ramping up and down the magnetic field, one observes no hysteresis for the lower boundary of the plateau. This absence of hysteresis as a function of H has already been observed in NMR experiments [8], in spite of the fact that the transition is of the first order, as indicated by the coexistence of the two phases. This contrasts with the two other transitions, which exhibit a rather strong hysteresis.

The position of the peaks found in the second derivative of $m_\perp$ (see fig. 1) are reported in fig. 2 to establish the field-temperature phase diagram. The transition temperatures delimiting the phase boundary between the paramagnetic and the ordered magnetic states (black triangles) are taken from NMR results [17]. The distance between the two extrema of the second derivative for a given transition should roughly give the range of magnetic field within which the two adjacent phases coexist. This is indeed in excellent agreement with the NMR data: at very low temperature, the coexistence between the uniform paramagnetic phase and the triplet superlattice starts at 26.6 T and the uniform phase disappears at 27 T [8]. The transition from the 1/8 plateau into the adjacent phase above was found to start at 28.3 T at 0.31 K both by NMR [17] and torque measurements. Considering the transition around 29.5 T, one observes that the hysteresis becomes much stronger below 200 mK. Indeed, a modification of the NMR lineshape has been observed at temperatures below 200 mK close to the beginning of the phase following the 1/8 plateau [17]. So, one cannot completely exclude the possibility that there is a new phase at low temperature, which would be seen by torque measurements only through the hysteresis of the transition around

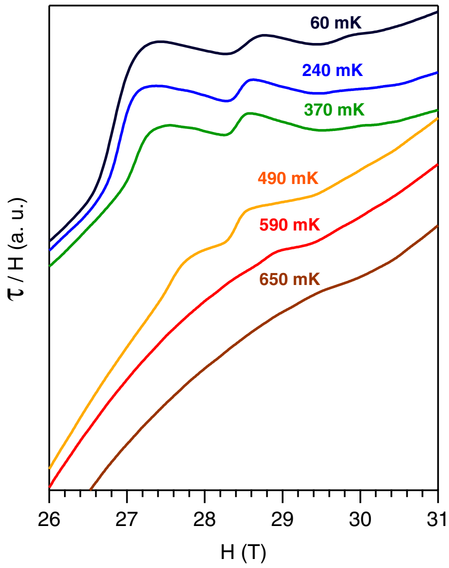

Fig. 3: (Colour on-line) Torque divided by field *vs.* field at different temperatures. The measurements were made for field sweep up. The curves have been arbitrary shifted for clarity.

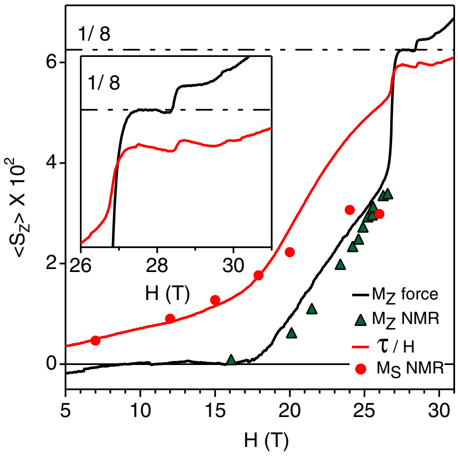

Fig. 4: (Colour on-line) Comparison of torque and magnetization. Magnetization was measured both by NMR technique, green triangles, and by torque in a field gradient, continuous black line. Those measurements are compared with torque divided by field, the continuous red line. The solid red circles correspond to the transverse staggered magnetization measured by ^{11}B NMR.

