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The main 1/2 magnetization plateau in Shastry-Sutherland magnets: Effect of the long-range Ruderman-Kittel-Kasuya-Yosida interaction

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Abstract – The ordering of the classical Ising model on the Archimedean lattice that is topologically equivalent to the Shastry-Sutherland one is studied in order to understand the fascinating magnetic properties experimentally observed in rare-earth tetraborides such as TmB_4. The long-range Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction which is expected to be predominant in these systems is taken into account, and the magnetization plateaus and rich ordering behaviors depending on the Fermi wave vector k_F are investigated in details by Monte Carlo simulation. The experimental 1/2 magnetization step can be qualitatively reproduced and its stability against the change of k_F is confirmed, suggesting that the coupling between conduction electrons and localized moments may play an important role in modulating the magnetization behaviors in these systems.

Introduction. – In the past few years, the magnetic orders in Shastry-Sutherland (S-S) magnets have drawn much attention due to the intrinsic effect of geometrical frustration [1–5]. Specifically, the so-called S-S lattice can be mapped as a square lattice with antiferromagnetic (AFM) couplings J_2 along the edges of the squares and diagonal AFM couplings J_1 in every second square, as shown in fig. 1(a) [6]. It has attracted great interest since its experimental realization in $SrCu_2(BO_3)_2$ which exhibits a fascinating sequence of magnetization (M) plateaus at fractional values of the saturated magnetization (M_s) in 1991 [7,8]. On the other hand, similar magnetization behaviors have also been observed in another representative S-S magnet: rare-earth tetraborides RB_4 (R = Tb, Dy, Er, Tm, etc.) with the magnetic moments located on an Archimedean lattice (fig. 1(b)) that is topologically

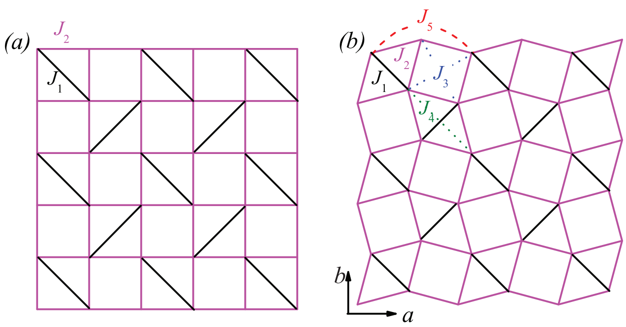

Fig. 1: (Color online) (a) The Shastry-Sutherland lattice with the diagonal coupling of J_1 along the edges of the squares and J_2 diagonally, and (b) the topologically identical structure realized in the (001) plane of rare-earth tetraborides with the additional interactions J_3, J_4 and J_5.

equivalent to the S-S one [9–12]. For example, the magnetization multi-plateaus at $M/M_s = 1/2$, 1/7, 1/9, etc., have been experimentally reported in TmB_4, and

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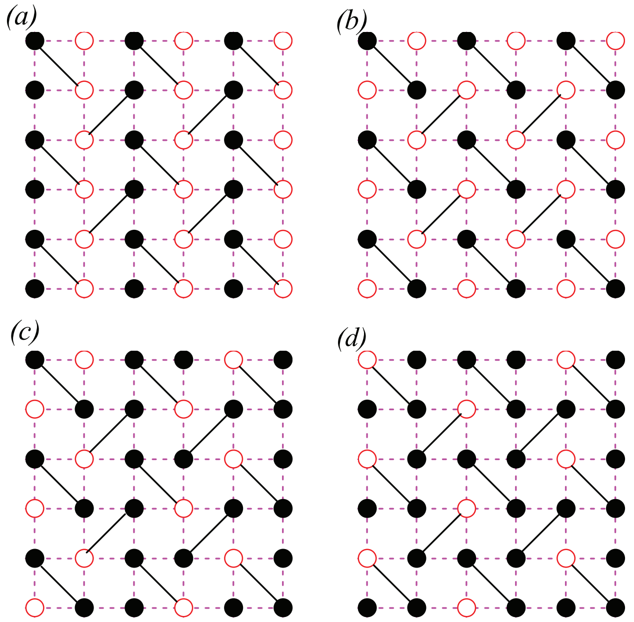

Fig. 2: (Color online) Spin configurations in (a) the collinear state, (b) the Néel state, (c) the UUD state, and (d) the FI state. Filled and open circles represent the up-spins and the down-spins, respectively.

several theoretical attempts to understand this fascinating phenomenon are reported [13–16].

