OFFPRINT

Dynamic magnetization process in the frustrated Shastry-Sutherland system TmB$_4$


EPL, 102 (2013) 37005

Please visit the new website
www.epljournal.org
The Editorial Board invites you to submit your letters to EPL

EPL is a leading international journal publishing original, high-quality Letters in all areas of physics, ranging from condensed matter topics and interdisciplinary research to astrophysics, geophysics, plasma and fusion sciences, including those with application potential.

The high profile of the journal combined with the excellent scientific quality of the articles continue to ensure EPL is an essential resource for its worldwide audience. EPL offers authors global visibility and a great opportunity to share their work with others across the whole of the physics community.

Run by active scientists, for scientists

EPL is reviewed by scientists for scientists, to serve and support the international scientific community. The Editorial Board is a team of active research scientists with an expert understanding of the needs of both authors and researchers.
Six good reasons to publish with EPL

We want to work with you to help gain recognition for your high-quality work through worldwide visibility and high citations.

1. **Quality** – The 40+ Co-Editors, who are experts in their fields, oversee the entire peer-review process, from selection of the referees to making all final acceptance decisions.

2. **Impact Factor** – The 2010 Impact Factor is 2.753; your work will be in the right place to be cited by your peers.

3. **Speed of processing** – We aim to provide you with a quick and efficient service; the median time from acceptance to online publication is 30 days.

4. **High visibility** – All articles are free to read for 30 days from online publication date.

5. **International reach** – Over 2,000 institutions have access to EPL, enabling your work to be read by your peers in 100 countries.

6. **Open Access** – Articles are offered open access for a one-off author payment.

Details on preparing, submitting and tracking the progress of your manuscript from submission to acceptance are available on the EPL submission website www.epletters.net.

If you would like further information about our author service or EPL in general, please visit www.epljournal.org or e-mail us at info@epljournal.org.

---

**EPL is published in partnership with:**

- European Physical Society
- Società Italiana di Fisica
- EDP Sciences
- IOP Publishing

---

“We’ve had a very positive experience with EPL, and not only on this occasion. The fact that one can identify an appropriate editor, and the editor is an active scientist in the field, makes a huge difference.”

Dr. Ivar Martin
Los Alamos National Laboratory, USA
Visit the EPL website to read the latest articles published in cutting-edge fields of research from across the whole of physics.

Each compilation is led by its own Co-Editor, who is a leading scientist in that field, and who is responsible for overseeing the review process, selecting referees and making publication decisions for every manuscript.

- Graphene
- Liquid Crystals
- High Transition Temperature Superconductors
- Quantum Information Processing & Communication
- Biological & Soft Matter Physics
- Atomic, Molecular & Optical Physics
- Bose–Einstein Condensates & Ultracold Gases
- Metamaterials, Nanostructures & Magnetic Materials
- Mathematical Methods
- Physics of Gases, Plasmas & Electric Fields
- High Energy Nuclear Physics

If you are working on research in any of these areas, the Co-Editors would be delighted to receive your submission. Articles should be submitted via the automated manuscript system at www.epletters.net

If you would like further information about our author service or EPL in general, please visit www.epljournal.org or e-mail us at info@epljournal.org

Image: Ornamental multiplication of space-time figures of temperature transformation rules (adapted from T. S. Bíró and P. Ván 2010 EPL 89 30001; artistic impression by Frédérique Swist).
Dynamic magnetization process in the frustrated Shastry-Sutherland system TmB$_4$

W. C. Huang$^1$, L. Huo$^1$, J. J. Feng$^1$, Z. B. Yan$^2$, X. T. Jia$^3$, X. S. Gao$^1$, M. H. Qin$^{1(\text{a})}$ and J.-M. Liu$^{2(\text{b})}$

$^1$Institute for Advanced Materials, South China Academy of Advanced Photonics Engineering, South China Normal University - Guangzhou 510006, China
$^2$Laboratory of Solid State Microstructures, Nanjing University - Nanjing 210093, China
$^3$School of Physics and Chemistry, Henan Polytechnic University - Jiaozuo 454000, China

received 4 February 2013; accepted in final form 18 April 2013
published online 15 May 2013

