Spin, charge and lattice dynamics of magnetization processes in frustrated Shastry-Sutherland system TmB_4

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TmB_4 - the metallic tetraboride with a geometrically frustrated Shastry-Sutherland lattice (SSL) has attracted recently a lot of attention. It exhibits a strong Ising-like anisotropy along the *c*-axis and a rather complex phase diagram of the ordered magnetic phase below $T_N \approx 11.7$ K in which besides the main magnetization (M) plateau also small fractional plateaus can be observed. To learn more about the properties of fractional plateaus we have investigated the dynamics of magnetization processes in TmB_4 by M vs. H (applied magnetic field) measurements at different temperatures and different field sweep rates (*swr*) 1.8 K. In addition, experiments of the charge carrier mobility and magnetostriction as a function of temperature and magnetic field were carried out to specify the interplay between magnetic structure and the charge dynamics, and the role of the magneto-elastic interaction in this compound, respectively. It was shown that the magnetization values of fractional plateaus become elevated with the increase of temperature at which magnetization takes place and with the decrease of magnetic field sweep rate, which points to inconsistency with theoretical predictions based on Glauber dynamics. Moreover, it turned out that the transitions between magnetic phases during magnetization processes are accompanied by an anomalous behavior of charge mobility and distinctive variations of magnetostriction.

Introduction

Geometrically frustrated magnetic systems provide a rather fertile ground for the observation of new states of matter. The interplay between competing interactions which cannot be simultaneously optimized, the applied magnetic field, and often also enhanced quantum fluctuations due to low dimensionality result into a variety of phases with properties that are not found in non-frustrated systems. Typical examples of such phases are e.g. spin-liquid phases on triangular and Kagome lattices, spin-ice structures on pyrochlore lattices, and various magnetization plateaus on the two-dimensional frustrated Shastry-Sutherland lattice (SSL) [1, 2]. As regards the case of SSL, magnetization plateaus observed in $SrCu_2(BO_3)_2$ have attracted a considerable interest due to their similarity to quantum Hall effect phenomena [3-6].

Recently, rare-earth tetraborides RB_4 (R = Tm, Er, Ho, Dy, Tb) have been identified to belong to the SSL family of quantum magnets. Even if they share the same frustrated magnetic lattice, the phase diagrams of the RB_4 magnetic compounds show also behaviors distinct from that of $SrCu_2(BO_3)_2$. In insulating $SrCu_2(BO_3)_2$ the exchange interaction is of the Heisenberg type, but on the other hand, the RB_4 magnets are metallic and the exchange interaction between their magnetic moments is of the long range Ruderman-Kittel-Kasuya-Yosida (RKKY) type mediated by conduction electrons [7]. Probably the most investigated among RB_4 compounds, thulium tetraboride TmB_4, which orders antiferromagnetically at T_N = 11.7 K, has attracted attention for its rich phase diagram [8-11] which is strongly biased by crystal field effects at Tm^{3+} ion sites that lift the degeneracy of the J = 6 multiplet and lead to a M_J = ±6 ground state doublet [9, 12]. TmB_4 thus exhibits strong Ising anisotropy in which the saturation in external field H_{sat} along the *c* axis is at least 10 times smaller than this in the perpendicular *a-b* plane. At temperatures below T_{N2} = 9.7 K, where another antiferromagnetic (Néel) phase becomes stable, depending on applied field H the magnetization M exhibits various plateaus: a wide main plateau M/M_{sat} = 1/2 in fields above about 18 kOe, and narrow hysteretic fractional plateaus at M/M_{sat} = 1/11, 1/9, 1/8 and 1/7 in fields between about 13 kOe and 18 kOe [9, 10, 13]. Very recently in [14] the 1/8 fractional plateau phase of TmB_4 has been viewed as an example of emergence of an Archimedean lattice. As theoretical approaches on SSL in the Ising approximation cannot provide the M/M_{sat} = 1/2 plateau, several theoretical groups have shown that the existence of such a plateau may be obtained by considering longer-range interactions [14-17]. But, the nature of fractional plateaus has still not been satisfactory elucidated [14-17]. Further details about the magnetic structure and other properties of TmB_4, and related magnetic tetraborides [18-22], as well as about the current theoretical approaches can be found elsewhere [23-27].