29.5 T. This should deserve further investigation. We now consider the temperature dependence of $m_\perp$ shown in fig. 3. There are two remarkable features in these data. First, one can see that the signature of the 1/8 plateau has fully disappeared at 590 mK, while there is still some reminiscence of the adjacent phase at this temperature. This again is in excellent agreement with previous NMR data [8], but contrasts with earlier measurements made in pulsed magnetic field performed at 1.4 K [6,16], in which the signature of the plateau is still visible. Since the gap between the lowest triplet excitations branch and the singlet state decreases toward a very small value [14,18] as the magnetic field is approaching the value corresponding to the entry of the plateau, the "high temperature" observation of the plateau in pulsed magnetic field can be interpreted in terms of adiabatic (isentropic) cooling of the spin system [19,20]. As long as only the lowest triplet branch and the singlet state are necessary to describe the system, this is an analog of the cooling of paramagnetic salts by adiabatic demagnetization. The second remarkable feature in fig. 3 is that between 370 mK and 490 mK the slope of τ/H changes from negative to positive before the complete melting of the magnetic superlattice. This corresponds to the regime in which the triplet superlattice and the paramagnetic phase coexist, which was found by NMR to extend between 360 mK and 520 mK at 27.6 T [21]. Such a change of sign, related to the coexistence of the superlattice and the uniform phase, can only be explained if the contribution to τ/H of the superlattice has a negative slope with increasing H, while that of the uniform paramagnetic phase has a positive one. This is rather surprising, since within the plateau one would expect the torque signal to remain constant. In order to elucidate this issue, we have performed a "true magnetization" measurement in which the bending of the cantilever is now dominated by the force $F_z = -M_z \mathrm{d}B_z/\mathrm{d}z$. The results are shown in fig. 4, together with a torque measurement recorded at the same temperature. In addition, solid triangles show the amplitude of $\langle S_z \rangle$ as determined by NMR. The field dependence of the longitudinal magnetization M_z actually strongly differs from the results obtained by torque. M_z is *flat* within the 1/8 plateau, as expected, and within the adjacent phase between 28.4 and $\simeq 29.5$ T it is nearly flat with only a small increase as approaching the upper boundary. The magnetization data are in excellent agreement with the NMR results. In particular, both techniques reveal a large jump of the magnetization just before the 1/8 plateau, in contrast to all previously reported data [6,16].

We also remark that recently it has been proposed that the 1/8 plateau could be preceded by a 1/9 plateau, and followed by a 1/7 plateau [22]. Indeed, the authors of ref. [22] report three transition lines around 27.1, 29 and 30.3 T. It turns out that by reducing these three fields values by 2% we recover the phase boundaries values reported in this letter. These latter values agree with those determined by NMR, a technique which inherently always provides a precise determination of the magnetic field. Therefore, the values reported in ref. [22] appear to result from an incorrect field scale. Furthermore, looking carefully at the pulsed field measurements of ref. [6], the magnetization values measured for the 1/4 and 1/3 plateaus clearly indicate that the phase extending between 26.7 and 28.3 T can only correspond to the 1/8 plateau. As far as the adjacent phase is concerned, our measurements of the longitudinal magnetization demonstrate that it cannot correspond to a 1/7 plateau, since the increase of M/M_{sat} is too small.

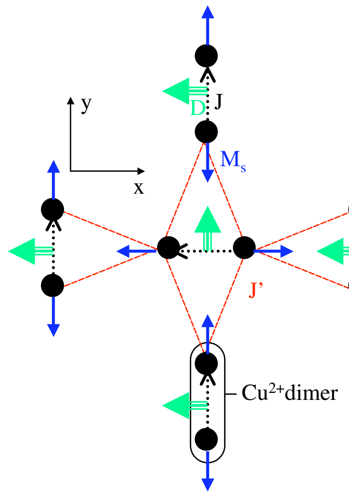

Fig. 5: (Colour on-line) Schematic structure of the Cu^{2+} dimers. The figure sketches the orthogonal network of Cu^{2+} dimers. Black circles represent the Cu^{2+} ions. Intradimer interactions are represented by dotted black lines, and interdimer interaction J' by dashed red lines. Thick green arrows indicate the direction of **D**-vectors for the intradimer Dzyaloshinskii-Moriya interaction $\mathbf{D}\cdot\mathbf{S}_i\times\mathbf{S}_j$, where the bond "direction" $i\rightarrow j$ is shown by black dotted arrows. The solid blue arrows represent the staggered magnetization M_s induced by a field applied along the c-axis (z-axis), within the field range below the first magnetization plateau. Interdimer DM interactions, which are less effective to generate a transverse magnetization, are not shown here.