PACS 75.10.Hk – Classical spin models
PACS 07.55.Db – Generation of magnetic fields; magnets
PACS 71.10.Hf – Non-Fermi-liquid ground states, electron phase diagrams and phase transitions in model systems

Abstract – The dynamic magnetization behaviors of the classical Ising model on the Shastry-Sutherland lattice with additional long-range interactions are investigated by means of the Glauber dynamics, in order to understand the fascinating magnetization plateaus and the hysteresis loop observed in TmB$_4$. With this algorithm, the experimental $1/n$ ($n = 7, 9, 11$) magnetization plateaus as well as the main $1/2$ one can be reproduced at low temperatures. Furthermore, the hysteresis loop can also be well explained by the present theory. It is indicated that the formation of domain walls due to the non-equilibrium magnetization process may be responsible for the emergence of the fractional plateaus.

Copyright © EPLA, 2013

Introduction. – Geometrically frustrated spin systems which exhibit very rich magnetic properties have drawn considerable attentions during the last several decades [1]. For example, various experimental and theoretical explorations have been devoted to the emergence of multi-step magnetization ($M$) curves in frustrated systems such as the triangular spin-chain system Ca$_3$Co$_2$O$_6$ [2–5] and the Shastry-Sutherland (S-S) magnets [6–11]. So far, the step-like magnetization curves observed in Ca$_3$Co$_2$O$_6$ are generally believed to be caused by the non-equilibrium magnetization dynamics [12,13], while those in S-S systems are far from being fully understood and remains to be checked.

The S-S lattice [14] has attracted special attentions since its experimental realization in the compound SrCu$_2$(BO$_3$)$_2$ in which a fascinating sequence of magnetization plateaus at fractional values of the saturated magnetization ($M_S$) have been reported [15,16]. Most recently, similar magnetic behaviors have been identified in rare-earth tetraborides RB$_4$ ($R =$ Tb, Dy, Ho, Tm, etc.) with the rare-earth moments located on a lattice which is topologically equivalent to the S-S lattice [8,9,17–20]. For instance, the fractional magnetization plateaus at $M/M_S = 1/2, 1/3, 1/9$ and $1/11, \ldots$ have been experimentally observed at temperature ($T$) below 4 K in TmB$_4$ [9]. In contrast to SrCu$_2$(BO$_3$)$_2$ with Cu$^{2+}$ ions carrying a quantum spin $1/2$, TmB$_4$ presents a large total magnetic moment ($\sim 6.0 \mu_B$) and can be considered as a classical spin system, triggering an extensive theoretical investigation of classical spin models on the S-S lattice [21–28].

It is experimentally indicated that TmB$_4$ is of strong easy-axis anisotropy caused by crystal field effects. Based on this point, the magnetization process of the classical Ising model on the S-S lattice was investigated using the tensor renormalization group approach, and a single magnetization plateau at $M/M_S = 1/3$ was predicted at low $T$ for certain coupling constants [21]. In fact, the ground states of the Ising model on the S-S lattice were investigated most recently and the existence of a single $1/3$ plateau was rigorously proved [22,23]. The effect of further-neighbor interactions was suggested to eventually explain the magnetization plateaus in TmB$_4$, and three different ground states with $M/M_S = 1/2$ were recognized when the additional third-neighbor interaction was considered. On the other hand, the quantum spin-$1/2$
Ising-like XXZ model with additional interactions on the S-S lattice was studied using the quantum Monte Carlo method, and the plateau at $M/M_S = 1/2$ was identified [29–31]. It was believed that the emergence of the $M/M_S = 1/2$ plateau may be due to the quantum fluctuations and long-range interactions. In our earlier work, the presence of the $M/M_S = 1/2$ plateau was also confirmed when the additional long-range interactions were taken into account in the classical spin model [32].