Unlike $SrCu_2(BO_3)_2$ with Cu^{2+} ions carrying a quantum spin $S = 1/2$, TmB_4 presents a large total magnetic moment (the magnetic moment of Tm^{3+} is $\sim 6.0\mu_B$), and can be considered as a classical spin system. In addition, TmB_4 exhibits a strong easy-axis anisotropy due to the strong crystal field effects, thus it can be reasonably described by the classical Ising model. The magnetization process of the classical Ising model on the S-S lattice has been studied using the tensor renormalization-group approach, and various spin orders have been revealed [17]. Under low magnetic field (h), either the collinear state (fig. 2(a)) or the Néel state (fig. 2(b)) can be developed depending on the value of J_1/J_2 (filled and open circles represent the up-spins and the down-spins, respectively). When h is increased, a single magnetization plateau at $M/M_\mathrm{s} = 1/3$ resulting from a particular spin state in which each triangle contains two up-spins and one down-spin (the UUD state, fig. 2(c)) was predicted. Most recently, the ground states of the Ising model on the S-S lattice are investigated and the existence of a single 1/3 plateau has been rigorously proved [18]. In addition, the ferrimagnetic (FI) state (fig. 2(d)) resulting in the $M/M_\mathrm{s} = 1/2$ step has been predicted when the additional interactions are considered. In our earlier reports, the effects of the further interactions on the magnetization process have been systematically discussed via Monte Carlo simulations [19,20]. On the other hand, the quantum spin-(1/2) Ising-like XXZ model with additional interactions on the S-S lattice was visited using the quantum Monte Carlo method, and quantum spin fluctuations and long-range interactions are believed to play an important role in the emergence of the $M/M_\mathrm{s} = 1/2$ plateau [21,22].

All these theoretical works suggested that additional long-range interactions were indispensable to understand the main $M/M_\mathrm{s} = 1/2$ plateau experimentally observed in TmB_4. However, the origin of the additional interactions is not clear in physics so far. On the other hand, it is reasonable to expect that the long-range Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction may predominate in a metal system such as TmB_4, as will be explained below. The so-called RKKY interaction which refers to a coupling between localized inner electron spins in a metal by means of an interaction through the conduction electron was first introduced to explain the unusual width of the resonance absorption line of Ag, and was believed to be more suitable for rare-earth metals [23]. For example, the RKKY coupling between the localized $4f$ electron through the $6s$ conduction one may be dominant in TmB_4 and may play an important role in its magnetization process, as suggested in earlier work [11]. Thus, a systematic study of the effect of the RKKY interaction on the magnetic properties is seriously needed to completely understand the magnetization process of this system. However, the itinerant character of electrons is generally ignored in the most previous spin models for such a system, and few works on this subject have been reported as far as we know. In order to clarify this issue, we start with a classical Ising model on the two-dimensional (2D) Archimedean lattice interacting with the long-range RKKY interaction, and then extensive simulations on the magnetization behaviors are performed. The main $M/M_\mathrm{s} = 1/2$ plateau observed in experiments can be qualitatively reproduced, and rich ordering behaviors are revealed depending on the parameters used in the simulation.

The remainder of this paper is organized as follows: in the following section, the model and the simulation method will be presented and described. The third section is devoted to the simulation results and discussion. Finally, the conclusion is presented.

