Large magnetoresistance and critical spin fluctuations in GdI_2

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(Received 12 February 2001; published 24 July 2001)

The effect of the large negative magnetoresistance in layered ferromagnetic metal GdI_2 has been explained on the basis of the s-f exchange model. Assuming the Ornstein-Zernike form of the spin-spin correlation function for the two-dimensional case, we calculate the resistivity in the vicinity of the ferromagnetic transition. The influence of the magnetic field was taken into account via the generalized dynamical-scaling approach and molecular-field approximation. The results are found to be in good agreement with experiment.

DOI: 10.1103/PhysRevB.64.064425 PACS number(s): 75.70.Pa, 75.50.Cc, 75.40.Gb

A layered magnetic system exhibits many interesting and promising properties. Perhaps the most exciting among them is the effect of the large negative magnetoresistance observed in manganites[1] and rare-earth-based layered materials[2,3] associated with a significant decrease in the resistivity on applying a magnetic field. Despite a large number of investigations the underlying physics of this phenomenon in manganites is still far from being understood. Furthermore, the situation there is more complicated due to the fact that MnO_6 centers are Jahn-Teller active. This introduces lattice degrees of freedom that complicate the physical picture.

It is therefore instructive to investigate materials that show large negative magnetoresistance where most of the physical properties are determined by magnetic interactions alone. One particular example we will consider here is the rare-earth-based layered ferromagnetic metal GdI_2 that undergoes a transition to ferromagnetism close to room temperature.[2,4] GdI_2 has a hexagonal-close-packed structure with $P6_3/mmc$ point-group symmetry. It comprises GdI_2 sheets with the Gd in the centers of the trigonal I_6 prisms formed by iodine ions. In addition, each Gd atom is surrounded by six other Gd atoms in the plane. The lattice parameters are $a=4.0775(4)$ Å and $c=15.041(1)$ Å.

The electrical resistivity versus temperature shows a broad anomaly centered at the Curie temperature T_c. This anomaly shifts towards higher temperatures when a magnetic field is applied and almost flattens at 7 T. The magnetoresistance exceeds the value of 60% at room temperature and high magnetic field.[2]

It is of general interest to investigate this compound in order to understand the nature of the large negative magnetoresistance in magnetic-layered rare-earth-based systems. The non-Jahn-Teller nature of the GdI_6 center and $5d$ character of conduction electrons there allows to exclude the effect of the strong electron-phonon interaction on the magnetoresistance properties. On the other hand, having $4f^7$ localized spins with $S=7/2$ and $5d$-conduction electrons as found in local-spin-density-approximation (LSDA) calculations,[2] the present case seems to resemble that of manganites with a localized t_{2g} shell with $S=3/2$ and $3d$-e_g conduction electrons. This suggest some similarities in the mechanism responsible for the large negative magnetoresistance phenomenon in GdI_2. Note, however, that in GdI_2 the s-f exchange interaction is not of the ferromagnetic Hund's type and is rather expected to be antiferromagnetic.

In this paper we show that the anomalous peak of the electrical resistivity around T_c in GdI_2 can be explained as a result of a strong scattering of conduction $5d$ electrons by localized $4f^7$ electrons with $S=7/2$ due to the specific topology of the Fermi surface of the conduction band. This results from the quasi-two-dimensional band structure[2] and the remarkable fact that conduction electrons do not occupy a broad s band like in Gd metal but a narrow $5d_{z^2}$ band. The presence of a weak magnetic field causes changes in the damping of spin excitations and introduces a nonzero magnetization above T_c. This results in the suppression of the scattering mechanism. In the case of large magnetic fields the localized spins are aligned along the field direction and spin fluctuations are strongly suppressed. This leads to the absence of scattering by the magnetic moments and a decrease of the magnetic resistivity.

In our theoretical analysis of the magnetoresistive properties of GdI_2 we start from the two-dimensional tight-binding model for $5d_{z^2}$-conduction electrons in the basal plane of a hexagonal lattice,

$$H_0=\sum_{\mathbf{k}\sigma}\epsilon_{\mathbf{k}}c^{+}_{\mathbf{k}\sigma}c_{\mathbf{k}\sigma}, \tag{1}$$

where

$$\epsilon_{\mathbf{k}}=2t[\cos(\mathbf{ka})+\cos(\mathbf{kb})+\cos\{\mathbf{k}(\mathbf{a}-\mathbf{b})\}] \tag{2}$$

is the energy dispersion with t being the hopping term between nearest neighbors. The fundamental translation vectors in the basal plane are $\mathbf{a}=a(1,0)$ and $\mathbf{b}=a(1/2,\sqrt{3}/2)$.

