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Monte Carlo study of magnetic resistivity in semiconducting MnTe

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We investigate in this paper properties of the spin resistivity in magnetic semiconducting MnTe of the NiAs structure. MnTe is a crossroad semiconductor with a large band gap. It is an antiferromagnet with a Néel temperature around 310 K. Due to this high Néel temperature, there are many applications using its magnetic properties. The method we use here is Monte Carlo simulation, in which we take into account the interaction between itinerant spins and lattice Mn spins. Our results show a very good agreement with experiments on the shape of the spin resistivity near the Néel temperature.

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I. INTRODUCTION

Spin resistivity in materials has been the subject of intensive studies both experimentally and theoretically for more than five decades. Experiments have been performed to determine the spin resistivity ρ in many magnetic materials, from metals to semiconductors. The rapid development of the field is due mainly to its many applications, in particular, in spintronics. One interesting aspect of magnetic materials is the existence of a magnetic phase transition from a magnetically ordered phase to the paramagnetic (disordered) state. Depending on the material, ρ can show a sharp peak at the magnetic transition temperature T_C,[1] or just a change of its slope, or an inflexion point. The latter case gives rise to a peak of the differential resistivity $d\rho/dT$.[2,3] Very recent experiments, such as those performed on ferromagnetic $SrRuO_3$ thin films,[4] Ru-doped induced ferromagnetic $La_{0.4}Ca_{0.6}MnO_3$,[5] antiferromagnetic ϵ-$(Mn_{1-x}Fe_x)_{3.25}Ge$,[6] semiconducting $Pr_{0.7}Ca_{0.3}MnO_3$ thin films,[7] superconducting $BaFe_2As_2$ single crystals,[8] and $La_{1-x}Sr_xMnO_3$[9] compounds, show different forms of anomaly of the magnetic resistivity at the magnetic phase transition temperature.

The properties of the total resistivity stem from different kinds of diffusion processes: scattering of the itinerant spins by phonons, by lattice magnons, by impurities and defects, etc. Each contribution has, in general, a different temperature dependence. Let us summarize the most important contributions at a low temperature (T) in the expression

$$\rho(T) = \rho_0 + AT^2 + BT^5 + C \ln \frac{\mu}{T}, \qquad (1)$$

where A, B, and C are constants. The first term is T independent; the second term, proportional to T^2, represents the scattering of itinerant spins at low T by lattice spin waves. Note that the resistivity caused by a Fermi liquid is also proportional to T^2. The T^5 term corresponds to low-T resistivity in metals. This is due to the scattering of itinerant electrons by phonons. Note that at high T, metals show a linear-T dependence. The ln term is the resistivity due to the quantum Kondo effect at very low T. For the magnetic contribution to the total resistivity, the T^2 term has been obtained from the magnon scattering by Kasuya.[10] However, at high T, in particular, in the region of the phase transition, much less is known. The general idea that the magnetic resistivity is a function of the spin-spin correlation was introduced by de Gennes and Friedel.[11] According to this idea, the magnetic resistivity should behave as the magnetic susceptibility, thus it should diverge at T_C. Fisher and Langer[12] and Kataoka[13] have suggested that the range of spin-spin correlation changes the shape of ρ near the phase transition. The resistivity due to magnetic impurities has been calculated by Zarand *et al.*[14] as a function of Anderson's localization length. This parameter expresses, in fact, the kind of correlation sphere induced around each impurity. Their result that the resistivity peak depends on this parameter is in agreement with the spin-spin correlation idea. In our previous works[15–17] we have studied the spin current in ferromagnetic thin films by Monte Carlo (MC) simulations. The behavior of the spin resistivity as a function of T has been shown to be in agreement with the main experimental features and theoretical investigations mentioned above. We have introduced in these works the picture of scattering of itinerant spins by magnetic defect clusters which are known to be formed in the transition temperature region. The size of each defect cluster expresses a kind of correlation between spins.

In antiferromagnets much less is known because very few theoretical investigations have been carried out. Haas[18] has shown that while in ferromagnets the resistivity ρ shows a sharp peak at the magnetic transition of the lattice spins, in antiferromagnets there is no such a peak. The alternate change of sign of the spin-spin correlation with distance may have something to do with the absence of a sharp peak.

