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## Reply to the Comment by S.E. Sebastian and N. Harrison

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In their comment, Sebastian and Harrison (SH) suggest that our torque and force measurements in  $\text{SrCu}_2(\text{BO}_3)_2$  [1] are incorrect due to a non-linear regime in the cantilever operation and deny the importance of Dzyaloshinski-Moriya (DM) interaction in torque measurements. In this reply, we show that their arguments are incorrect and why neither the torque measurements [1, 2] nor the magnetization measurements in pulsed magnetic field [3, 4] can give access to the field dependence of the longitudinal magnetization at thermodynamic equilibrium.

First of all, any non-linearity is perfectly excluded in all our measurements. Our largest signal reached only 10% of the whole linear response range, and the variation of the capacitance has always been smaller than 1% of the zero field capacitance. This means that the deflection of the cantilever was smaller than  $10^{-4}$  rad, which is comparable to the numbers given in the comment ( $0.5 \times 10^{-4}$  rad). Next, the angle  $\theta$  between the  $c$ -axis of the sample and the applied magnetic field  $H$  in our experiment was smaller than  $0.4^\circ$ , which is much smaller than the  $2^\circ$  mentioned in [2] and comparable to the  $\ll 1^\circ$  declared for the experiments reported in the comment.

SH base their interpretation of the torque results on the assumption that DM effects are negligible, as their data follow closely those obtained by inductive method in pulsed magnetic field, which couples only to the total magnetization  $M_z$ . This statement is obviously wrong since in the gapped phase below 16 T the pulsed field measurements correctly record zero magnetization, while the torque measurements *do* record a non-zero signal - that is the signal due to DM interaction. Indeed, Ref. [2] explicitly mentions that at low fields the data have been corrected (put to zero) by hand, and in the Fig. 1 of the comment it is obvious that below 16 T there is an important variation of the torque signal. Furthermore, in the same figure the steep jump towards the 1/8 plateau

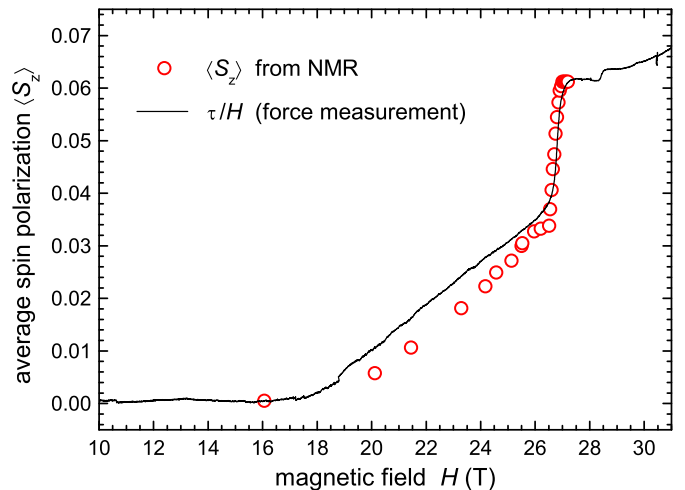


Fig. 1: **Magnetization from NMR and force measurements** The field dependence of magnetization, and in particular the jump towards the 1/8 plateau, recorded by our force measurement [1] is in excellent agreement with values determined from the NMR data of Ref. [5] as explained in the text.

is obviously *not* the same in the torque and in the pulse field data. So the main assumption of SH is contradicted by their own data. Finally, Fig. 3 in Ref. [3] shows that below 30 T the magnetization measurements do *not* scale with the  $g$ -tensor anisotropy, at variance with their claims.

SH criticize the large jump in the magnetization curve observed in our force measurements [1], in which the deviation of the cantilever is dominated by the force  $F = \frac{dH}{dz} M_z$ . The variation of  $M_z$  during this "jump" indeed corresponds to the coexistence of the uniform magnetization phase and the 1/8 plateau phase as already shown by copper NMR [5]. Kodama *et al.* have shown that up to

26.5 T the system is in a uniform phase in which all copper electronic spins bear identical magnetization  $g\mu_B S_z$ . In this phase,  $S_z$  values have been accurately determined by NMR and found to increase up to the value of 0.034 at 26.5 T (Fig. 4A of Ref. [5]). Between 26.5 T and 27 T, the increase of the volume fraction  $x(H)$  of the plateau phase as a function of  $H$  has also been determined by NMR (Fig. 4C of Ref. [5]). One can thus easily deduce the field dependence of the average bulk magnetization  $\langle S_z \rangle = 0.034(1-x) + 0.0625x$ , which is reported in Figure 1. Clearly, the agreement between the force measurement and the NMR data is excellent, meaning that the rapid variation of  $M_z(H)$  in the field range 26.5–27 T *does* correspond to the physics of the system. This clearly indicates that all previously reported measurements, whatever using torque technique in static field or flux integration in pulsed field, were unable to give the correct equilibrium  $M_z(H)$  dependence.

