Torque anomalies at magnetization plateaux in quantum magnets with Dzyaloshinskii-Moriya interactions

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Abstract – We investigate the effect of Dzyaloshinskii-Moriya (DM) interactions on torque measurements of quantum magnets with magnetization plateaux in the context of a frustrated spin-1/2 ladder. Using extensive DMRG simulations, we show that the DM contribution to the torque is peaked at the critical fields, and that the total torque is non-monotonous if the DM interaction is large enough compared to the g-tensor anisotropy. More remarkably, if the DM vectors point in a principal direction of the g-tensor, torque measurements close to this direction will show well-defined peaks even for small DM interaction, leading to a very sensitive way to detect the critical fields. We propose to test this effect in the two-dimensional plateau system SrCu$_2$(BO$_3$)$_2$.

The investigation of quantum magnets in high magnetic fields is a very active field of research thanks to a number of recent and remarkable discoveries ranging from Bose-Einstein condensation [1] to magnetization plateaux [2,3], and to the on-going search for the analog of supersolid phases [4–7]. A central piece of information is provided by the magnetization as a function of the external field. However, technical requirements have lead many groups to measure the torque rather than the magnetization. In SU(2)-invariant magnets, it does not matter since the torque is rigorously proportional to the magnetization along the field. In the presence of anisotropic interactions, such as Dzyaloshinskii-Moriya (DM) interactions [8], this is no longer the case and an additional response is obtained which needs to be considered carefully. This DM contribution to the torque has already been investigated in the context of molecular magnets [9,10] and of the spin ladder compound Cu(Hp)Cl [11]. In the latter case, the DM interactions were assumed not to compete with the exchange, so that their additional effect on the torque is a relatively smooth contribution in the intermediate phase. Recent torque measurements on SrCu$_2$(BO$_3$)$_2$, a quasi-two-dimensional quantum magnet with several magnetization plateaux [2,3], call for further investigation of the issue. In particular, the field dependence of the torque reported in refs. [12,13] is quite different, especially regarding the behavior inside the plateaux. In Sebastian et al.’s data, the torque is never flat but increases quite significantly inside the plateaux, while in Levy et al.’s data, it actually decreases inside the 1/8 plateau. Since DM interactions have been unambiguously identified in SrCu$_2$(BO$_3$)$_2$ [14–16], the torque is not expected to be simply proportional to the magnetization, which leaves the door open to anomalous behavior depending on the details of the experiments, in particular the orientation of the field. However, the field dependence of the torque around magnetization plateaux has never been investigated so far.

In this letter, we investigate this issue in the context of the frustrated ladder depicted in fig. 1 and defined by the

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Hamiltonian \((h \equiv 1)\)
\[
\mathcal{H} = J \sum_j \mathbf{S}_{j,1} \cdot \mathbf{S}_{j,2} + \sum_j D_j \cdot \left( \mathbf{S}_{j,1} \times \mathbf{S}_{j,2} \right) \\
+ J_\parallel \sum_j \left( \mathbf{S}_{j,1} \cdot \mathbf{S}_{j+1,1} + \mathbf{S}_{j,2} \cdot \mathbf{S}_{j+1,2} \right) \\
+ J_\perp \sum_j \left( \mathbf{S}_{j,1} \cdot \mathbf{S}_{j+1,2} + \mathbf{S}_{j,2} \cdot \mathbf{S}_{j+1,1} \right) \\
- N \mu_B \mathbf{H} \cdot \mathbf{S},
\]
(1)
\[
\mathbf{S} = \frac{1}{N} \sum_j \left( \mathbf{S}_{j,1} + \mathbf{S}_{j,2} \right).
\]
(2)

This frustrated ladder has a magnetization plateau at \(1/2\) if \(J_\parallel\) and \(J_\perp\) are not too different (their ratio should be between \(1/3\) and \(3\) in the limit \(J_\parallel, J_\perp \ll J\) \([17]\), and it is amenable to the density matrix renormalization group method (DMRG). It is thus an ideal minimal model to study subtle effects related to plateaux in frustrated quantum magnets. We set \(J_\parallel \equiv 1\) throughout the paper and we will concentrate on the case \(J_\parallel = J_\perp\) for which the plateau is largest. We assume the \(g\)-tensor \(\mathbf{g}\) to be diagonal but to possess an asymmetry \(\delta g\) between the \(x\)-\(y\) and the \(z\) component. Due to the DM interaction, the SU(2) symmetry of the original Heisenberg model is not present and the DMRG calculations can be very demanding, restricting us to treat only rather small systems with up to 49 rungs.

