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ORIGINAL PAPER

Formation of Magnetization Plateaus in Rare Earth Tetraborides: Exact Diagonalization and Quantum Monte Carlo Studies

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Abstract

A relatively simple model based on the coexistence of spin and electron subsystems is proposed for a description of magnetization processes in metallic rare earth tetraborides. The model takes into account the Ising interaction between the localized spins, the Hubbard interaction between the itinerant electrons, and the Ising interaction between electron and spin subsystems. To solve this model, a combination of small cluster exact diagonalization calculations and quantum Monte Carlo simulations is used. Particular attention is paid to a description of correlation effects (the Hubbard interaction) on formation and stabilization of magnetization plateaus with fractional magnetizations. It is shown that the Hubbard interaction significantly stabilizes the 1/2 magnetization plateau and simultaneously suppresses the 1/3 magnetization plateau, in accordance with experimental measurements in rare earth tetraborides.

Keywords Magnetization plateaus · Shastry-Sutherland lattice · Rare earth tetraborides

1 Introduction

In the past decade, a considerable amount of effort has been devoted to understanding of the multitude of anomalous magnetic properties of the so-called Shastry-Sutherland rare earth metal tetraborides. One of the most interesting of them is surely the formation of fascinating sequences of magnetization plateaus with fractional magnetizations. They are observed practically for all members of the metallic RB_4 family group (R = Tb, Dy, Ho, Er, Tm, etc.) and form a wide spectrum of magnetization curves in these materials. For instance, a single plateau at one-half of the saturation magnetization (m/m_s = 1/2) has been observed in ErB_4 [1]. In TmB_4, in addition to an extended 1/2-plateau a narrow plateau with fractional value of m/m_s = 1/8 for temperatures below 4 K has been detected [2, 3], and in TbB_4 very complex sequence, consisting of several magnetization plateaus with fractional magnetizations $m/m_s = 1/2, 4/9.1/3, 2/9, 7/9$ is found [4].

It is supposed that the anomalous properties of these systems are caused by the geometrical frustration that leads to an extensive degeneracy in the ground state. Indeed, all above mentioned compounds have the tetragonal structure (space group P4/mbm) with magnetic ions R^{3+} located on an Archimedean lattice that is topologically equivalent to the so-called Shastry-Sutherland lattice [5] that exhibits the strong geometrical frustration (see Fig. 1).

Moreover, since there are the strong crystal field effects, these compounds can be described in terms of an effective spin-1/2 Shastry-Sutherland model under strong Ising anisotropy. This is, for example, the case of TmB_4 and ErB_4 where the easy-magnetization axis is normal to Shastry-Sutherland planes [1]. Thus, the study of the Ising model was the first and natural step toward a complete understanding of magnetization processes in these materials. This model has been solved numerically [6, 7] as well as analytically [8] with a conclusion that only the m/m_s = 1/3 plateau is stabilized by the nearest J_1 and next nearest J_2 interactions. The existence of the magnetization plateau at only 1/3 of the saturation magnetization and its absence at 1/2 of the saturation magnetization, as observed for example in ErB_4 and TmB_4, indicates that it is necessary to go beyond the classical Ising limit to reach the correct description of the magnetization processes in rare earth tetraborides. The three main generalizations

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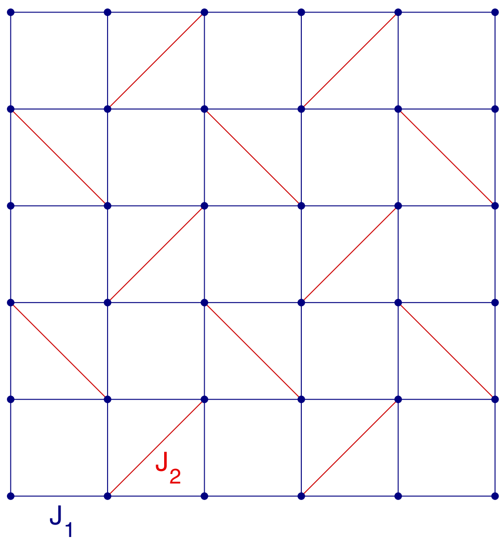

Fig. 1 The Shastry-Sutherland lattice with the first (J_1) and the second (J_2) nearest neighbor couplings

have been proposed to reach this goal. The first such a generalization has been done by Meng and Wessel [9] who studied the spin-1/2 easy-axis Heisenberg model on the Shastry-Sutherland lattice with ferromagnetic transverse spin exchange using quantum Monte Carlo and degenerate perturbation theory. Besides the magnetization plateau at 1/3 of the saturation magnetization they found a further narrow plateau at 1/2, which persists only in the quantum regime.