In this paper, we are interested in antiferromagnetic MnTe, a well-studied semiconductor with numerous applications due to its high Néel temperature. The pure MnTe has either a zinc-blende structure[19] or a hexagonal NiAs one.[20] We confine ourselves to the case of the hexagonal structure. For this case, the Néel temperature is $T_N = 310$ K.[20] Hexagonal MnTe is a crossroad semiconductor with a big gap (1.27 eV) and a room-temperature carrier concentration of $n = 4.3 \times 10^{17}$ cm^{-3}.[21,22] Without doping, MnTe is nondegenerate. In doped cases,[23–26] band tails created by doped impurities can more or less cover the gap. But these systems, which are disordered by doping, are not the purpose of our present study. So, in the following we study only pure MnTe. The behavior of the spin resistivity ρ in MnTe as a function of T has been shown experimentally.[27–31] In our previous paper,[32] we carried out a theoretical study with Boltzmann's equation using numerical data for a cluster distribution obtained by MC simulations. We could only compare our result with old experimental data available at that time below the transition temperature.[27] After the publication of our paper, new experimental results of He *et al.*[31] become available for the whole range of temperature for pure MnTe. This motivated the present work.

In this paper, we use the same model as in our previous work[32] for MnTe. But unlike that previous paper, which used the approximate Boltzmann's equation, we use here direct MC simulations without recourse to approximations. In addition, we take into account the lattice magnetic relaxation time in the calculation of the spin resistivity. The tuning of the relaxation time allows an excellent agreement with the experimental resistivity of MnTe as shown below.

In Sec. II, we present our model and describe our MC method. Results are reported and compared with experimental resistivity of MnTe in Sec. III. Concluding remarks are given in Sec. IV.

II. MODEL AND METHOD

A. Model

The hexagonal NiAs-type structure of MnTe is shown in Fig. 1.[20] It is composed of ferromagnetic xy hexagonal planes antiferromagnetically stacked in the c direction. The nearest-neighbor (NN) distance in the c direction is $c/2 \simeq 3.36$ Å shorter than the in-plane NN distance, which is $a = 4.158$ Å. Neutron scattering experiments show that the main exchange interactions between Mn spins in MnTe are (i) interaction between NNs along the c axis with the value $J_1/k_B = -21.5 \pm 0.3$ K, (ii) ferromagnetic exchange $J_2/k_B \approx 0.67 \pm 0.05$ between in-plane neighboring Mn (they are next NN by distance), and (iii) third NN antiferromagnetic interaction $J_3/k_B \simeq -2.87 \pm 0.04$ K.

The interactions J_1, J_2, and J_3 are indicated in Fig. 1. In addition, the spins are lying in the xy planes perpendicular to the c direction, with a small in-plane easy-axis anisotropy D.[20] We note that the values of the exchange integrals given above have been deduced from experimental data by fitting with a formula obtained from a free spin-wave theory.[20] Other fittings with mean-field theories give slightly different values.[21] Therefore, care should be taken when using values deduced from experimental data, keeping in mind that they depend on the models used and approximations involved in the fitting.

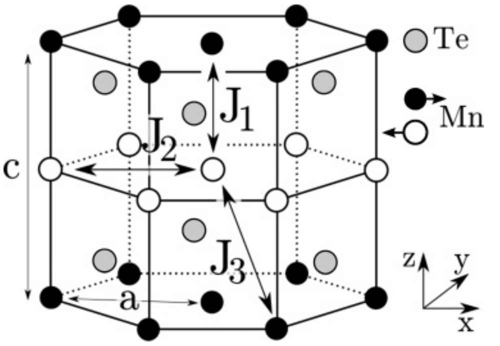

FIG. 1. Structure of MnTe of the NiAs type. Antiparallel spins are shown by black and white circles. The nearest-neighbor (NN) interaction is labeled J_1; the next NN interaction, J_2; and the third NN interaction, J_3.