So far, the main magnetization plateau at $M/M_S = 1/2$ and some of other small fractional plateaus in TmB$_4$ can be reproduced by the consideration of the long-range interactions for certain coupling constants. However, the origins of other small fractional plateaus are still under debate. Generally speaking, a frustrated spin system can be easily trapped into metastable states at low $T$ and is hard to relax to the equilibrium state. The time available experimentally may not be sufficient for the spin rearrangement, even though the energy difference between plateaus may be very small. Thus, it is reasonable to assume that the spins in TmB$_4$ are easily trapped into a metastable state rather than into the equilibrium one at low $T$, which, to some extent, can be also favored by the obvious hysteresis loop observed in earlier experiments. Furthermore, the formation and the motion of domain walls due to the non-equilibrium magnetization process may play an important role in the emergence of the magnetization plateaus, which has been verified in the study of the triangular spin-chain compound Ca$_3$Co$_2$O$_6$ [13,33,34].

As a matter of fact, the $M/M_S = 1/8$ plateau has been predicted in the XXZ model on the S-S lattice for a short relaxation time, indicating that this plateau may arise from non-equilibrium state [31]. Thus, one may question if the non-equilibrium magnetization dynamics is also crucial for the understanding of the fascinating magnetic fluctuations and long-range interactions in TmB$_4$. A detailed discussion of this question is definitely necessary for S-S magnets and other similar frustrated spin systems.

The remainder of this paper is organized as follows: In the second section, the model and the simulation method will be presented and described. The third section is attributed to the simulation results and discussion. At last, the conclusion is presented in the fourth section.

Model and method. – In the presence of the long-range interactions and $h$, the Hamiltonian can be described as follows:

$$H = J_1 \sum_{\langle i,j \rangle_1} S_i \cdot S_j + J_2 \sum_{\langle i,j \rangle_2} S_i \cdot S_j + J_3 \sum_{\langle i,j \rangle_3} S_i \cdot S_j + J_4 \sum_{\langle i,j \rangle_4} S_i \cdot S_j - h \sum_i S_i^z,$$

(1)

where $\langle i,j \rangle_1, \langle i,j \rangle_2, \langle i,j \rangle_3,$ and $\langle i,j \rangle_4$ denote the summations over all pairs on the bonds with $J_1$, $J_2$, $J_3$ and $J_4$ couplings, respectively, as shown in fig. 1. $S_i$ represents the Ising spin with unit length on site $i$, $h$ is applied along the $+z$ axis and $S_i^z$ denotes the $z$ component of $S_i$. $J_1 = 1$ is the antiferromagnetic (AFM) coupling, the coupling ratio $J_2/J_1 = 1$ is expected from the crystal structure of TmB$_4$, similar with earlier estimation [30,31], the AFM $J_3 = 0.15J_1$ and the ferromagnetic (FM) $J_4 = -0.15J_1$ are estimated to qualitatively reproduce the experimental results. To investigate the magnetization dynamics of the spin system, the simulation is carried out by a single spin-flip rate in the Glauber form [13,34]. The spins are assumed to interact not only with the neighbors and external magnetic field but also with a heat reservoir, based on the Glauber theory [35]. The probability of a spin flip of the $i$-th spin per Monte Carlo step (MCS) can be described as

$$W_i = \frac{\alpha}{2} \left[ 1 - S_i \tanh \left( \frac{D}{k_B T} + \frac{\mu h}{k_B T} \right) \right],$$

(2)
Dynamic magnetization process in the frustrated Shastry-Sutherland system \textit{TmB$_4$}

Fig. 2: (Color online) Magnetization curves for (a) different magnetic-field sweep rates at $T = 0.01$, and (b) enlargements of magnetization curves at different temperatures at a fixed sweep rate of 1/60 MCs$^{-1}$. 

with

$$D = J_1 \sum_{\langle i,j \rangle_2} S_j + J_2 \sum_{\langle i,j \rangle_2} S_j + J_3 \sum_{\langle i,j \rangle_3} S_j + J_4 \sum_{\langle i,j \rangle_4} S_j,$$

where $\alpha = 0.5$ MCs$^{-1}$ is the constant of the interaction of a spin with the heat reservoir, $k_B = 1$ is the Boltzmann constant, $\mu = 1$ is the magnetic moment of the Tm ion. In earlier work, the Glauber-type form of the spin-flip probability has been discussed in detail and successfully used in the frustrated spin-chain system $\text{Cu}_3\text{Co}_2\text{O}_6$ [13,34]. Similarly, a reasonable value $\alpha = 0.5$ MCs$^{-1}$ is used in our simulation to qualitatively coincide with the experimental results.

