Magnetic-phase transition in the magnetic-polaron system studied with the Monte Carlo method: Anomalous specific heat of EuB_6

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We have investigated the phase transition in the magnetic-polaron system to understand the anomalous behavior of the specific heat C in EuB_6 near the magnetic-phase transition temperature T_C. By employing the exact diagonalization and the Monte Carlo method, we have found a new temperature scale T_p, at which C has its maximum value and the magnetic polarons become percolated. We have demonstrated that, upon cooling, the isolated magnetic polarons become linked to yield the finite magnetization at T_C, and then the linked magnetic polarons eventually merge to show the percolating behavior with the C peak at T_p ($T_p < T_C$). The validity of the Fisher-Langer relation in the magnetic-polaron system is also discussed.

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I. INTRODUCTION

Interest in the magnetic polaron has been revived because physical properties of some candidate spintronic materials are thought to be governed by the magnetic polarons.[1] Diluted magnetic semiconductors (DMS)[2–4] and colossal magnetoresistance (CMR) materials are good examples.[5] The concept of the magnetic polaron was proposed a long time ago.[6,7] However, the understanding of the magnetic polaron and related physical phenomena is still far from complete. The comprehensive description of the magnetic-phase transition in a magnetic-polaron system is lacking. It was suggested that the magnetic-phase transition is induced by the percolation phenomena of magnetic polarons upon cooling,[4,8] but the detailed process is yet to be resolved. Furthermore, there have been no theoretical studies on the thermal properties of the magnetic-polaron system. The specific heat provides the information of the energy of a system. It is thus expected that the specific heat behavior would provide a very important clue to the understanding of the magnetic and percolation transitions in the magnetic-polaron system.

One of the reasons that makes the physics of the magnetic polaron difficult is the complicated coupling of the magnetic polaron with lattice or disorder.[9,10] For the concentrated system, one should also consider the electron-electron correlation. In this respect, EuB_6, which is a low carrier-density metallic system, is ideal to study the physics of magnetic polarons, independently of other degrees of freedom. EuB_6 experiences the ferromagnetic (FM) and insulator-metal transitions near $T_C \approx 15.5$ K (T_C: Curie temperature). EuB_6 also shows very large negative magnetoresistance near T_C,[11] similarly to CMR perovskite manganites. The magnetic and transport properties of EuB_6 have been described well based on the magnetic polaron model.[12–17]

EuB_6 reveals an intriguing thermal property, which has not been well understood. As shown in the top of Fig. 1, the specific heat of EuB_6 exhibits a broad peak at $T_p \approx 10.5$ K in addition to a small peak at $T_C \approx 15.5$ K.[18–23] The differential resistivity $d\rho/dT$ in the bottom of Fig. 1 also shows an extraordinary feature, indicating two consecutive transitions in EuB_6.[23,24] The deviated feature of T_p from T_C in EuB_6 ($T_p = 0.6 - 0.7 T_C$) is unusual, since, in normal magnetic systems, the peak position of the specific heat coincides with T_C. It is also notable that, distinctly from the normal metallic magnetic systems, the Fisher-Langer relation[25] does not apply to the EuB_6 system. According to Fisher and Langer,[25] $d\rho/dT$ would have a similar behavior to the magnetic specific heat near T_C. The anomalous specific heat data suggest that EuB_6 would not be a simple collinear ferromagnet and the magnetic-phase transition would not be of the second-order type.[19]

Süllow *et al.*[22] once proposed that the spin reorientation from the [100] to the [111] direction would produce the two-peak structure in the specific heat. In the later publication, Süllow *et al.*[23] proposed another interpretation for the two-peak structure in $d\rho/dT$: the higher peak at $T_{C1} = 15.5$ K represents the emergence of the spontaneous magnetization accompanied by the metallization, while the lower peak at $T_{C2} = 12.6$ K represents the bulk magnetic transition.[26] They tried to explain the specific heat peak at T_p as arising from the bulk FM transition at T_{C2}. However, there seems to be too large a separation between $T_p \approx 10.5$ K of the former and $T_{C2} = 12.6$ K of the latter. Further, there has been no concrete theoretical support for this interpretation.