Model and method. – The Hamiltonian can be written as

$$H = \sum_{ij} J(r_{ij}) S_i \cdot S_j - h \sum_i S_i, \tag{1}$$

where S_i represents the Ising spin with unit length on site i, r_{ij} is the distance between the sites i and j, the long-range RKKY coupling between the spins at the sites i and j is described by [24]

$$J(r_{ij}) = -J_0 a^3 \left[\frac{\cos(2k_F r_{ij})}{r_{ij}^3} - \frac{\sin(2k_F r_{ij})}{2k_F r_{ij}^4} \right], \tag{2}$$

where J_0 is the constant that describes the intensity of the RKKY exchange interaction, a is the lattice constant which is set to be 1 for the sake of briefness, k_F is the Fermi wave vector. One may note that k_F is one of the most

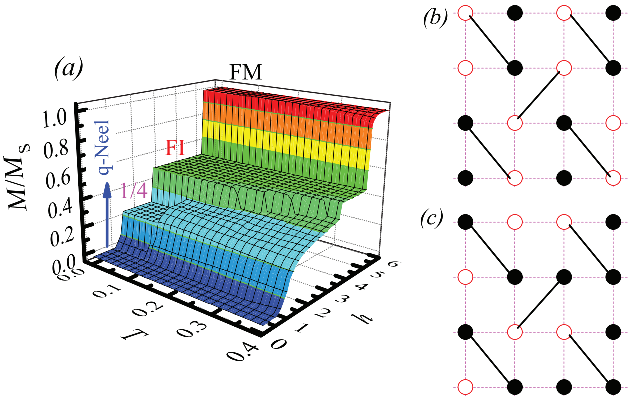

Fig. 3: (Color online) (a) Magnetization $M/M_{\rm s}$ *vs.* temperature T and magnetic field h. Spin configurations in (b) the q-Néel state and (c) the 1/4 state.

important parameters to tune the oscillation of the RKKY interaction. In our simulation, a smaller $k_F = 2\pi/1.243$ in units of a^{-1} than that in Tm is chosen due to the fact that the free-electron density of TmB_4 is lower than that of Tm [25]. Naturally, we also simulate the model with other values of k_F in order to examine the stability of the spin orders. In addition, J_0 is selected to satisfy the nearest-neighbor interaction $J_1 = 1$ and the choice of other values will not affect our conclusion at all.

Furthermore, it is noted that the ratio J_1/J_2 is nearly equal to 1 in TmB_4 and ErB_4, which is also realized in our simulation. In our simulation, the distance between two sites is calculated on the Archimedean lattice, while the spin configurations are discussed in its topologically equivalent S-S one in order to help one to easily understand the spin orders. The simulation is performed on an $L \times L$ ($L = 24$ is chosen unless stated elsewhere) lattice with periodic boundary conditions using the standard Metropolis algorithm and the parallel tempering algorithm [26,27]. In order to save CPU time, a cut-off radius $R_{ij} = 6$ is chosen. It will be checked later that the choice of R_{ij} never affects our conclusion. An exchange sampling will be taken every 10 standard Monte Carlo steps. The initial 1×10^4 Monte Carlo steps are discarded for equilibrium consideration and another 1×10^4 Monte Carlo steps are retained for statistic averaging of the simulation.

Simulation results and discussion. – Figure 3(a) shows the calculated M as a function of temperature (T) and h for $k_F = 2\pi/1.243$. At low T ($T < 0.1$), four magnetization steps can be observed respectively. Interestingly, instead of the collinear state or Néel state, a particular AFM state (q-Néel), with spin configuration (fig. 3(b)) which is similar to that of the Néel state, is developed and results in the $M = 0$ step for small h. When h increases to about 1.2, M switches to $M = M_{\rm s}/4$ resulting from the 1/4 state (the spin configuration is shown in fig. 3(c)). Subsequently, the magnetization step at $M = M_{\rm s}/2$ caused by the FI state is observed as h is further increased to about 2.4, which is similar to the earlier report [21]. Finally, M switches to $M_{\rm s}$ above $h \approx 4.8$, demonstrating the replacement of the FI state by the ferromagnetic (FM) one. It is noted that the 1/4 state is not much stable and can be completely destroyed by thermal disturbance when T increases to about 0.1. Contrarily, the FI state with the $M = M_{\rm s}/2$ step is rather stable even at $T = 0.4$, although the width of the step is noticeably decreased.