To supplement the finite-size DMRG data, we have also performed a mean-field calculation which generalizes the ansatz used in ref. \([18]\) in order to deal with the tilted field. It is based on a product of rung wave functions,
\[
|\psi\rangle = \prod_j |\phi\rangle_j,
\]
(3)
\[
|\phi\rangle_j = a|S\rangle_j + b|T^{-1}\rangle_j + c|T_0\rangle_j + d|T_1\rangle_j,
\]
with the singlet and triplet eigenstates of the dimer on rung \(j\) given by \(|S\rangle_j = 1/\sqrt{2}(|\downarrow\uparrow\rangle_j - |\uparrow\downarrow\rangle_j|, \quad |T^{-1}\rangle_j = \frac{1}{\sqrt{2}}(|\downarrow\downarrow\rangle_j + |\uparrow\uparrow\rangle_j), \quad |T_0\rangle_j = \frac{1}{\sqrt{2}}(|\downarrow\uparrow\rangle_j + |\uparrow\downarrow\rangle_j), \quad |T_1\rangle_j = |\uparrow\downarrow\rangle_j\). For the case \(J_\parallel = J_\perp\) and \(D = 0\), this ansatz provides an exact solution [17], and it is expected to provide a good approximation for the physically relevant case \(D \ll J\). Comparison with the finite-system DMRG data shows good agreement, giving us confidence that the results presented are not affected by strong finite-size effects.

In the following we focus on the magnetic response of the system to an external magnetic field with arbitrary orientation. First we discuss our results in the presence of \(D\) only, and afterwards go to the more general case with an additional finite \(\delta g\). The symmetry analysis for a single dimer with DM interaction and \(\delta g = 0\) certifies that the results are independent of the absolute orientation of \(\mathbf{H}\) and \(\mathbf{D}\) as long as the angle between them is the same [19] for which reason we confine ourselves to treat only the case depicted in fig. 1. If not mentioned otherwise, we present results for \(D = 0.03\) and \(J_\parallel = J_\perp = 0.1\), i.e., we restrict ourselves to the case of a strongly frustrated ladder, which is relevant when having in mind the frustrated plateau system SrCu$_2$(BO$_3$)$_2$. We concentrate on the uniform magnetization, which is defined as
\[
\tilde{m}_u := -\frac{1}{N} \langle \mathcal{H} \rangle / \langle \partial \mathcal{H} \rangle = \mu_B g \langle \mathbf{S} \rangle.
\]
(4)