The lattice Hamiltonian is given by

$$\mathcal{H} = -J_1 \sum_{(i,j)} \mathbf{S}_i.\mathbf{S}_j - J_2 \sum_{(i,m)} \mathbf{S}_i.\mathbf{S}_m - J_3 \sum_{(i,k)} \mathbf{S}_i.\mathbf{S}_k - D \sum_i \left(S_i^x\right)^2, \qquad (2)$$

where $\mathbf{S}_i$ is the Heisenberg spin at lattice site i, $\sum_{(i,j)}$ is made over the NN spin pairs $\mathbf{S}_i$ and $\mathbf{S}_j$ with interaction J_1, and $\sum_{(i,m)}$ and $\sum_{(i,k)}$ are made over the next-NN and third-NN pairs with interactions J_2 and J_3, respectively. $D > 0$ is an anisotropy constant which favors the in-plane x easy-axis spin configuration. The Mn spin is experimentally known to be of the Heisenberg model with magnitude $S = 5/2$.[20]

The interaction between an itinerant spin and surrounding Mn spins in semiconducting MnTe is written as

$$\mathcal{H}_i = -\sum_n J(\vec{r} - \vec{R}_n)\mathbf{s} \cdot \mathbf{S}_n, \qquad (3)$$

where $J(\vec{r} - \vec{R}_n) > 0$ is a ferromagnetic exchange interaction between the itinerant spin $\mathbf{s}$ at $\vec{r}$ and the Mn spin $\mathbf{S}_n$ at lattice site $\vec{R}_n$. The sum on lattice spins $\mathbf{S}_n$ is limited at some cutoff distance as discussed later. We suppose that $J(\vec{r} - \vec{R}_n)$ is weak enough to be considered a perturbation to the lattice Hamiltonian:

$$J(\vec{r} - \vec{R}_n) = I_0 \exp[-\alpha(\vec{r} - \vec{R}_n)], \qquad (4)$$

where I_0 and α are constants. We choose $\alpha = 1$ for convenience. The choice of I_0 should be made so that the interaction $\mathcal{H}_i$ yields an energy much lower than the lattice energy due to $\mathcal{H}$ (see discussion of the choice of variables in Refs. 33 and 34).

Since in MnTe the carrier concentration is $n = 4.3 \times 10^{17}$ cm^{-3}, low with respect to the concentration of its surrounding lattice spins, $\simeq 10^{22}$ cm^{-3}, we do not take into account the interaction between itinerant spins. The contribution of that interaction to the energy of an itinerant spin is negligible.

B. Method

We perform MC simulations on a sample of dimension $L \times L \times L$, where L is the number of MnTe cells in the x, y, and z directions. Note that each cell contains two Mn atoms and two Te atoms (see Fig. 1). Periodic boundary conditions are applied in all directions. The itinerant electrons move in the system under an electric field $\vec{\epsilon}$ applied along the x direction,

$$\mathcal{H}_E = -e\vec{\epsilon}.\vec{\ell}, \qquad (5)$$

where $-e$ is the charge of the electron and $\vec{\ell}$ the displacement vector of the electron. Each electron spin $\mathbf{s}$ of magnitude 1/2 interacts with neighboring lattice Mn spins within a sphere of radius D_1 according to Eq. (3). Note that the periodic boundary conditions are applied to the lattice so that electrons near (and at) the system boundaries "see" the same environment as if they are deep inside the system. For that purpose only the lattice is "translated" in all directions by the periodic boundary conditions, while electrons that leave the system at one end will re-enter the system at the opposite end so that the total number does not change. They are never outside the system. The electric field acts on them inside the system. We take in the following the Ising model for electron spin. In doing so, we neglect quantum effects, which are, of course, important at very low temperatures but not in the transition region at room temperature, where we focus our attention.

The MC technique we use for the transport is a multistep averaging which has been shown to reduce statistical fluctuations efficiently.[33,34] The reader is referred to our early works for a detailed description of our method as well as the effects of changing physical parameters such as D_1 in antiferromagnets.

We briefly summarize our method here. The procedure of our simulation consists in equilibrating first the lattice at a given temperature T without itinerant electrons. When equilibrium is reached, we randomly add N_0 polarized itinerant spins into the lattice. Each itinerant electron interacts with lattice spins in a sphere of radius D_1 centered at its position. We next equilibrate the itinerant spins using the following updating: we calculate the energy E_{old} of an itinerant electron taking into account the spin-lattice interaction described above. Then we perform a trial move of length ℓ taken in an arbitrary direction with random modulus in the interval $[R_1,R_2]$, where $R_1 = 0$ and $R_2 = a$, a being the lattice constant. Note that the move is rejected if the electron falls in a sphere of radius r_0 centered at a lattice spin or at another itinerant electron. This excluded space emulates the Pauli exclusion. We calculate the new energy E_{new} and use the Metropolis algorithm to accept or reject the electron displacement. We choose another itinerant electron and begin the procedure again. When all itinerant electrons are considered, we say that we have made an MC sweeping, or one MC step per spin We have to repeat a large number of MC steps per spin to reach a stationary transport regime. We then perform the averaging to determine physical properties such as magnetic resistivity, electron velocity, and energy as functions of temperature.