As TmB_4 belongs to frustrated spin systems in which the interplay between competing interactions cannot be simultaneously optimized, one can expect that it may be trapped in meta-stable states from which it is at low temperatures hard to relax into the equilibrium ground state. One may therefore question if non-equilibrium magnetization dynamics could influence the properties of fractional magnetization plateaus. A detailed discussion of this question has been performed in [28]. In order to clear this point, authors of [28] studied the magnetization dynamics of a classical Ising model on the SSL with additional long-range interactions using the Glauber dynamics which has been successfully used to study magnetic properties of the triangular spin-chain compound $Ca_3Co_2O_6$ [29]. This algorithm allowed them to investigate the dependence of magnetization curves as a function of temperature and magnetic field sweep rate. It was shown that the experimental magnetization plateaus at fractional values of M/M_{sat} = 1/7, 1/9, and 1/11 followed by the main magnetization plateau at M/M_{sat} = 1/2 could be reproduced at low temperatures for certain magnetic field sweep rates. In addition, the hysteresis loop observed in [9, 30] could also be qualitatively explained using this simulation. It was also indicated that the formation of domain walls due to non-equilibrium magnetization processes might be responsible for the emergence of fractional plateaus.

To verify these predictions for TmB_4, especially the dependence of magnetization curves as a function of temperature and magnetic field sweep rate, we have investigated the dynamic magnetization process by M vs. H measurements at different field sweep rates *swr* (*swr* equal to 200 Oe/s, 20 Oe/s and 2 Oe/s) at temperatures between 1.8 K and 4.2 K. Moreover, experimental studies of the field dependence of charge carrier mobility and of magnetostriction as a function of temperature and magnetic field were carried out to get new information about charge and lattice dynamics in various magnetic phases, respectively.

Experimental

The used TmB_4 single crystals were grown by an inductive, crucible-free zone melting method. Samples were cut from the same batch and their residual resistivity ratio was larger than 100, documenting their high quality. $M(H)$ magnetization measurements were performed using a Quantum Design MPMS-3 system. A four probe method with a rotating sample was used to measure the Hall-effect at low temperatures in magnetic fields up to 80 kOe [31]. Dilatometry measurements were carried out using a capacitance dilatometer [32] with a resolution of $\Delta l/l \approx 10^{-9}$.

Results and discussion

The spin dynamics of TmB_4 was investigated by performing $M(H)$ magnetization measurements at different temperatures below T_{N2} (see Fig. 1) and at different magnetic field sweep rates *swr* = 2, 20 and 200 Oe/s (see Fig. 2). Fig. 1 illustrates the field dependence of magnetization for $H \parallel c$ in decreasing magnetic field. In this case was the sample first heated up to 30 K, then cooled down in zero field to a certain temperature (4 K, 3 K or 2 K), and subsequently magnetized (up to 70 kOe) and demagnetized with a sweep rate of *swr* = 200 Oe/s. It can be seen that the value of the observed fractional plateau (its M/M_{sat} value) depends on temperature at which the zero field cooled sample was magnetized and demagnetized. With decreasing temperature the M/M_{sat} value of the fractional plateau decreases (e. g. at 3 K M/M_{sat} = 1/7 and at 2 K M/M_{sat} = 1/9). These results are inconsistent with predictions which follow from ref. 28 where for the same sweep rate an increase of M/M_{sat} with decreasing temperature of magnetization is expected. Fig. 2 shows the evolution of the fractional plateau M/M_{sat} = 1/9 as a dependence of various magnetization (and demagnetization) sweep rates. In this case again, the sample was first heated up to 30 K and then zero-field cooled to 1.8 K with a sweep rate of 200 Os/s. Subsequently the sample was magnetized up to 70 kOe and demagnetized back to zero field with a certain sweep rate (*swr* = 200, 20, 2 or 1 Oe/s). The obtained results show interesting dependencies. On the virgin magnetization curve at highest sweep rate (200 Oe/s) no fractional plateau develops, it appears only on the demagnetization curve. However, with decreasing sweep rates (*swr* = 20, 2, 1 Oe/s) fractional plateaus appear both on magnetization and demagnetization dependencies. Moreover, with decreasing sweep rate the hysteresis between magnetization and demagnetization curves is shrinking. Notice also that during demagnetization all M/M_{sat} *vs. H* dependencies remain almost the same which may be related to the fact that fractional plateaus lie directly below the main plateau phase with a M/M_{sat} = ½ magnetization. All this shows a much richer behavior as proposed in [28]. Thus, as a result, not a full agreement between obtained experimental results and the theoretical prediction based on Glauber dynamics has been observed regarding the magnetization processes in TmB_4 at different temperatures and at different magnetic field sweep rates. This difference may be associated with the existence and properties of domain walls which were in [28] suggested to be responsible for the emergence of fractional plateaus.