Our simulation is performed on an $L \times L$ ($L = 96$ is chosen unless stated otherwise) lattice with periodic boundary conditions. The simulation is started from the saturated magnetization state under high $h$, which is in the best accordance with the real case. Then the magnetization curves in the decreasing $h$ at different sweep rates for various temperatures are studied to investigate the magnetization dynamics in detail. In addition, the sweep rate is defined by $1/\text{m MCs}^{-1}$ which means that the $1/\text{m}$ of $h$ unit is updated per MCs.

**Simulation results and discussion.** Figure 2(a) shows the calculated $M/M_S$ as a function of $h$ for different magnetic-field sweep rates at a low temperature $T = 0.01$. Three steps can be observed for $h = 0$ for different sweep rates of 1/6000 MCs$^{-1}$. When $h$ decreases down to $\sim 4.75$, $M$ reaches the plateau at $M/M_S = 1/2$, and then falls down to the step at $M = 0$ below $h \sim 2.2$. More interestingly, the step at $M = 0$ decomposes into two substeps (zero and nonzero) separated at $h \sim 1$ when the magnetic-field sweep rate is increased. The height of the nonzero substep increases with increasing sweep rate. When the magnetic-field sweep rate increases up to 1/60 MCs$^{-1}$, the $M/M_S = 1/9$ plateau reported in experiments can be well reproduced in addition to the major magnetization plateaus at $M = 0$ and $M/M_S = 1/2$. Furthermore, the dynamic magnetization curves in response to $T$ at the constant sweep rate of $1/60$ MCs$^{-1}$ are also investigated, and the simulated results are presented in fig. 2(b). It is clearly demonstrated that the nonzero substep is heightened as $T$ decreases. When $T$ falls down to 0.008, a magnetization step at $M/M_S = 1/7$ is observed at intermediate $h$ range, which is consistent with experimental observation [9]. In addition, some additional narrow plateaus can also be noticeable in our simulation, which deserves to be checked in further experiments.

As stated earlier [32], the ferrimagnetic (FI) state spin arrangement consisting of alternative AFM and FM stripes is more favored than the FM state when $h$ is decreased down to the first critical field, while the Neel state is likely stabilized below the following critical field. For the extremely slow sweep rate (1/60000 MCs$^{-1}$), the single-domain FI state with the plateau at $M/M_S = 1/2$ and the Neel state with the $M = 0$ plateau are, respectively, stabilized below these two critical fields, leading to the three-step magnetization curve which is similar to that obtained by the Monte Carlo simulation. To uncover the origin of the nonzero substep, the specimens of configurations for various plateaus under different $h$ at the magnetic-field sweep rate of 1/60 MCs$^{-1}$ are presented in fig. 3. It is clearly shown that the Neel state grows at a lot of nucleation centers when $h$ falls down to the second critical field, resulting in the domain formation. A mixed state with the Neel order and domain walls constructed of polarized spin chains is responsible for the emergence of the $M/M_S = 1/9$ plateau in the magnetization curve, as shown in fig. 3(c). The domains become smaller with increasing magnetic-field sweep rate, leading to the heightening of the nonzero substep. When $h$ is further decreased, the domain walls almost disappear (fig. 3(d)), and the magnetization plateau at $M = 0$ can be observed. On the other hand, the domain boundary mobility may
be greatly decreased as $T$ decreases. Thus, the domains become smaller with decreasing $T$, making the additional magnetization steps more apparent, as confirmed in our simulation (fig. 2(b)). It is noted that the temperature at which the experimental $M/M_S = 1/7$ plateau is observed is higher than that of the $M/M_S = 1/9$ one. The inconsistency between the present theory and the experiment may be due to the fact that the disorder effect caused by the inhomogeneity in realistic materials is completely ignored in our simulation. However, our work clearly indicates that the non-equilibrium magnetization dynamics may play an important role in the appearance of the fractional magnetization steps in TmB$_4$.