In Fig. 1 the results for ϵ_k of a tight-binding calculation together with a resulting Fermi surface are shown. The agreement with LSDA calculations was found for $t=180$ meV.[5] One can notice the following important details. The conduction electrons at the Fermi surface in GdI_2 lie much closer to a Γ point of the Brillouin zone (BZ) than in any ordinary rare-earth metal like Gd, for example. Therefore, one would expect that conduction electrons are strongly

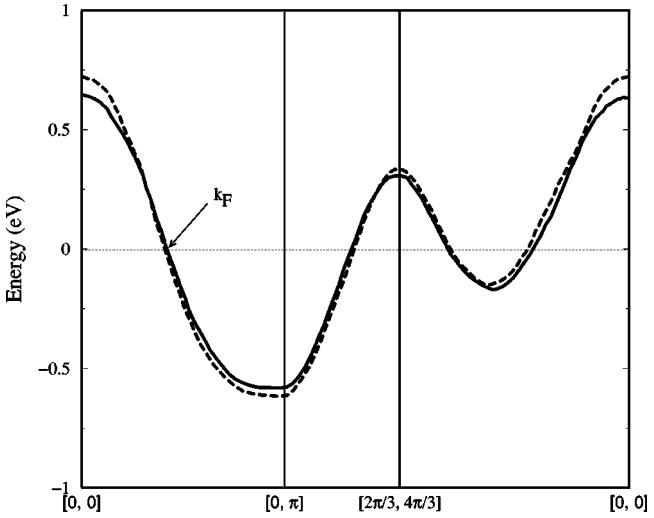

FIG. 1. Calculated tight-binding energy dispersion ϵ_k for the triangular plane of GdI_2 (solid curve) for $t=180$ meV along the path $(0,0)\rightarrow(\pi,0)\rightarrow(4\pi/3,2\pi/3)\rightarrow(0,0)$. For comparison, the LSDA results (Ref. 2) are also shown (dotted curve). The arrow indicates the position of Fermi radius in **k** space with respect to a Γ point of the first Brillouin zone.

coupled to the ferromagnetic fluctuations ($\mathbf{q}\approx 0$) that play an important role close to the Curie temperature.

The interaction between conduction $5d_{z^2}$ and localized $4f^7$ electrons can be taken into account via a phenomenological *s-f*-like exchange model,

$$H_{s\text{-}f}=\sum_{\langle \mathbf{k},\mathbf{k}'\rangle} I(\mathbf{k},\mathbf{k}')e^{-i(\mathbf{k}-\mathbf{k}')\mathbf{R_n}}\mathbf{S_n}\mathbf{s_{kk'}}, \tag{3}$$

where $\mathbf{S}=7/2$ is the total spin of localized electrons and

$$s^z_{\mathbf{k}'\mathbf{k}}=\frac{1}{2}[c^+_{\mathbf{k}'\uparrow}c_{\mathbf{k}\uparrow}-c^+_{\mathbf{k}'\downarrow}c_{\mathbf{k}\downarrow}],$$

$$s^+_{\mathbf{k}'\mathbf{k}}=c^+_{\mathbf{k}'\uparrow}c_{\mathbf{k}\downarrow},$$

$$s^-_{\mathbf{k}'\mathbf{k}}=c^+_{\mathbf{k}'\downarrow}c_{\mathbf{k}\uparrow}, \tag{4}$$

are spin operators of conducting electrons in the second-quantization form. The constant of the *s-f* exchange, $I(\mathbf{k})$, is considered to be small enough with respect to the bandwidth of conduction electrons.

We can explore here the perturbation theory, which is well investigated in the case of $I\ll t$.[6] For example, the most interesting second-order perturbation results in the effective Ruderman-Kittel-Kasuya-Yasida (RKKY) interaction of localized spins via conduction electrons. This leads also to a ferromagnetic Curie temperature of localized electrons close to room temperature in GdI_2.

The electrical resistivity of GdI_2 can be analyzed using the following formula:

$$\rho=\rho_{res}+\rho_{ph}+\rho_m, \tag{5}$$

where ρ_{res} is the residual resistivity due to impurities, ρ_{ph} is the lattice resistivity due to a phonon and it varies roughly linearly with temperature above the Debye temperature, and ρ_m is the magnetic resistivity. The anomalies in the electrical resistance around Curie temperature in GdI_2 are certainly due to magnetic *s-f* exchange interaction. At 0 K in a ferromagnetic metal, all the spins are aligned, the system is completely periodic, and the magnetic resistivity is equal to zero. If temperature increases, the spins would fluctuate around their average value resulting in the nonzero scattering of conduction electrons by localized spins. Since the fluctuations become very strong around T_c, one would expect the strongest scattering there.