The second type of generalizations represent various extensions of the Ising model with the third, fourth and even fifth nearest neighbors [7, 8, 10]. It was shown that these extensions are able to describe some of the individual plateaus observed in rare earth materials, as well as the partial sequences consisting of two or even three right magnetization plateaus, but it is questionable if it is the intrinsic property of a model, or only a consequence of the large number of variables (fitting parameters) that enter to the model as interaction parameters.

And finally, the third type of generalizations are based on the assumption that for a correct description of magnetization processes in rare earth tetraborides one should take into account both spin and electron subsystems since these compounds are not insulating, but metallic [11, 12]. Such an model has been also examined in our previous paper [13]. It takes into account the Ising interaction between the first (J_1) and second (J_2) nearest neighbor spins on the Shastry-Sutherland lattice, the kinetic energy of the noninteracting electrons, that hop on nearest and next nearest lattice sites with probabilities t and t', as well as the Ising type interaction J_z between the electron and spin subsystems. The Hamiltonian of this model can be written as

$$H = \sum_{ij\sigma} t_{ij} d^+_{i\sigma} d_{j\sigma} + J_z \sum_i (n_{i\uparrow} - n_{i\downarrow}) S^z_i - h \sum_i (n_{i\uparrow} - n_{i\downarrow}) + \sum_{ij} J_{ij} S^z_i S^z_j - h \sum_i S^z_i \,, \tag{1}$$

where $d^+_{i\sigma}$, $d_{i\sigma}$ are the creation and annihilation operators of the itinerant electrons in the d-band Wannier state at site i, $n_{i\sigma} = d^+_{i\sigma} d_{i\sigma}$ and $S^z_i = \pm 1/2$ denotes the z-component of a spin-1/2 degree of freedom on site i of a square lattice. Numerical studies that we have performed with this Hamiltonian showed [13] that the model based on the coexistence and coupling between the electron and spin subsystems can yield answers on some fundamental questions concerning the formation and stabilization of magnetization plateaus with fractional magnetizations in metallic rare earth tetraborides. In particular it is shown that just the electron spin coupling is responsible for the appearance of the 1/2 plateau that suppresses the stability region of the 1/3 plateau and thereby points to possible mechanism that could lead to a complete disappearance of the 1/3 plateau on the magnetization curve and its substitution by the 1/2 plateau in accordance with experimental measurements in ErB_4 and TmB_4. Unfortunately, the reduction of the 1/3 plateau phase and the stabilization of the 1/2 plateau phase due to the spin-electron Ising coupling is still too small to reach the accordance between the theoretical and experimental results and therefore a further generalization of the model is needed.

2 Model and Methods

The most crude approximation, from the point of view of the above discussed model seems to be the fact that spins are considered as interacting, but electrons not. This deficiency of the model can be easily removed, by including the Hubbard type interaction between up and down spin electrons on the same site, but such an extension significantly complicates numerical calculations since the model becomes now fully quantum. Indeed, with the Hubbard interaction term the model Hamiltonian (1) takes the form:

$$H = \sum_{ij\sigma} t_{ij} d^+_{i\sigma} d_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} + J_z \sum_i (n_{i\uparrow} - n_{i\downarrow}) S^z_i - h \sum_i (n_{i\uparrow} - n_{i\downarrow}) + \sum_{ij} J_{ij} S^z_i S^z_j - h \sum_i S^z_i \,, \tag{2}$$

which can be further simplified as follows:

$$H = H_H + H_I = \sum_{ij\sigma} t'_{ij\sigma} d^+_{i\sigma} d_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} - h \sum_i (n_{i\uparrow} - n_{i\downarrow}) + \sum_{ij} J_{ij} S^z_i S^z_j - h \sum_i S^z_i \,, \tag{3}$$

where $t'_{ij\sigma} = t_{ij} + J_z\sigma S_i^z\delta_{ij}$ represent new hopping amplitudes of the itinerant electrons which depend now explicitly on the particular spin distribution $s = \{S_1^z, S_2^z \dots S_L^z\}$. Thus for a given spin distribution s the model Hamiltonian (2) can be considered as the ordinary Hubbard model with generalized hopping amplitudes and its solution reduces to finding a configuration s_{min}, which minimizes the ground state energy of the model. For given s the ground state energy can be found in principle exactly by the exact diagonalization Lanczos method [14], but since the hopping amplitudes depend now explicitly on s, the Lanczos procedure has to be used many times (strictly said 2^L times, when J_z and h are switched off and $L2^L$ times otherwise), which imposes severe restrictions on the size of clusters that can be studied within the exact diagonalization method ($L = 12$). Therefore, for large clusters we use another method, and namely, the combination of the Projector Quantum Monte Carlo (PQMC) method [15] and well controlled numerical algorithm that we have initially elaborated and used for a determination of ground states of the spinless [16] as well as spin-one-half [17] Falicov Kimball model. The algorithm can be summarized as follows: (i) Chose a trial spin configuration $s = \{S_1^z, S_2^z \dots S_L^z\}$ and calculate the spin energy $E_s = \sum_{ij} J_{ij} S_i^z S_j^z$. (ii) Having U, J_z and the total number of electrons ($N_\uparrow + N_\downarrow = L$) fixed, find (by using the PQMC) the ground state energies $E(N_\uparrow)$ of the generalized Hubbard model for all $N_\uparrow = 1 \dots L$. (iii) For given h find the total energy of the electron subsystem $E_e(N_\uparrow^0)$ by taking the minimum of $E(N_\uparrow) - h(N_\uparrow - N_\downarrow)$ as well as the total energy of system $E_t(s) = E_e(N_\uparrow^0) - E_s - h\sum_i S_i^z$. (iv) Generate a new spin configuration s_{new} by flipping a randomly chosen spin and calculate new total energy of the system $E_t(s_{new})$. If $E_t(s_{new}) < E_t(s)$ the new spin configuration is accepted, otherwise it is rejected. Then, the steps (ii)–(iv) are repeated until the convergence (for given U and J_z) is reached. Of course, one can flip instead of one spin in step (iv) simultaneously two or more spins, thereby the convergence of method is improved.

3 Results and Discussion

To reveal the possible effects of the Coulomb interaction between electrons on formation of magnetization plateaus within the model Hamiltonian (2) we have started our study on small finite cluster of $L = 12$, where the exact results can be obtained by the Lanzos method. In the next we also use these results for testing the accuracy of results obtained by PQMC method within the above describe algorithm. The $L = 12$ cluster that we have used in our numerical calculations has an exotic shape (see Fig. 2) and belongs to the class of the so-called tilted lattices, which are used frequently in the two-dimensional numerical simulations on small clusters [14].

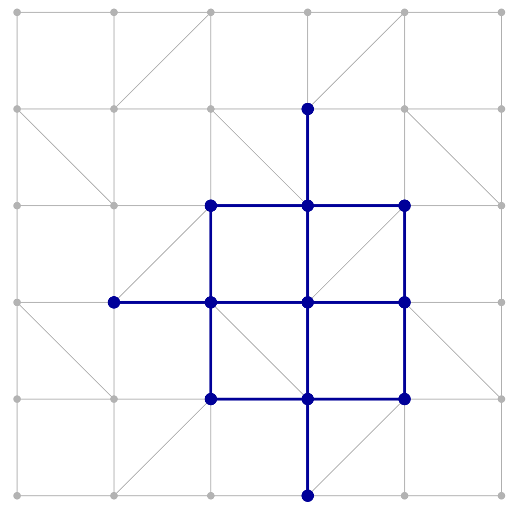

Fig. 2 The $L = 12$ site cluster used in our numerical calculations

To minimize the number of model parameters we have used in all our next numerical calculations the following set of input values $J_1 = J_2 = 1, t = t' = 1, J_z = 4$ (only U is left as a free parameter), for which the largest effect of the electron spin coupling on the stabilization of the 1/2 plateau phase have been observed for $U = 0$ [13].