In general, for finite \(\delta g\) a magnetization perpendicular to \(\mathbf{H}\) is induced, causing a torque
\[
\tau = \tilde{m}_u \times \mathbf{H},
\]
(5)
which gives access to the uniform magnetization perpendicular to \(\mathbf{H}\) obtained as \(m_u^\parallel = \tau / H\). If \(D = 0\), in order to minimize the Zeeman-term in the Hamiltonian, the spins align in the direction of the vector \(\mathbf{H}\). For a better comparison of our results obtained at various angles and for different values of \(\delta g\), let us introduce the effective field
\[
\mathbf{H}^{\text{eff}} := \mu_B g \mathbf{H},
\]
(6)
In the following, when \(\delta g \neq 0\), we discuss the dependence \(\tilde{m}_u(H^{\text{eff}})\). If \(D = 0\) and \(\delta g \neq 0\), the components of the uniform magnetization parallel and perpendicular to \(\mathbf{H}\) are obtained as
\[
\tilde{m}_u^{\parallel} = |\langle \mathbf{S} \rangle| \mu_B \sqrt{g^2 + \cos^2 \alpha \delta g(2g + \delta g)} \left( \begin{array}{c} 0 \\ \sin \alpha \cos \alpha \end{array} \right),
\]
(7)
\[
\tilde{m}_u^{\perp} = |\langle \mathbf{S} \rangle| \mu_B \sin \alpha \cos \alpha \delta g(2g + \delta g) \left( \begin{array}{c} 0 \\ -\sin \alpha \cos \alpha \end{array} \right).
\]
(8)
Both quantities are proportional to \(|\langle \mathbf{S} \rangle|\) and hence \(m_u^{\parallel}\) and \(m_u^{\perp}\) have the same dependence on the magnitude of the field \(H^{\text{eff}}\), so that \(m_u^{\parallel}\) can be obtained by measuring the torque. For a better comparison of the results for different parameters, we rescale the magnetizations by their angular- and \(g\)-dependence, i.e., if \(\delta g \neq 0\) we analyze
\[
\tilde{m}_u^{\parallel, \text{rescaled}} = \frac{|\tilde{m}_u^{\parallel}|}{\sqrt{g^2 + \cos^2 \alpha \delta g(2g + \delta g)}},
\]
(9)
\[
\tilde{m}_u^{\perp, \text{rescaled}} = \frac{|\tilde{m}_u^{\perp}|}{\sin \alpha \cos \alpha \delta g(2g + \delta g)},
\]
(10)
as a function of
\[
H^{\text{eff}} = \mu_B \mathbf{H} \sqrt{g^2 + \cos^2 \alpha \delta g(2g + \delta g)}. \tag{11}
\]
Before we go to the general case with finite \(\delta g\) and finite \(D\), we first discuss the illustrative case of a system with DM interactions only. To do this, it is useful to recall the results obtained for an isolated dimer with \(D \neq 0\), \(\delta g = 0\) [19]. For perturbatively weak fields one finds
\[
\tilde{m}_u \sim \left( \mathbf{D} \times \mathbf{H} \right) \times \mathbf{D},
\]
(12)
leading, in general, to a magnetization perpendicular to $\vec{H}$. Note, however, that in this case it is not possible to determine $m_{\parallel}^u$ by measuring $\tau$, since $m_{\parallel}^u(H) \neq m_{\perp}^u(H)$ due to the lack of SU(2) symmetry. This is demonstrated in fig. 2 where we present DMRG results for the case $D = 0.03$ and $\delta g = 0$. As can be seen, the field dependences of $m_{\parallel}^u$ and $m_{\perp}^u$ differ fundamentally. In $m_{\parallel}^u$, a plateau is obtained which is smooth due to the DM interaction, while in $m_{\perp}^u$ pronounced peaks appear at the critical fields confining the plateau. This can be understood by considering the angular dependence of the torque $\tau(\alpha)$. As discussed in ref. [19] for the case of a single dimer, it changes drastically when leaving the limit of perturbatively small $H$ and going to the critical field $H_c$: one finds

$$\tau(\alpha) \sim \begin{cases} \sin \alpha \cos \alpha & \text{for } H < H_c, \\ \sin \alpha & \text{for } H = H_c. \end{cases}$$

This leads to a clear peak in $m_{\perp}^u$ at $H_c$. Note that in fig. 2(b) we present $m_{\perp}^u$ rescaled by $\sin \alpha \cos \alpha$, taking into account the angular dependence for $H < H_c$. As can be seen, the same dependence on $\alpha$ is found for all values of $H$ sufficiently away from the critical points. In fig. 3 we compare the angular dependence of $m_{\perp}^u$ at the critical point $\mu_B g H_{c,1} = 1$ and inside the plateau for $\mu_B g H = 1.1$.