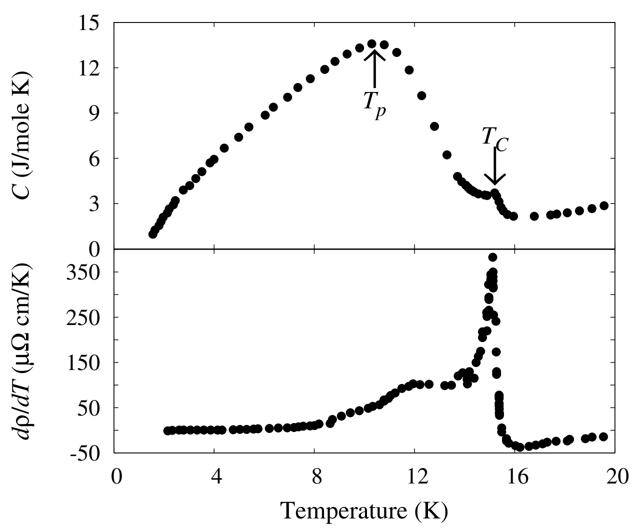

FIG. 1. Anomalous specific heat C (top) and differential resistivity $d\rho/dT$ (bottom) for EuB_6 (reproduced from Ref. 23).

To clarify the origin of the anomalous specific heat in EuB_6, we have investigated systematically magnetic, transport, and thermal properties of the magnetic-polaron system using the unbiased numerical simulation method. We have identified a new temperature scale T_p, at which the percolation of the magnetic polarons occurs below the magnetic-phase transition temperature T_C. We propose that the percolation temperature T_p corresponds to the peak position of the anomalous specific heat observed in EuB_6.

II. MAGNETIC, TRANSPORT, AND THERMAL PROPERTIES IN THE MAGNETIC-POLARON SYSTEM

To describe the physics in the magnetic-polaron system, we have employed the following FM Kondo lattice model:

$$H = -t \sum_{\langle i,j\rangle\sigma} (c_{i\sigma}^{\dagger} c_{j\sigma} + \text{H.c.}) - J_H \sum_i \vec{\sigma}_i \cdot \vec{S}_i, \quad (1)$$

where $c_{i\sigma}^{\dagger}$ creates a charge carrier with spin σ at site i, and $\vec{\sigma}_i$ and $\vec{S}_i$ are spins of the carrier and local moments, respectively. t and J_H represent the hopping parameter and the Hund-type coupling between the carrier spin and local moments. t is used as an energy unit ($t=1$), and we have assumed a very large J_H/t ratio as much as 10.[27] $\langle i,j\rangle$ runs over nearest-neighbor lattice pairs. For local $\vec{S}_i$, the Ising-type spin is adopted ($S_z = \pm 1$). The local spin magnitude in EuB_6 is larger than 1, but it is well known that the overall result is not sensitive to the spin size: calculations with the classical spin ($S=\infty$) and the Ising-type spin give equivalent results.[16,28–30] The carrier-carrier interaction is neglected because we are interested in the dilute carrier limit. Furthermore, the large value of J_H reduces excessively the probability of double occupancy in the dilute system, and so the on-site Hubbard repulsion can be safely neglected.[31]

To solve the above model Hamiltonian, we have performed the Monte-Carlo simulation using the standard metropolis algorithm[32] on the 8×8 two-dimensional square lattice with the periodic boundary condition. Although usual magnetic-polaron systems are three dimensional, the calculations are often performed on one- or two-dimensional systems. It is because the magnetic-polaron model does not depend much on the dimensionality and the lattice type.[28–30] The small lattice size is justified because the size of the magnetic polaron is small enough with respect to the lattice size near T_C. The electronic energy was calculated through the exact diagonalization method.[33] The resistivity was obtained by the Kubo formula and the f-sum rule.[34] Finite broadening of 0.2 was assumed for delta functions to consider broadening effects that are not included in the model. All the data were obtained while cooling, but the calculation is reversible for the temperature variation.

The above model Hamiltonian yields a phase diagram that describes a transition from the high-temperature paramagnetic (PM) state to the low-temperature FM state through the magnetic-polaron state.[16] The magnetic-polaron state is characterized by a hump or a peak in the temperature-dependent resistivity curve near T_C. We have examined the behavior of specific heat near T_C. The magnetic susceptibility χ and the specific heat C were calculated from $\chi = [\langle M^2\rangle - \langle M\rangle^2]/Nk_BT$ and $C = [\langle E^2\rangle - \langle E\rangle^2]/N(k_BT)^2$, where M and E are the local magnetic moment and the energy of the system, respectively. N represents the number of sites. The specific heat can also be calculated from the temperature derivative of the energy. This method gives equivalent results, but with much larger statistical error.