The results of our exact numerical calculations on finite cluster of $L = 12$ sites are summarized in Fig. 3, were the final magnetization curves as functions of external magnetic field h are plotted for $U = 0$ and $U = 2$.

Before discussing the effects of Coulomb interaction on formation of magnetization plateaus, let us note some interesting facts that can be found by comparing our zero-U results obtained on very small cluster of $L = 12$ and ones obtained in our previous paper [13] for much larger cluster consisting of $L = 140 \times 140$ sites. Such a comparison shows that despite the very small size of cluster used in the current paper, it is able to reproduce, at least qualitatively, all main features of magnetization curve found on much larger cluster, including the appearance of the 1/2 plateau phase, its stabilization at the expense of the 1/3 plateau phase, as well as the appearance of the secondary structure, observed at m/m_s=1/5,1/7 on $L = 140 \times 140$ cluster and at m/m_s=1/6 on $L = 12$ cluster. This indicates that results presented in Fig. 3, for finite U, do not have to be considered as informative only, but can reveal real trends also valid for macroscopic systems. Two possible trends are obvious, the increasing Coulomb interaction (i) stabilizes the 1/2 plateau phase and (ii) suppresses the 1/3 plateau phase, in accordance with our assumptions that the above proposed Hamiltonian (2) could model more realistically situation in rare earth compounds than one without the Hubbard interaction term (Eq. 1). Since it is generally accepted that these materials belong to the class of strongly correlated

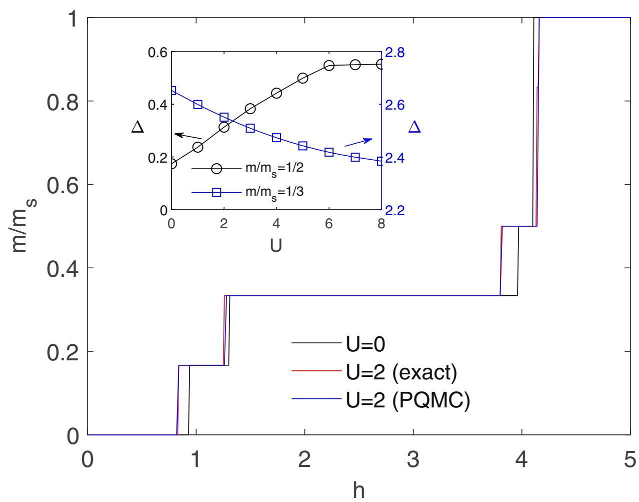

Fig. 3 The magnetization curves of the spin-electron model (2) calculated for two different values of U by exact diagonalization method (exact) and projector quantum Monte Carlo method (PQMC) on the $L = 12$ site cluster. The inset shows the width of the 1/2 and 1/3 plateau as a function of the Coulomb interaction U

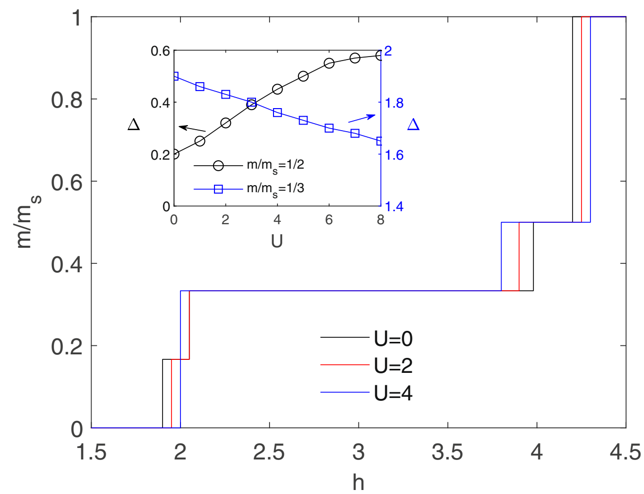

Fig. 4 The magnetization curves of the spin-electron model (2) calculated for three different values of U by the PQMC method on the $L = 12 \times 12$ site cluster. The inset shows the width of the 1/2 and 1/3 plateau as a function of the Coulomb interaction U