In the latter case, the finite-size DMRG and the mean-field results are in excellent agreement with each other and with the dependence $\tau(\alpha) \sim \sin \alpha \cos \alpha$ and we see that finite-size effects play a minor role. At the critical field, however, we see that also for the ladder systems a completely different angular dependence is obtained. For small $\alpha$, it follows $\sin \alpha$, while for larger angles $m_{\perp}^u$ deviates to smaller values. Note that at the critical point the results of the DMRG and of the mean-field calculations show qualitatively similar behavior, but the mean-field value is smaller than the DMRG result. Comparison of DMRG results for 25 and 49 rungs shows that finite-size effects are minimal also in this case, so that we believe that these results should be representative for the thermodynamic limit. Despite the deviation from $\sin \alpha$, for large $\alpha$ the resulting $m_{\perp}^u$ at the critical field and inside the plateau differ by an order of magnitude, providing an explanation for the peaks visible in fig. 2(b).

In the following we test how a finite $\delta g$ changes the picture obtained for finite $D$ only. In fig. 4 we show our results for ladder systems when $\alpha = 10^\circ$ and $0.005 < \delta g < 0.2$, i.e. we go to a realistic value of $\delta g = 10\%$ while keeping $D = 0.03$. If $\delta g < D$, the peaks in $m_{\perp}^u$ at the critical fields are well visible. For $\delta g > D$, however, the peaks vanish. In this case, the magnetization curves resemble the SU(2) symmetric case up to a smoothening of the plateau, and we obtain $m_{\perp}^{u,\text{rescaled}}(H_{\text{eff}}) \approx m_{\perp}^{u,\text{rescaled}}(H_{\text{eff}}^\text{eff})$.

The remarkable angular dependence of the torque obtained for systems with $\delta g = 0$ has also very important consequences in the presence of $g$-tensor anisotropy. If we now keep realistic values of both anisotropies $\delta g = 10\%$ and $D = 0.03$ and increase $\alpha$, for angles $\alpha > 80^\circ$ pronounced peaks in $m_{\perp}^u$ at the critical fields become visible again, as can be seen in fig. 5. We find that $m_{\perp}^{u,\text{rescaled}}(H_{\text{eff}}) \neq m_{\perp}^{u,\text{rescaled}}(H_{\text{eff}}^\text{eff})$, as in the case with DM anisotropy only. This shows that care has to be taken when interpreting results of torque measurements.
Fig. 4: (Colour on-line) Rescaled uniform magnetization as obtained by eqs. (9) and (10) parallel (a) and perpendicular (b) to $\vec{H}$ for $D = 0.03$, $\alpha = 1^\circ$, and $0.005 \leq \delta g \leq 0.2$. The solid lines are the results of the mean-field calculation, the points the results of DMRG calculations on 49 rungs. The black line shows the exact magnetization parallel to $\vec{H}$ when $D = \delta g = 0$. Note that in (a) the mean-field results lie on top of each other.

and that, in general, it is not possible to safely conclude on the field dependence of $m_u^\parallel$ by considering the field dependence of the torque.

In conclusion, by investigating the interplay of DM and $g$-tensor anisotropies on the magnetic response of a strongly frustrated ladder, we have clarified in which cases the torque can be used as a good approximation to the magnetization when DM is present. For this to be the case, two conditions must be fulfilled: i) The ratio of the DM interaction to the exchange coupling $D/J$ should be smaller than (or at most comparable to) the $g$-tensor anisotropy; ii) The angle between the magnetic field and the DM vector should not be too small. While the first condition is often fulfilled (the DM interaction is typically a few percent of the exchange while the $g$-tensor anisotropy is about 10% for Cu$^{2+}$), the second condition may or may not be satisfied depending on the experimental conditions. Interestingly enough, the fact that the field dependence of the torque is quite different from that of the magnetization when the field is almost parallel to the DM vector could be an advantage. Indeed, in this geometry we predict the torque to have well pronounced peaks at the critical fields that delimitate the plateau. This effect could thus be used to locate with a high precision the critical fields, a difficult task if only magnetization data are available since DM interactions can lead to a significant rounding at the boundaries of the plateaux. In view of the controversies regarding the plateau structure of SrCu$_2$(BO$_3$)$_2$, it would thus be particularly interesting to perform torque measurements with the field perpendicular to the layers and parallel (or almost parallel) to the intra-dimer DM vector of one type of dimers, and it is our hope that the present paper will encourage such an investigation.

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