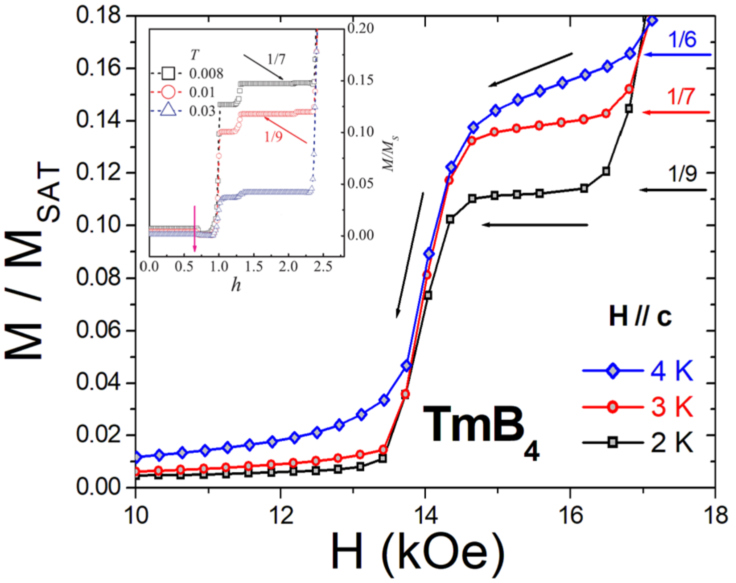

Fig. 1: Field dependences of demagnetization curves at various temperatures for $H \parallel c$ (virgin curves from 0 to 70 kOe are not displayed for clarity). In this case the sweep rate for all dependencies was 200 Oe/s. In the inset the proposed field dependencies of fractional plateaus at different temperatures according to [28] are shown. It can be seen e.g. that the 1/7 plateau is expected at lower temperature than the 1/9 one, on the contrary to obtained experimental results.