In the following, we explain the possible origin of discrepancies between different techniques. In pulsed magnetic field measurements, during the fast increase of the field the energy levels of the lower triplet states are lowered towards the singlet level, but their populations have not enough time to relax to its thermal equilibrium value (adiabatic process). The effective temperature describing this non-equilibrium level populations is thus much lower than that of the experiment. This explains the observation of 1/8 magnetization plateau in pulsed magnetic field experiments performed at 1.4 K [3], 1.6 K and 0.6 K [4], although this phase is *not* stable above 0.55 K. This also implies that the values of  $M_z$  in these experiments are incorrect.

Let us come to the torque measurements, and why the results of different groups can differ. There are three contributions to the torque applied on the cantilever. The first one is well known and related to the anisotropy of the diagonal part of the  $g$ -tensor. The second one is due to the field gradient ( $dH/dz \neq 0$ ) if the sample is not rigorously at the center of the field. The third one is related to the presence of the DM interaction and the staggered  $g$ -tensor in the system. They induce a staggered transverse magnetization, but also a *uniform* transverse magnetization contributing to the torque [6]. This contribution vanishes at zero  $\theta$ , but is otherwise always present in experimental results [1]. The interplay between the three contributions to the torque is very dependent on the position  $z$  of the sample with respect to the center of the field and the angle  $\theta$ , which can be varied independently. We first focus on the first two terms. For small values of  $\theta$ , the torque divided by the magnetic field can be written as:  $\tau/H = [(g_c - g_a)/g_c \sin 2\theta + l/H dH/dz]M_z$ , where  $l$  is the length of the cantilever,  $g_c = 2.28$  and  $g_a = 2.05$  [7]. Close to the center the field profile is parabolic, so that  $dH/dz = z d^2H/dz^2$ . Clearly one can find a position  $z_0$  where the two terms cancel:  $z_0 = -2\theta/l (g_c - g_a)/g_c [d^2H/dz^2]^{-1}$ . For typical values of  $\frac{d^2H}{H dz^2} = -50$  ppm/mm<sup>2</sup> and  $l = 5$  mm,

this leads to  $z_0$  (mm) =  $14 \theta(^{\circ})$ , e.g.  $z_0 = +6$  mm for  $\theta = 0.4^{\circ}$ . That is, depending on the position of the sample, one can not simply predict the relative magnitude and sign of the contribution proportional to  $M_z$  and that due to the transverse magnetization induced by the DM interaction. This can well explain why our torque measurements exhibit a negative slope within the plateau, while those of SH exhibit a positive slope. Unfortunately, there is no calculation yet of the transverse uniform magnetization generated by the DM and the staggered  $g$  tensor on a Shastry-Sutherland lattice, in presence of a superstructure of the magnetization corresponding to a plateau at  $M_z/M_{saturation} = 1/8$  [5] or  $1/9$  [8].

One puzzling feature in our results, however, is that the signal amplitudes obtained in the force and in the torque measurements are comparable, although in principle one would expect the contribution of the DM interaction to be much smaller [6]. Since we have not done a systematic study as a function of the angle  $\theta$  and the position  $z$ , we cannot disentangle the relative contributions of the longitudinal magnetization and the transverse one. Furthermore, it has been shown recently [9] that in frustrated spin ladders the DM interaction can give a huge contribution to the torque when the angle between the magnetic field and the DM vector is small. In  $\text{SrCu}_2(\text{BO}_3)_2$  the interdimer DM vector is nearly parallel to the  $c$ -axis [10], and may thus be a possible source of a strong torque signal. As regards our force measurement, considering their agreement with the NMR data, they appear to give a correct field dependence of the magnetization, at least up to the 1/8 plateau. The theoretical prediction for the absolute value of the magnetization in the plateaus in  $\text{SrCu}_2(\text{BO}_3)_2$  is currently controversial [2, 8, 11], which makes their experimental determination highly desirable. This requires a special experimental setup to minimize as much as possible any torque contribution with respect to the effect of force due to the field gradient applied on the sample [12].

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