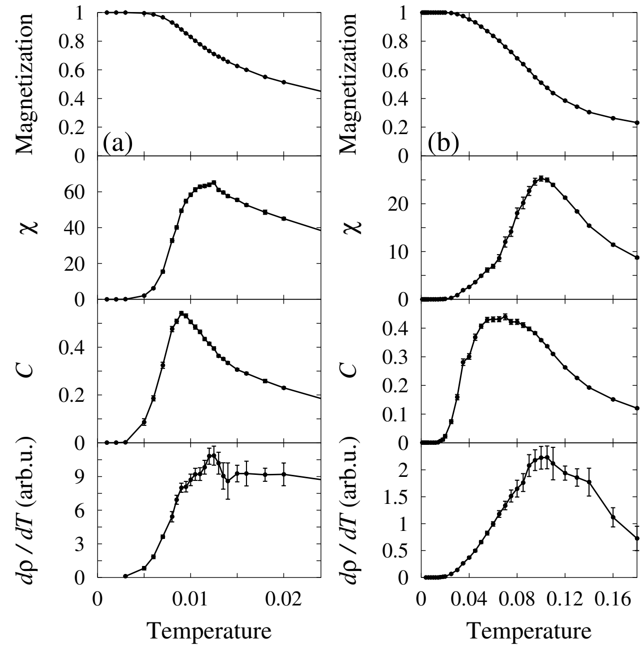

FIG. 2. (a) Magnetization, magnetic susceptibility (χ), specific heat (C), and the temperature derivative of resistivity ($d\rho/dT$) for a one-carrier magnetic-polaron system ($n=0.0156$). The calculation was performed on the 8×8 square lattice. (b) The same as (a) but for a five-carrier magnetic-polaron system ($n=0.0781$).

Figure 2 presents the magnetization of local moments, the magnetic susceptibility (χ), the specific heat (C), and the temperature-derivative of resistivity ($d\rho/dT$) near T_C, which are obtained for systems with one and five carriers ($N_c = 1,5$). T_C is determined by the peak position of χ. Peaks of C for $N_c=1$ and $N_c=5$ are located at $T_p=0.72T_C$ and at $T_p = 0.70T_C$, respectively. Noteworthy is that the behaviors of calculated χ and C are quite similar to those of experimental χ and C for EuB_6. The amount of deviation between T_p and T_C is close to the observation $T_p = 0.6-0.7T_C$. Due to the limited resolution of the calculation, it is difficult to identify a small peak at T_C in the C curve. The similar results were obtained for other carrier densities ($N_c=3,4$) except for $N_c = 2$.[35] We also considered smaller lattices (6×6 and 4×4 square lattices) and obtained equivalent results.

Numerical calculations with the conventional magnetic model Hamiltonian sometimes yield different peak positions for the susceptibility and the specific heat due to the finite-size effect. The deviation between the peaks, however, does not exceed several percent. In contrast, the deviation between T_C and T_p in the present case is about 30%, and it becomes as much as 80% for the case of $N_c=2$. In addition, the deviation is very robust with a change in the lattice size. Thus, we believe that the deviation of the specific heat peak from magnetic T_C is an intrinsic feature in the magnetic-polaron system, and the anomalous specific heat in EuB_6 can be described properly in terms of the magnetic-polaron model.

The resistivity near T_C decreases monotonically upon cooling. The differential resistivity $d\rho/dT$ in the bottom panels of Fig. 2 shows a maximum at T_C and behaves quite similarly to the magnetic susceptibility χ. According to Fisher and Langer,[25] in normal metallic magnetic systems, $d\rho/dT$ would have a similar scaling behavior to the magnetic part of specific heat due to the short-range magnetic fluctuations near T_C. Although $d\rho/dT$ shows its maximum at T_C, the deviation of the maximum $d\rho/dT$ from T_p in Fig. 2 indicates that the Fisher-Langer relation[25] does not hold in the magnetic-polaron system. It is not surprising, however, because in the magnetic-polaron system, the spatial inhomogeneity—which was not considered by Fisher and Langer—would play an important role during the magnetic transition.