We emphasize here that in order to have sufficient statistical averages for microscopic states of both the lattice spins and the itinerant spins, we use the following procedure: after averaging the resistivity over N_1 steps for "each" lattice spin configuration, we again thermalize the lattice with N_2 steps in order to take another disconnected lattice configuration. Then we take back the averaging of the resistivity for N_1 steps for the new lattice configuration. We repeat the cycle, $N_1 + N_2$, N_3 times, usually several hundred thousand times. The total MC steps for averaging is about 4×10^5 steps per spin at each temperature in our simulations. This procedure greatly reduces the thermal fluctuations observed in our previous work.[16,17] Of course, the larger N_1 and N_3 are, the better the statistics becomes. The question is, What is the correct value of N_1 for averaging with each lattice spin configuration at a given T? This question is important because it is related to the relaxation time τ_L of the lattice spins compared to that of the itinerant spins, τ_I. The two extreme cases are as follows. (i) $\tau_L \simeq \tau_I$; one should take $N_1 = 1$, namely, the lattice spin configuration should change with each move of itinerant spins. (ii) $\tau_L \gg \tau_I$; in this case, itinerant spins can travel in the same lattice configuration many times during the averaging. In a word, this technique consists in changing the lattice spin configuration as often as the temperature-dependent relaxation time allows. The relaxation time of the lattice spin system is expressed as[35]

$$\tau_L = \frac{Q}{|1 - T/T_N|^{z\nu}}, \tag{6}$$

where Q is a constant, ν the correlation critical exponent, and z the dynamic exponent. From this expression, we see that as T tends to T_N, τ_L diverges. This phenomenon is known as the critical slowing-down. For the Heisenberg model, $z\nu = 1.38$ ($\nu = 0.704$ and $z = 1.97$).[36] We have previously shown that τ_L strongly affects the shape of ρ.[37] However, as shown in the following, the choice of Q is rather physically natural: we know that far in the paramagnetic phase, the lattice spins are strongly disordered. They fluctuate rapidly in the thermal bath. It is reasonable therefore to assume that $\tau_L = 1$ (smallest value in MC time) at $T = 2T_N$. With this assumption, we have $Q = 1$ as seen from Eq. (6). We use this choice in the following.

III. RESULTS

The resistivity $\rho(T)$ is calculated by counting the number of itinerant spins $n(T)$ that cross a unit surface perpendicular to the electric field, per MC time unit. In order to take spatial averages, we put such surfaces at three positions at equal distance along the sample length in the x direction. The resistivity is given by

$$\rho(T)^{-1} = \frac{n(T)e^2\tau_I}{m}, \tag{7}$$

where m is the electron mass.

We have calculated the spin resistivity of hexagonal MnTe using the exchange integrals taken from Ref. 20. These values are given above. As stated before, the values of the exchange interactions deduced from experimental data depend on the model Hamiltonian, in particular, on the spin model and the approximations. We emphasize here, again, that in semiconducting MnTe we neglect the interaction between itinerant electrons due to their low concentration. They interact only with the lattice spins. In the calculation of the resistivity $\rho(T)$, we count, among N_0 itinerant spins, only the number of electrons which cross a unit surface perpendicular to the electric field, per unit time. This number, $n(T)$, depends on the temperature. Using $n(T)$, one obtains $\rho(T)$ by Eq. (7).

Note that in semiconductors, the carrier concentration depends on T. In a simple theoretical model for a semiconductor, we have the expression $n(T) = 2[m_e k_B T/2\pi\hbar^2]^{3/2} \exp[-(E_g - E_F)/k_B T]$, where m_e is the electron effective mass in the conduction band, E_g the energy gap, and E_F the Fermi level.[38] But this theoretical formula is from a first approximation. It does not correspond to real semiconductors, where, unfortunately, no correct formulas are available. In our model, the temperature dependence of $n(T)$ comes from the simulation; it is not introduced by hand. As stated above, $n(T)$ is counted in the simulation at each T. So, our method of direct computation of $n(T)$ without approximation is very helpful for comparison with experiments.