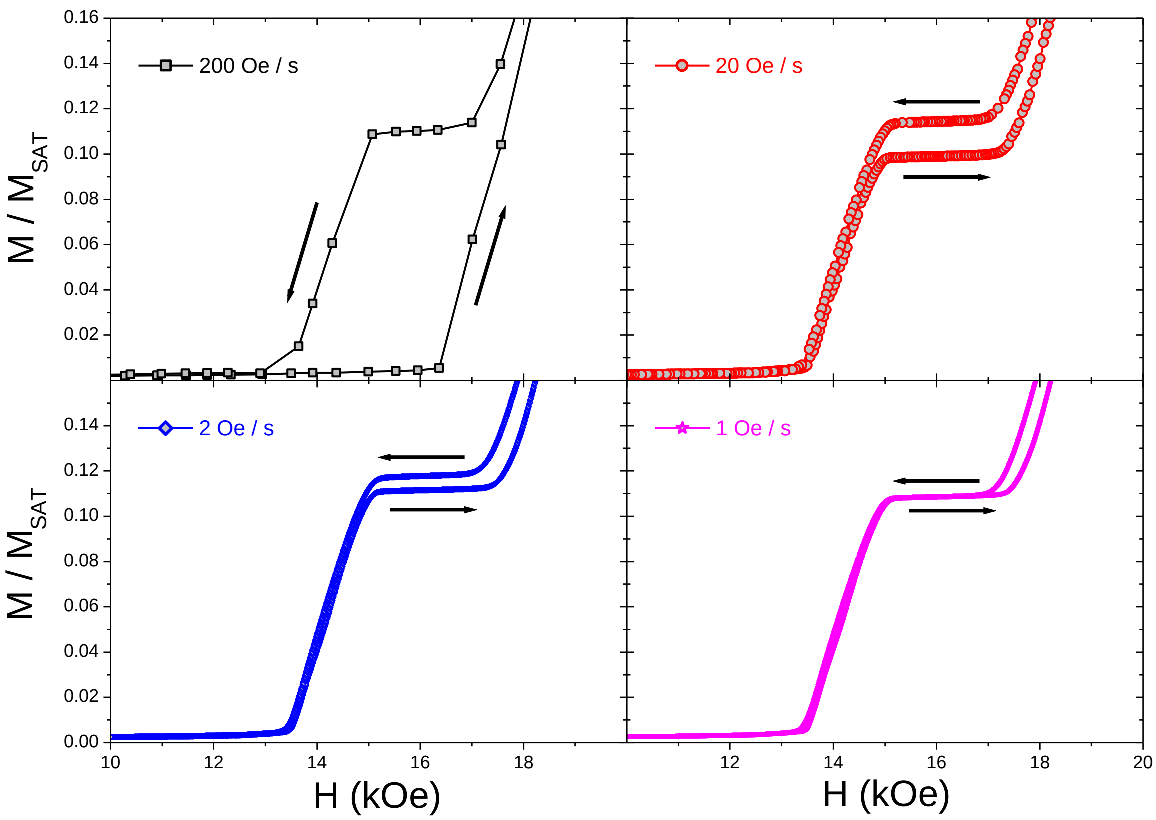

Fig. 2: Field dependences of magnetization curves at 1.8 K at various magnetization sweep rates.

In Fig. 3 the field dependence of the Hall coefficient R_H and the corresponding charge mobility μ_H at various temperatures as features of charge dynamics during magnetization processes are displayed. At temperatures well below T_N (at 1.85 K and 3.2 K) sharp kinks on $R_H(H)$ and $\mu_H(H)$ dependencies can be observed with increasing field as boundaries between different magnetic phases of TmB_4 are being crossed (gradually between the antiferromagnetic Néel phase and the fractional plateau phase at ~14 kOe, between the fractional plateau and the half plateau phase at ~20 kOe, and between the main plateau phase and the magnetically saturated state at ~38 kOe). These results correspond to those obtained in [13, 19] and point to significant presence of the intrinsic anomalous Hall-effect in the ordered phase, which depends on M [13, 33] (due to diverse magnetic arrangements below T_N). Not fully understood appears to be the region above the magnetization saturation, where between ~38 kOe and ~44 kOe a very sharp increase of R_H and μ_H is observed before both dependencies reach the monotonous high field behaviour. From Hall coefficient relation $R_H = -1/nq$, where n denotes the concentration and q the charge of carriers, the concentration of conduction electrons in TmB_4 was estimated to be $n \approx 2.5 \times 10^{21}$ cm^{-3}.