The differential cross section that determines the magnetic resistivity can be calculated in Born approximation and one obtains[7]

$$\frac{d\sigma}{d\Omega}=\frac{\sigma_0}{4\pi}\sum_n e^{i\mathbf{q}\cdot\mathbf{R}_n}\left[\frac{\langle \mathbf{S}_0\mathbf{S}_n\rangle-\langle \mathbf{S}\rangle^2}{S(S+1)}\right] \tag{6}$$

with

$$\sigma_0=\frac{1}{4\pi}\left[\frac{mI}{\hbar}\right]^2 S(S+1), \tag{7}$$

where m is an effective mass, $\langle \mathbf{S}_0\mathbf{S}_n\rangle$ is a spin-spin correlation function, and $\langle \mathbf{S}\rangle$ is the average value of the spin in the ordered phase. The main result of expression (6) is that the temperature dependence of the magnetic resistivity in the paramagnetic phase is determined only by the corresponding temperature dependence of the spin-spin correlation function of the localized electrons. In the ferromagnetic region $\langle \mathbf{S}\rangle$ increases continuously to reach S at $T=0$. Of course, at low enough temperatures Eq. (6) is not valid due to the neglect of the additional scattering of carriers on the spin waves, which is beyond the scope of the present analysis.

The calculation of the spin-spin correlation function of localized spins in layered GdI_2 requires obeying the Mermin-Wagner theorem. This will invalidate application of the ordinary molecular-field approximation (MFA). However, due to three-dimensional coupling one would again have a nonzero Curie temperature. Therefore it is useful to reexamine MFA results in order to see the importance of the strong fluctuations around T_c and their effect on the conduction electrons already at this stage.

In ferromagnetic metals like GdI_2 or ordinary Gd we have above the Curie temperature $\langle \mathbf{S}\rangle=0$. The short-range fluctuations of the spins are directly included in MFA by introducing the molecular field $\mathbf{H}_n$ at point n caused by the nearest-neighboring spins $S_{n'}$ in the following manner

$$\langle \mathbf{S}_n\rangle=SB_s\left[\frac{\mathbf{S}\mathbf{H}_n}{k_BT}\right], \tag{8}$$

where $B_s(x)$ is a Brillouin function. The molecular field $\mathbf{H}_n$ is equal to

$$\langle \mathbf{H}_n\rangle=\beta\frac{1}{Z_n}\sum_{n'\neq n}\langle \mathbf{S}_{n'}\rangle \tag{9}$$

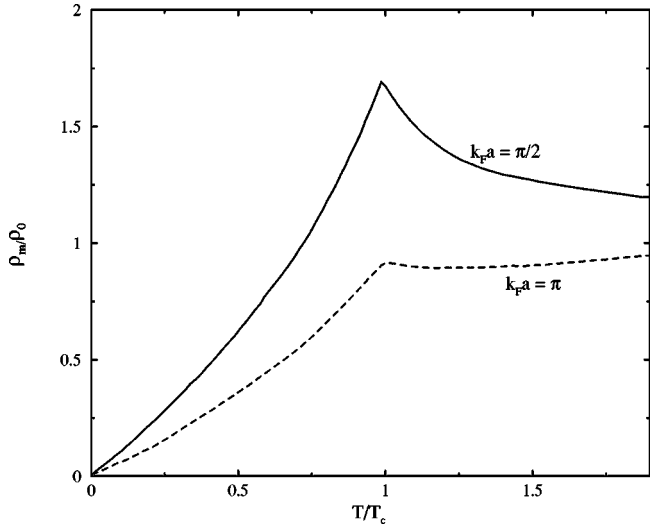

FIG. 2. Calculated temperature dependence of the normalized magnetic resistivity in MFA approximation for $k_F a=\pi$ (dotted curve) and $k_F a=\pi/2$ (solid curve). ρ_0 is the value of magnetic resistivity above T_c without correlations (Ref. 8).

with β being a constant and Z_n being the number of the nearest neighbors of n. After a few straightforward calculations one arrives at the following result:[8]

$$\sum_n \frac{\langle \mathbf{S}_0 \mathbf{S}_n \rangle}{S(S+1)} e^{i\mathbf{q}\cdot\mathbf{R}_n} = \left[1 - \frac{T_c \sin\left(2k_F a \sin\frac{\theta}{2} \right)}{T 2k_F a \sin\frac{\theta}{2}} \right]^{-1}, \tag{10}$$

where k_F is a Fermi wave vector of conduction electrons, a is the lattice constant in the plane and θ is the angle between $\mathbf{k}$ and $\mathbf{k}'$. Inserting Eqs. (8) and (10) into the scattering cross section (6) we can calculate the resistivity as

$$\rho_m = \frac{m}{z e^2 \tau}, \tag{11}$$

where z is the number of electrons per atom and τ the relaxation time given by

$$\frac{1}{\tau} = \frac{\hbar k_F}{m} 2\pi \int_0^\pi \sin\theta d\theta (1-\cos\theta) \frac{d\sigma}{d\Omega}. \tag{12}$$