We have also examined the effect of the impurity potential, which turned out to give a resistivity peak above T_C.[16] We considered one impurity with an attractive potential of $\varepsilon_0=-1.5$ for the system with $N_c=1$. We have obtained $T_C=0.009$ and $T_p=0.007$. Thus, the impurity reduces both T_C and T_p, but the discrepancy ratio between the two is retained.

III. MAGNETIC AND PERCOLATION TRANSITIONS

We have explored the magnetic and thermal properties further to get an insight into the unusual transition in the magnetic-polaron system. The magnetic susceptibility and the specific heat describe the fluctuation of the magnetization and the energy of the system, respectively. The magnetic susceptibility is maximized at T_C, where the magnetic correlation length diverges. In the magnetic polaron model, the energy of the system is given by the kinetic energy of carriers in the background of local moments [see Eq. (1)]. The specific heat would be maximized when the carriers are delocalized with the occurrence of polaron percolation at T_p. Therefore, the deviation of T_p from T_C implies that the polaron percolation does not occur simultaneously with the FM long-range ordering of the local magnetic moments.

This situation is clearly seen in Fig. 3, which provides the change of the magnetic-polaron state as a function of temperature for $N_c=2$. It is seen that the magnetic polarons formed at $T\sim0.1$ grow in size upon cooling so as to extend over the whole lattice at $T\sim0.0085$. Based on this result, we can derive the schematic phase diagram as shown in the bottom of Fig. 3. The magnetic polarons are formed at T^* much higher than T_C.[36] These polarons are isolated from one another and the magnetization directions of the polarons are random. Let us call this state an isolated magnetic-polaron (MP) phase. As they grow in size upon cooling, the large FM cluster appears through the polaron overlaps, which can be called the linked MP phase. T_C is defined by this phenomenon. In the linked MP phase, the carriers are still confined to the magnetic-polaron clusters, and so the metallic and magnetic regions are seen to be separated from the insulating and paramagnetic regions.[16] Further cooling induces all the linked magnetic polarons to be merged so that the polaron

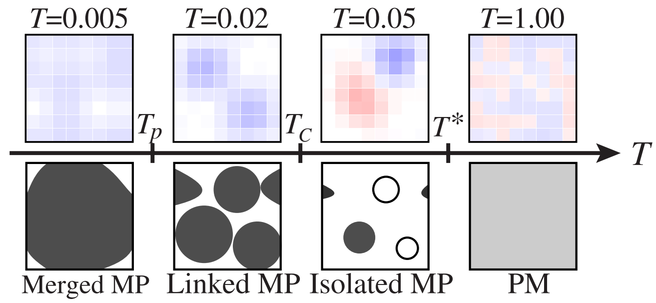

FIG. 3. (Color online) (Top) Typical magnetic polaron (MP) state at each temperature for $N_c=2$. Red and blue colors at each cell represent the carrier density at the cell with opposite spin directions. (Bottom) Schematic phase diagram that describes the unusual magnetic-phase transition in the magnetic-polaron system. There are three different temperature scales, T^*, T_C, and T_p. Upon cooling, the high-temperature PM state transforms to the isolated MP state at T^*, and then to the linked MP state at T_C, and eventually to the merged MP state at T_p. Solid and empty objects represent magnetic polarons with opposite magnetization.

percolation occurs at T_p. In this merged MP phase, the carriers are fully delocalized and the energy is reduced appreciably. The concept of the magnetic polaron becomes meaningless below T_p.

To prove the above hypothesis, we have studied the percolation-spanning probability (p) of magnetic polarons as a function of temperature. The polaron size can be determined by the distribution of the carrier density. That is, if the carrier density of a connected site is higher than $1/\sqrt{e}$ of the maximum density in the lattice, the site is regarded as a part of the polaron. As illustrated in the inset of Fig. 4, the percolation is defined by the existence of a large polaron that spans the whole lattice. When the percolation occurs, the polaron size diverges. The percolation-spanning probability p was calculated from $p=N_p/N_{\mathrm{MCS}}$, where N_{MCS} and N_p represent the total number of Monte-Carlo steps and the number of percolated states at each temperature, respectively.