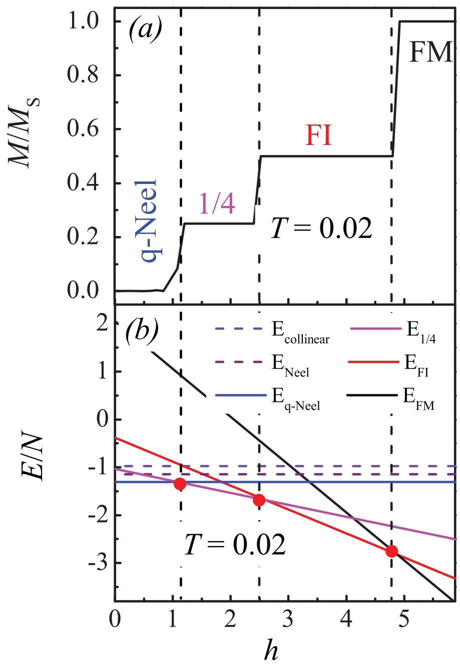

Fig. 4: (Color online) (a) Magnetization $M/M_{\rm s}$ and (b) the local energies as a function of h for different spin orders at $T = 0.02$.

Similarly [17], the ground states and the transition h between two successive plateaus can be estimated by comparing the energies at different spin configurations which are confirmed in our simulation. Specifically, the magnetization curve at $T = 0.02$ is shown in fig. 4(a) in which three transitions h are recognized. Accordingly, fig. 4(b) shows the calculated energies as a function of h for six possible states including the collinear state, the Néel state, the q-Néel state, the 1/4 state, the FI state and the FM state. The energy of the q-Néel state is lower than that of the collinear state or Néel state, leading to its stabilization for small h. When h increases to the first transition field $h \approx 1.2$, the 1/4 state is in the lowest energy, resulting in the emergence of the magnetization plateau at $M = M_{\rm s}/4$. Similarly, the 1/2 plateau can be observed due to the fact that the FI state is in the lowest energy when h is further increased to the second transition h. Finally, the FM state is developed when the static magnetic energy is predominant above $h \approx 4.8$.

In an earlier work [20], it has been proved that an AFM J_3 or a FM J_5 (fig. 1(b)) tends to stabilize the $M = M_{\rm s}/2$ plateau together with the destabilization of the $M = M_{\rm s}/3$

Table 1: The k_F-dependence of the exchange interactions J_3, J_4 and J_5, the spin orders for $M = 0$, and the main magnetization steps observed at low T.

$2\pi k_F^{-1}$	1.035	1.055	1.065	1.100	1.120	1.220	1.243	1.250	1.260
J_3	−0.0187	−0.1534	−0.2307	−0.6246	−1.1167	0.2992	0.1135	0.0707	0.0168
J_4	−0.1245	−0.0585	−0.0162	0.2143	0.5012	−0.1973	−0.0771	−0.0488	−0.0128
J_5	−0.0094	−0.0800	−0.1180	−0.2783	−0.4325	−0.1114	−0.1451	−0.1503	−0.1546
Spin order for M =0	Néel		q-Néel			q-Néel			Néel
Main steps	$M/M_s = 1/3$				None	$M/M_s = 1/2$, $1/4$		$M/M_s = 1/2$	

$2\pi k_F^{-1}$	1.270	1.300	1.360	1.390	1.410	1.420	1.430	1.450	1.460
J_3	−0.0304	−0.1437	−0.2963	−0.3501	−0.3802	−0.3937	−0.4063	−0.4290	−0.4392
J_4	0.0187	0.0934	0.1800	0.1991	0.2037	0.2036	0.2019	0.1934	0.1865
J_5	−0.1560	−0.1457	−0.0813	−0.0346	0.0005	0.0191	0.0384	0.0792	0.1007
Spin order for M =0	Néel								
Main steps	M/M_s = 1/2	M/M_s = 5/12, 1/6	$M/M_s = 1/2$			$M/M_s = 1/2$			

one, because more local AFM J_3 or FM J_5 interactions can be satisfied in the FI state than those in the UUD one. To some extent, the presence of AFM J_3 or FM J_5 interactions may be understood as a consequence of the oscillating nature of the RKKY interaction. In our simulation, these couplings are calculated to be $J_3/J_1 = 0.1135$ and $J_5/J_1 = -0.1451$ for $k_F = 2\pi/1.243$, leading to the stabilization of an extended $M = M_s/2$ plateau in the absence of the $M = M_s/3$ one at low T.