$SrCu_2(BO_3)_2$ crystallizes in a tetragonal structure with alternative layers of Sr and $Cu(BO_3)$ planes along the c-axis. At low temperature, due to the buckling of the BO_3-Cu-O-Cu-BO_3 bonding, the ab-plane is no longer a mirror plane [23], allowing the existence of an in-plane intradimer DM interaction as shown on fig. 5, as well as a staggered g-tensor. Interdimer DM interactions are also present [24], but their role is mainly to partially restore some kinetic energy to the triplets. The strong difference between the longitudinal magnetization $M_z(H)$ and the torque signal as shown in fig. 4 is indeed the signature of the intradimer DM interaction and the staggered g tensor. The presence of the intradimer DM interaction has been shown to generate a transverse staggered magnetization, observed by ^{11}B NMR and computed by exact diagonalization [14]. While this transverse staggered magnetization has no effect on the torque, it has been shown recently that there is an additional *uniform* transverse component generated by the DM interaction, which is smaller by an order of D/J [15]. In the low field limit and for an isolated dimer, this component for each dimer has the symmetry of $\mathbf{D}\times\mathbf{D}\times\mathbf{H}$. Within the low-field approximation, the torque per spin dimer can be expressed as

$$\tau=(\chi_{ab}-\chi_c)\sin\theta\cos\theta H^2-g\mu_{\mathrm{B}}D^2/4J^3\sin\theta\cos\theta H^2,$$

where θ is the angle between the c-axis and the applied magnetic field, and χ is the part of the susceptibility

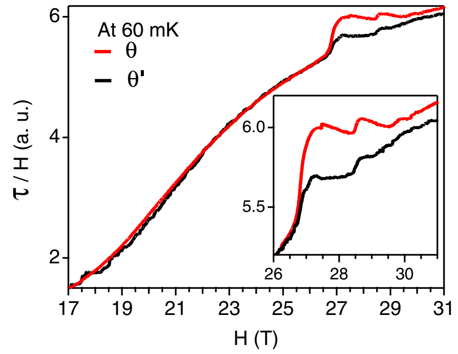

Fig. 6: (Colour on-line) Dzyaloshinski-Moriya contribution within the 1/8 plateau and its adjacent phase. Torque data have been recorded at $\theta=+0.4°$ and at $-|\theta'|$. The latter data have been rescaled to give the same signal at 17 and 26 T. Two signals strongly differ within the plateau and its adjacent phase, which is attributed to a symmetry breaking.

which only depends on the symmetric part of the g-tensor.

The first term is the standard contribution which is proportional to the longitudinal magnetization for small values of θ, while the second one results from the DM interaction. Both terms have the same angular dependence, and vanish for $\theta=0$ ($H\parallel c$). In fig. 4 the field dependence of τ/H is compared to that of the staggered magnetization determined from NMR measurements [14]. In the low field limit, in which the torque signal is only (as long as $M_z=0$) or mainly due to the DM interaction, their variation is quite similar, which is in agreement with the theory predicting that both quantities vary linearly with H. When $M_z\neq 0$ the torque becomes the sum of two contributions and a direct comparison is no longer possible. Exact diagonalization calculations are required to determine the full field dependence of the "uniform" transverse magnetization in the Shastry-Sutherland geometry.