In addition, the hysteresis loop is also studied in our work, and the results can qualitatively reproduce the experimental observations [9,19]. Figure 4(a) shows the hysteresis loop at the magnetic-field sweep rate of 1/60 M Cs$^{-1}$ at $T = 0.01$. The nonzero substep emerges in both field-decreasing and field-increasing branches of the magnetization curve. The nonzero substep ($M/M_S = 1/9$) in the field-decreasing branch is much higher than that ($M/M_S = 1/11$) in the field-increasing branch, in a good agreement with the experimental report [9]. This phenomenon demonstrates that the domain structures can be strongly affected by the initial state at a high magnetic-field sweep rate. In fact, the spin configurations of the simulation reveal that the Neel domains for the nonzero substep in the field-increasing branch are generally larger than those in the field-decreasing branch, resulting in the emergence of the hysteresis loop. More interestingly, it is confirmed in our simulation that the value of plateau magnetization varies between different runs, similar with earlier experimental report [9]. The corresponding results are not shown here for brevity. Furthermore, it has been noticed in earlier experiment that the critical fields in the field-decreasing branch of the magnetization curve are respectively smaller than those in the field-increasing branch [19]. This magnetic behavior can also be well reproduced by the simulation.

As stated earlier, the time required for the spin rearrangement likely exceeds the time available experimentally. Thus, the non-equilibrium magnetization dynamics may be essential for the emergence of the fractional magnetization plateaus in TmB$_4$. This point has been confirmed in this work in which the non-equilibrium evolution is performed by means of the Glauber dynamics. The fractional magnetization plateaus and the hysteresis loop at low $T$ reported in experiments can be reproduced in our simulation. Thus, our work may provide a new insight into the study of the magnetization process of TmB$_4$, although not all the experimental results can be excellently explained based on the present theory.

At last, the dependence of the step-like magnetization feature on the lattice size $L$ has been investigated in order to exclude the artificial facts caused by the finite lattice size. Figure 4(b) shows the simulated magnetization curves for different $L$ ($L = 60, 96, 120$ and $150$) at $T = 0.01$ for the extremely fast magnetic-field sweep rate of 1/60 M Cs$^{-1}$. All the simulated curves for various $L$ are almost merged into one, indicating that the finite-size effect on the magnetization of the system is almost negligible and never affects our conclusion.

Conclusion. – In summary, we have investigated the classical Shastry-Sutherland Ising model with long-range interactions employing a Glauber-type form of the spin-flip probability in order to understand the dynamic magnetization process in TmB$_4$. Besides the main $M/M_S = 1/2$ plateau, other fractional magnetization plateaus at $M/M_S = 1/n$ ($n = 7, 9, 11$) observed in experiments can be reproduced in our simulation of the model at low temperatures for certain magnetic-field sweep rates. In addition, the hysteresis loop can be also well explained in the present theory. It is indicated that the magnetization dynamics may be essential for the emergence of those fractional magnetization plateaus. Thus, the present work may provide a new insight into the understanding of the magnetization process for frustrated S-S magnets and other similar frustrated spin systems.

***

This work was supported by the Natural Science Foundation of China (11204091, 11274094, 11234005), the National Key Projects for Basic Research of China (2011CB922101), China Postdoctoral Science Foundation funded project (2012T50684, 20100480768, 2011M500088), and the Priority Academic Program Development of Jiangsu Higher Education Institutions, China.

REFERENCES

Dynamic magnetization process in the frustrated Shastry-Sutherland system TmB₄