The stability of the FI state against the possible change of k_F has been examined, and the simulated results are summarized in table 1. The values of J_3, J_4 and J_5 are also given in order to help one to better understand the results. Similar results can be obtained for $k_F = 2\pi/1.220$ and $2\pi/1.250$ close to the case studied above. The q-Néel state is developed, resulting in the $M = 0$ step for small h. In addition, it is noted that the 1/4 state is so unstable and can be quickly suppressed with the change of k_F, as clearly shown in fig. 5(a) (dotted lines) which gives the estimated phase diagram in the (k_F, h)-plane at $T = 0.02$. In the intermediate h range, the $M = M_s/2$ plateau resulting from the FI state can be observed. However, when k_F is further decreased from $2\pi/1.2$, the system enters into a somewhat unphysical region in which the magnitude of J_3 or J_4 is several times larger than that of J_1. For example, $J_3 = -3J_1$ and $J_3 = -83J_1$ are obtained for $k_F = 2\pi/1.141$ and $k_F = 2\pi/1.152$, respectively, which is obviously inconsistent with experimental report.

Furthermore, the UUD state with the $M = M_s/3$ plateau can be developed when k_F is further decreased to $2\pi/1.100$. It is noted from table 1 that the FM J_5 decreases its magnitude and J_3 changes its sign from negative to positive with decreasing k_F. Thus, the FI state becomes unstable and can be replaced by the UUD one, leading to the emergence of the $M = M_s/3$ plateau. On

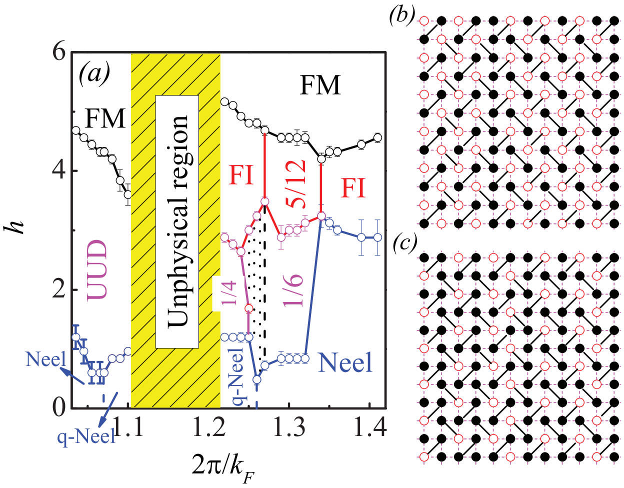

Fig. 5: (Color online) (a) The estimated phase diagram of the magnetization plateau in the (k_F, h)-plane at $T = 0.02$. The transition h is estimated from the magnetization curves and the positions of the peaks in the magnetic susceptibility. Spin configurations in (b) the 1/6 state and (c) the 5/12 state.

the other hand, different magnetic behaviors arise when k_F is further increased from $2\pi/1.250$. For example, the $M = M_s/6$ and $M = 5M_s/12$ plateaus are predicted for $k_F = 2\pi/1.300$. The spin configuration of the 1/6 state is shown in fig. 5(b), which is the same as in an earlier prediction [13]. In addition, the 5/12 state with the spin structure shown in fig. 5(c) can be developed in the intermediate-h region. This result is also confirmed by the energy comparison among those possible spin orders, although the corresponding results are not shown here. However, the FI state is revived again when k_F is increased to about $2\pi/1.350$. This phenomenon could be understood from two different points of view. On the one hand, the AFM J_3 is replaced by the FM one which favors the UUD state rather than the FI state, as discussed earlier. On the other hand, J_4 gradually decreases and eventually changes its sign from negative to positive at around $k_F \approx 2\pi/1.270$. Importantly, the AFM J_4 tends to favor the FI state rather than the UUD one due to the fact that more AFM J_4 interactions can be satisfied in the FI state. This viewpoint can be easily understood from the respective spin configurations which are shown in fig. 2. Finally, the FI state can be developed as a consequence of the strong competitions among the long-range interactions even for increasing k_F. Furthermore, either the Néel or the q-Néel state is recognized for small h at low T, demonstrating the fact that the collinear state is unfavorable throughout our simulation.