We now consider the τ/H variation within the plateau and its adjacent phase and try to understand the origin of the negative slope observed within the 1/8 plateau. Figure 6 shows torque measurements recorded at θ and at $-|\theta'|$. Since, as expected, the corresponding raw data have different sign, those corresponding to $-|\theta'|$ have been renormalized in order to give the same values at 17 and 26 T. One can see that the field variation of both signals are identical in the uniform phase, as expected if they only differ by a factor $\sin\theta\cos\theta$. However, they strongly differ within the 1/8 plateau and its adjacent phase, in which an extra contribution is observed. This is expected, at least for the 1/8 plateau, for the following reason. Within the uniform phase, the DM interaction is conserved by the three symmetry operations of the crystallographic structure: the mirror plane zx, the mirror plane yz and a C2 rotation at the intersection of these mirror planes. However, within the 1/8 plateau, the structure determined for the magnetic superlattice [8] has only one symmetry left, which is a C2 rotation around the middle of the most polarized dimer. We thus expect that the angular dependence of the torque signal becomes different, and starts to depend also on the angle between the projection of H on the ab-plane and the crystallographic axes. Recently, the investigation of frustrated ladders with DM interactions in a magnetic field [25] has been extended to the situation where the field is neither parallel nor perpendicular to the **D**-vector [26]. It has been shown that the torque induced by the DM interaction develops peaks upon entering and leaving the 1/2 magnetization plateau. While the torque produced by the misalignment of the field with a principal axis of the g-tensor increases monotonously with the field, the torque induced by the DM interaction is non-monotonous inside the plateau, in qualitative agreement with the present observation. Whether this anomalous contribution disappears or not above 29.5 T, where we know from NMR that the "magnetic crystal" persists [17], is not clear at the moment, and would require new measurements.

What is the nature of the phase adjacent to the 1/8 plateau? Recent NMR experiments have shown that the magnetic superlattice, analogous to a magnetic crystal, does not melt when additional triplets are introduced. One can then immediately suspect that this new phase is the analog of a supersolid phase, in which the additional triplets would undergo a Bose-Einstein condensation. However, the presence of the intradimer DM interaction and the resulting staggered magnetization break the $U(1)$ symmetry around the applied magnetic field and thus remove the continuous symmetry of a supersolid phase. So, some more sophisticated theoretical description of the exact nature of this phase has to be provided in the future.

In conclusion, we have determined the phase diagram of the Shastry-Sutherland quantum antiferromagnet $SrCu_2(BO_3)_2$ in the (H-T)-plane up to 31 T, using both magnetic torque and "pure longitudinal" magnetization measurements. We show that the torque measurements allow the detection of the phase transitions between successive quantum ground states, but cannot give access to the true variation of the longitudinal magnetization M_z. This is due to the existence of an intradimer Dzyaloshinskii-Moriya interaction, which generates an additional uniform transverse magnetization providing a strong contribution to the torque. In the low-field limit this contribution to τ/H scales linearly with the transverse staggered magnetization measured by NMR. The phase boundaries of the 1/8 magnetization plateau are found to be in agreement with NMR data: at 60 mK, the coexistence between the uniform paramagnetic phase and the 1/8 plateau extends from 26.6 to 27 T, and the plateau ends through a first-order transition starting at 28.3 T. The temperature corresponding to the complete melting of the spin superlattice in the 1/8 plateau is at most 570 mK. This demonstrates that the observation of the 1/8 plateau at 1.4 K in pulsed field measurements is due to isentropic adiabatic cooling of the spin system. The most important finding of this study is the evidence of a new phase adjacent to the 1/8 plateau and extending up to 29.3 T. The magnetization within this phase is nearly field independent and only increases when approaching its upper boundary. However, its value does not correspond to any simple rational value of the saturation magnetization. Recent NMR measurements show that the "magnetic crystal" does not melt in that phase. However, since the Dzyaloshinskii-Moriya interaction removes the continuous rotation symmetry in $SrCu_2(BO_3)_2$, this phase has to be more complex than a simple analog of a supersolid, which would correspond to the coexistence of the 1/8 plateau "magnetic crystal" and a Bose-Einstein condensate of the interstitial triplets. This clearly shows that our understanding of the physics of interacting hard-core bosons on a lattice has to be improved, and that further theoretical and experimental investigation are necessary to clarify the evolution of the quantum ground states between the 1/8 and the 1/4 plateaus in this model compound.

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