Figure 4 provides the percolation-spanning probability as a function of temperature for the magnetic-polaron systems with $N_c=1$ and $N_c=5$. Upon cooling, the high-temperature unpercolated state transforms into the percolated state. Although the transition is rather broad due to the limited lattice size, it is clear that the percolation occurs not at T_C, but at T_p. It becomes more evident in the temperature derivative of the percolation-spanning probability dp/dT. As shown in the lower panels of Fig. 4, the minima of dp/dT appear exactly at T_p. This feature is also observed for other carrier densities ($N_c=2,3,4$) and for smaller lattices (6×6 and 4×4 square lattices), reflecting the robustness of the result. Therefore, we can conclude that the specific-heat maximum at T_p represents the occurrence of the polaron percolation, which takes place after the magnetic transition.

This conclusion is consistent with the recent muon-spin rotation measurement,[37] which provides evidence for the existence of magnetic-phase separation between T_C and T_p. They observed that the magnetic-phase separation disappears below T_p. Obviously, the present conclusion is distinct from the proposal by Süllow *et al.*[23] Based on the two-peak struc-

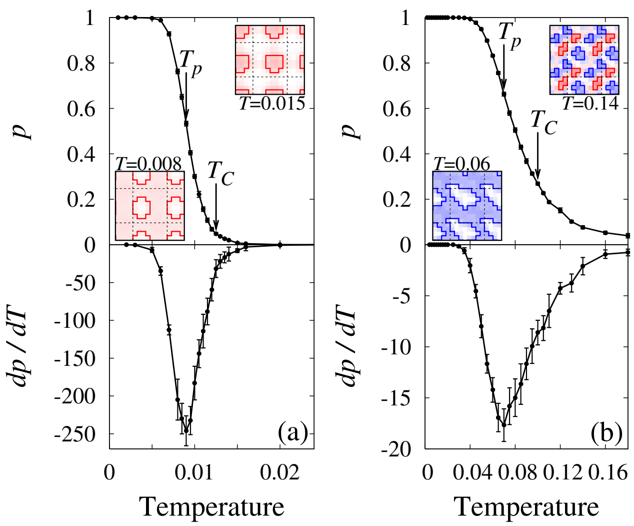

FIG. 4. (Color online) (a) The percolation-spanning probability p (upper panel) and the temperature derivative of that (lower panel) as a function of temperature for $N_c=1$ ($n=0.0156$). The percolation temperature T_p and the magnetic-phase transition temperature T_C obtained from Fig. 2 are also indicated in the upper panel. (b) The same as (a) but for $N_c=5$ ($n=0.0781$). The insets in (a) and (b) show examples of the unpercolated ($T>T_p$) and percolated ($T<T_p$) states. In the percolated state, magnetic polarons are merged into one magnetic-polaron cluster of infinite size. The periodic boundary condition was assumed.

ture in $d\rho/dT$, they proposed that the percolation occurs at $T_{C1}=15.5$ K and the bulk magnetic transition occurs at $T_{C2}=12.6$ K. The existence of the two-peak structure in $d\rho/dT$ is not well manifested in the calculation, and so it is difficult to identify T_{C2}. Nevertheless, the present calculation ascertains that the magnetic-phase transition temperature T_C is higher than the percolation-transition temperature T_p.

Note that the deviation of T_p from T_C is observed uniquely in EuB_6. In other magnetic-polaron systems such as doped perovskite manganites, pyrochlore $Tl_2Mn_2O_7$, and EuO, the peak of the magnetic specific heat is located at T_C.[38–41] It is likely that other interactions are operative in the above systems to smear out the magnetic-polaron effect. For doped perovskite manganites, the lattice effect is thought to play a more crucial role in the magnetic-phase transition than the magnetic-polaron effect.[9] In the case of low carrier-density systems, $Tl_2Mn_2O_7$ and EuO, it has been pointed out that the magnetic polaron alone cannot account for their high T_C's, and so other types of magnetic interactions, such as the FM superexchange or the FM indirect exchange, should be invoked.[40,42–44] Therefore, for the above systems, more studies on the magnetic-polaron model incorporating other types of interactions are requested. This fact highlights again the merit of EuB_6 as an ideal system to study the magnetic-polaron effect.

IV. CONCLUSION

We have demonstrated that the specific heat in the magnetic-polaron system has its maximum at T_p, where the polaron percolation occurs. The polaron percolation takes place below the magnetic-phase transition temperature T_C. The Fisher-Langer relation does not hold in the magnetic polaron system. The behavior of calculated C and χ based on the magnetic-polaron model is quite consistent with the experimental behavior in EuB_6, indicating that the anomalous magnetic and thermal properties of EuB_6 near T_C are explained well by the magnetic-polaron model.

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