However, one may note that some of the fractional plateaus at $M/M_s = 1/7$, $1/9$, etc. reported in experiments can not be simultaneously reproduced with $M/M_s = 1/2$ even when the long-range RKKY interaction is taken into account. This inconsistency between the experiment and the present simulation may be understood from the following two aspects. On the one hand, the time available in experiment may not be sufficient for the spin rearrangement for small h at low T, and the spins may be easily trapped into a metastable state rather than into the equilibrium one. Actually, the effect of the magnetization dynamics on the magnetic behaviors of a classical spin model on the S-S lattice has been investigated and the formation of domain walls due to the non-equilibrium magnetization process is suggested to be responsible for the emergence of the fractional plateaus [28]. On the other hand, it is noted that the disorder effect caused by the inhomogeneity inevitable in realistic materials is completely ignored in our simulation, which may also contribute to the inconsistency in some extent.

Anyway, the present work reveals a significant effect of the long-range RKKY interaction that is expected to be predominant in rare-earth tetraborides RB_4 due to their metallic character. The experimental $M = M_s/2$ plateau can be qualitatively reproduced and the FI state occupies a considerable region of the parameter space, strengthening our conclusion that this plateau may be caused by the long-range RKKY interaction. In other words, it is partially suggested that the additional interactions responsible for the 1/2 step in S-S magnets may be of the RKKY type. Definitely, some other factors may be needed to fully understand all the experimental results. For example, the magnetization dynamics or/and the inevitable disorder effect may be the potential choices. However, the related subject is beyond the scope of this study and will be discussed elsewhere.

Finally, in order to verify the reliability of our simulation, the dependences of the step-like magnetization feature on the cut-off radius R_{ij} and the lattice size L are also checked. The simulated magnetization curves for different R_{ij} ($R_{ij} = 6$, 7, and 8) for $k_F = 2\pi/1.243$ are presented in fig. 6(a). These curves perfectly coincide with each other, indicating that the choice of R_{ij} in this work

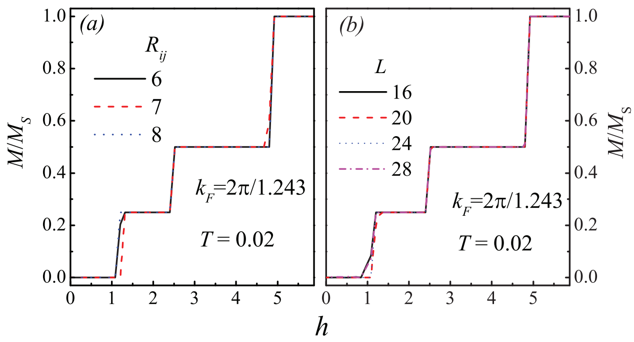

Fig. 6: (Color online) The calculated magnetization curves for (a) different cut-off radius R_{ij} and (b) different lattice sizes L at $T = 0.02$.

is reasonable. Moreover, fig. 6(b) shows the simulated magnetization curves for different L ($L = 16$, 20, 24, and 28). The simulated curves overlap, demonstrating that the finite-size effect on the magnetization properties in this model is negligible and our conclusion is reliable.

Conclusion. – In conclusion, we have studied the magnetic behaviors of the classical Ising model with the long-range RKKY interaction on the Archimedean lattice by means of Monte Carlo simulation in order to understand the magnetic process in rare-earth tetraborides such as TmB_4. The main magnetization plateau at $M = M_s/2$ observed in experiments can be qualitatively reproduced and is proved to be stable in a wide range of the parameter k_F. Thus, our work strongly suggests that the $M = M_s/2$ plateau in such a system may be resulting from the long-range RKKY interaction.

* * *

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