systems where $U >> t$, the effects of finite U can be indeed very strong. This is demonstrated in the inset to Fig. 3, where the width of the 1/2 plateau is plotted as a function of U. It is seen that the width of the plateau first rapidly increases with increasing U and then tends to saturated value, which is approximate 3 times larger against its value at $U = 0$, indicating that the Coulomb interaction should be certainly taken into account in the correct description of magnetization processes in rare earth tetraborides.

Of course it is natural to ask if these results persist also on much large clusters. To answer this question we have used the second method discussed above, which is based on the combination of the QMC and the well controlled algorithm. In this case the QMC simulations were performed using a projector algorithm which applies $\exp(-\theta H)$ to a trial wave-function (in our case the solution for $U = 0$). A projector parameter $\theta \sim 30$ and a time slice of $\Delta\theta = 0.05$ suffice to reach well converged values of the observables discussed here. This is illustrated in Fig. 3 where the quantum Monte Carlo results are compared directly with the exact diagonalization results obtained on the $L = 12$ cluster ($U = 2$) and a nice accordance of results is found over the whole region of h values. In the next step we have performed the same calculations on larger clusters consisting of $L = 12 \times 12$ sites. The resultant magnetization curves obtained by this method for several different values of U are displayed in Fig. 4 and they confirm practically all conclusions made on the base of exact-diagonalization calculations on the $L = 12$ cluster.

Again there are observed strong effects of the Coulomb interaction U on the stabilization of the 1/2 plateau and the suppression of the 1/3 plateau and it is interesting to note that only very small finite size effects are found on the width of the 1/2 plateau phase. This is explicitly demonstrated in the inset to Fig. 4, where the width of this plateau is plotted as a function of U and a nice qualitative and even quantitative accordance between results obtained on $L = 12$ and $L = 12 \times 12$ cluster is found indicating that these results can be satisfactorily extended on macroscopic systems. Thus we can conclude that the inclusion of the Coulomb interaction between up and down spin electrons of the Hubbard type leads in accordance with our suppositions to stabilization of the 1/2 plateau and suppression of the 1/3 plateau and makes model more realistic for description of magnetization processes in metallic rare earth tetraborides. However, it should be noted that despite strong effects of the Coulomb interaction on both 1/2 as well as 1/3 plateau they are still insufficient to suppress completely the 1/3 plateau phase, as observed in some rare earth compounds. The most probable explanation of this deficiency is too simplified description of electron-spin interaction, here considered of the Ising type instead of the more realistic Heisenberg one. Indeed, it is reasonable to assume that the replacement of the Ising interaction by the Heisenberg one could improve the accordance between the theoretical and experimental results, since as was shown by Meng and Wesell [9], the Heisenberg interaction itself produces the same effect as the Hubbard interaction (this result is intuitively expected since the Hubbard model in the strong-coupling limit and the half-filed band case can be mapped on the Heisenberg model [18]) and thus combined effects of both interactions could completely suppress the 1/3 plateau. The work in this direction is currently in progress.

In summary, we have proposed a relatively simple model for a description of magnetization processes in metallic rare earth tetraborides. The model is based on the coexistence of spin and electron subsystems that are present in these compounds and takes into account the Ising interaction between the localized spins, the Hubbard interaction between the itinerant electrons and the Ising interaction between electron and spin subsystems. Particular attention was paid to a description of correlation effects (the Hubbard interaction) on formation and stabilization of magnetization plateaus with fractional magnetizations. It was shown that the Hubbard interaction significantly stabilizes the 1/2 magnetization plateaus and simultaneously suppresses the 1/3 magnetization plateau, in accordance with experimental measurements in rare earth tetraborides, where the 1/2 plateau is the main magnetization plateau and the 1/3 plateau is not present at all or it is stable only on very narrow interval.

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