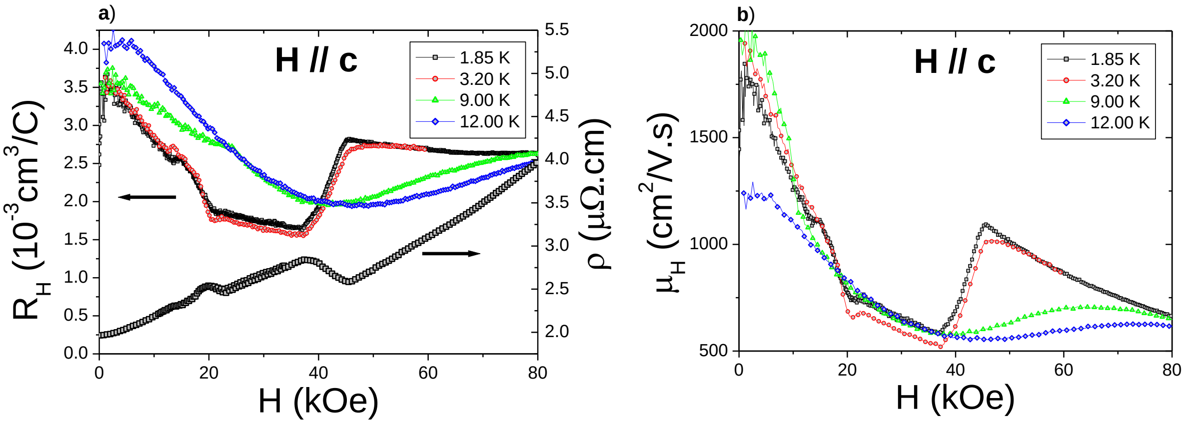

Fig. 3: Field dependence of the Hall coefficient $R_H(H)$ (a) and of the charge mobility $\mu_H(H)$ (b) as a function of magnetic field at different temperatures. The solid squares in (a) represent the longitudinal resistivity $\rho(H)$ field dependence at 1.85 K.

The features of lattice dynamics in magnetisation processes were investigated through magnetostriction measurements. The obtained results of magnetostriction / dilatation ($\Delta l/l$) along the c-axis for $H \parallel c$ are displayed in Fig. 4. They exhibit a strong dependence of $\Delta l/l$ on magnetic field (Fig. 4b) and suggest thus a rather large magneto-elastic interaction (interplay between spin and lattice degrees of freedom) in TmB_4. We assume that this interaction could in magnetically strongly anisotropic TmB_4 cause also lattice distortions which depend on field orientation (i.e. various distortions should be induced for $H \parallel c$ and for $H \perp c$). But, the confirmation of these considerations requires further investigation. Moreover, it is surprising that a hysteresis of magnetostriction as a function of magnetic field was observed also in the magnetically saturated phase above ~38 kOe (Fig. 4b).

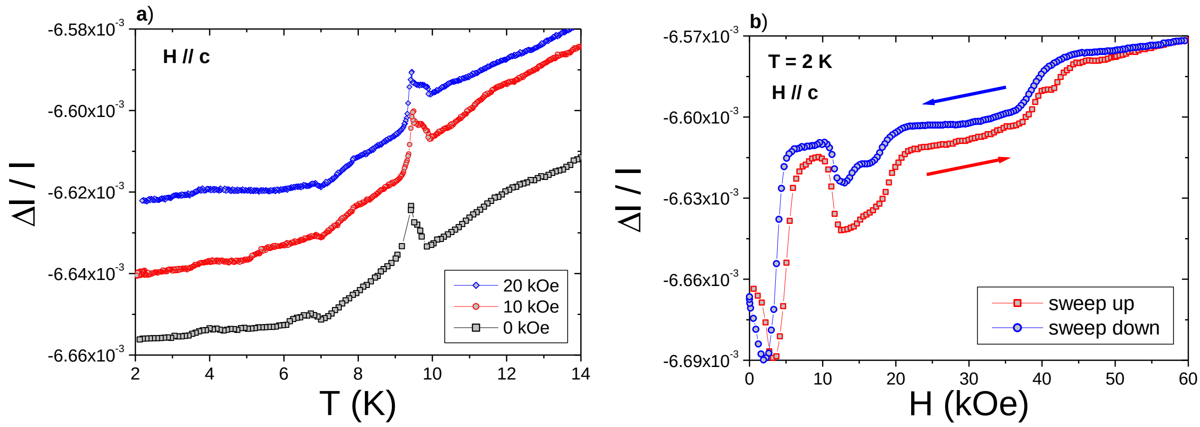

Fig. 4: Temperature dependences of dilatation *Δl/l* in different magnetic fields (a) and the field dependence of *Δl/l* at 2 K (b).

Conclusion

Our results have shown that the magnetization of fractional plateaus becomes elevated with the increase of temperature and that the behavior of magnetization curves depends considerably on magnetic field sweep rates. These results point to inconsistency with theoretical predictions based on Glauber spin dynamics. Open questions remain also in connection with the charge and lattice dynamics in TmB_4 which below and above T_N exhibit anomalous behavior as a function of applied magnetic field.

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