The result for the spin resistivity is shown in Fig. 2. Our result reproduces the experimental data $n(T) \simeq 4.3 \times 10^{17}$ cm^{-3} at room temperature. Using the experimental data in Fig. 2, the reader can obtain $n(T)$ at any other

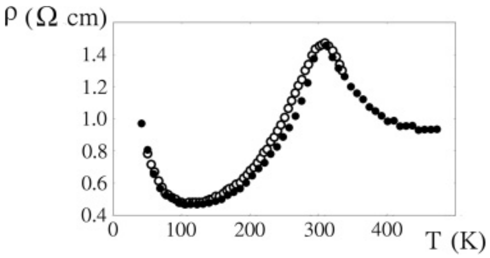

FIG. 2. Spin resistivity ρ versus temperature T. Filled circles are from Monte Carlo simulation; open circles are experimental data taken from He *et al.*[31] Parameters used in the simulation are $J1 = -21.5$ K, $J_2 = 2.55$ K, $J_3 = -9$ K, $I_0 = 2$ K, $D = 0.12$ K, $D_1 = a = 4.158$ Å, $E = 2 \times 10^5$ V/m, and $L = 30$.

temperature. Furthermore, with the value of ρ, we obtain a relaxation time of itinerant spin equal to $\tau_I \simeq 0.1$ ps, and a mean free path equal to $\bar{l} \simeq 20$ Å, at the critical temperature.

Let us make some comments on the result shown in Fig. 2.

(1) With J_3 slightly larger in magnitude than the value deduced from experiments, we find $T_N = 310$ K.

(2) ρ shows a pronounced peak, in excellent agreement with experiments.

(3) Note that the shape of the peak depends on Q. The value we used to obtain that agreement is $Q = 1$. A discussion of this value is given folllowing Eq. (6)

(4) In the temperature regions below $T < 140$ K and above T_N, the MC result is in excellent agreement with experiment, unlike in our previous work[32] using Boltzmann's equation.

(5) In the region 140 K $< T < T_N$ the MC result of ρ is slightly smaller than the experimental data. In the search for an explanation, we see that the magnetization obtained by the MC simulation, though in good agreement with experiments at low T and yielding the precise value of T_N, is slightly smaller than the experimental one in the intermediate temperature region. This is shown in Fig. 3. The magnetization deficit may be due to the fact that the magnetic anisotropy was too small, at $D = 0.12$ K, taken from Ref. 20; it is not strong enough to enhance the magnetization.

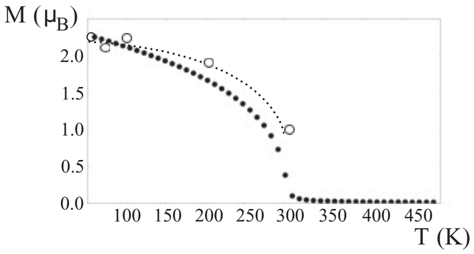

FIG. 3. Magnetization M (in units of μ_B) versus temperature T (in units of K). Filled circles are the results of Monte Carlo simulation; open circles are experimental data taken from Efrem D'Sa *et al.*[30] Parameters are the same as those given in the caption to Fig. 2.

(6) MnTe is a nondegenerate semiconductor with a large gap. In such a case, the Fermi level is in the gap, the conduction electron concentration is low (due to no doping), the Maxwell-Boltzmann distribution can be used for electrons in the conduction band instead of the Fermi distribution. As a consequence, quantum effects are not strong or are even absent at moderate temperatures. This may explain the good agreement of our result using the classical spin model for $T > 50$ K.

IV. CONCLUSION

We have shown in this paper the MC results of the spin resistivity ρ as a function of temperature in MnTe. We have taken into account the main interaction which governs the resistivity behavior, namely, the interaction between itinerant spins and lattice Mn spins. Our result is in agreement with experiments: it reproduces the correct Néel temperature as well as the shape of the peak at the phase transition. Note that the theory of Haas[18] predicts the absence of a peak in ρ in the temperature region of the phase transition for antiferromagnetic MnTe. We, finally, emphasize that our excellent agreement was possible because, most importantly, we have correctly taken into account the temperature dependence of the lattice spin relaxation time in the simulation.[37]

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