One can analyze now the temperature dependence of the magnetic resistivity for different ferromagnetic metals. The main difference between Gd and GdI_2 in the MFA is that in the first case the conduction electrons belong to a broad s band but in the second case they occupy a narrow $5d$ band. Therefore in the case of Gd the value of $k_F a$ appearing in Eq. (9) is about or slightly bigger than π. On the other hand, this constant in GdI_2 can be obtained from our tight-binding approach and it is approximately equal to $\pi/2$. In Fig. 2 we show the results of normalized magnetic resistivity versus temperature for both parameters. As a consequence in the case of GdI_2, the critical spin fluctuations around T_c affect the magnetic part of the resistivity much stronger. This leads to the anomalous peak around T_c for GdI_2 whereas in the case of Gd metal there is just a cusp reflecting the transition to the ferromagnetic state. The physical origin of this feature is due to the fact that in contrast to Gd, the carriers in GdI_2 at the Fermi level lie two times closer to a Γ point of the first BZ where the ferromagnetic fluctuations take place. Therefore, critical opalescence occurs around T_c resulting in the strong scattering of conduction electrons. We have to notice, however, that at small angles and long wavelength, the MFA approximation is no longer valid as it is seen from Eq. (10) and has to be improved.

In order to treat the behavior of the critical spin fluctuations of localized $4f^7$ electrons for the real layered structure of GdI_2 we use a following random-phase-approximation-like phenomenological ansatz for the spin susceptibility in the temperature region above T_c:

$$\chi(\mathbf{q},\omega) = \frac{\chi_{2d}(\mathbf{q})}{1 - J_\perp(\mathbf{q})\chi_{2d}(\mathbf{q})} \frac{i\gamma(\mathbf{q},\omega)}{\omega + i\gamma(\mathbf{q},\omega)}. \tag{13}$$

Here, $\gamma(\mathbf{q},\omega)$ is the damping function of the spin fluctuations, and $J_\perp(\mathbf{q})$ is the Fourier transform of the ferromagnetic exchange of localized spins between different layers. The denominator of Eq. (13) determines the ferromagnetic Curie temperature.

Generally the $\mathbf{q}$ dependence of $J_\perp(\mathbf{q})$ and $J_\parallel(\mathbf{q})$ are due to the RKKY interaction mechanism leading to an oscillating function. In the two-dimensional case it varies as $r^{-2}\sin k_F r$.[9] Simple analysis shows that the exchange between next-nearest neighbors is three times smaller than between nearest neighbors but it is still ferromagnetic. One has to assume also that $J_\perp(\mathbf{q})$ has its maximum at $\mathbf{q}=0$ resulting in a ferromagnetic transition at T_c. The Heisenberg type of the Hamiltonian with effective ferromagnetic exchange between neighboring $\mathbf{S}=7/2$ spins is the reasonable approximation.

The static part of the spin susceptibility in the plane, $\chi_{2d}(\mathbf{q})$, is assumed to have the Ornstein-Zernike form,

$$\chi_{2d}(\mathbf{q}) = \frac{\chi_{2d}(0)}{1 + \xi_{2d}^2 \mathbf{q}_\parallel^2} \tag{14}$$

with ξ_{2d} and $\chi_{2d}(0)$ being a magnetic correlation length and uniform spin susceptibility, respectively. The calculation of both quantities has to be done respecting the Mermin-Wagner theorem, i.e., the paramagnetic spin susceptibility and correlation length are divergent only at $T=0$ K. In particular, the following formula for the susceptibility was derived by several authors:[10–12]

$$\chi_{2d}(T) = C(T)^l \exp\left[\frac{C_1}{T}\right], \tag{15}$$

where the values of l, C, and C_1 depend on the level of approximation. For a two-dimensional ferromagnet on a triangular lattice with large spin $S=7/2$, one can use for example either Schwinger bosons[13] or the modified spin-wave theory by Takahashi.[12] Since the results of both approaches are identical, we use the latter.

We refer the reader for the technical aspects to the original work of Takahashi.[12] We notice only that within the usual Holstein-Primakoff transformations the Mermin-Wagner theorem can be taken into account via introducing the additional *constraint* of zero magnetization in the z direction. Therefore, a new density matrix is obtained by minimizing the free energy under this *constraint* with the help of a Lagrange multiplier μ. This results in a self-consistent equation for the chemical potential and new expressions for the thermodynamical characteristics like spin susceptibility and correlation length.[14]

For the ferromagnetic Heisenberg Hamiltonian on the triangular plane the calculation of spin susceptibility and correlation length is a straightforward extension of the original work for the square plane[12] with the only difference in energy dispersion for the spin waves,

$$\hbar\omega_{\mathbf{q}} = J(0)\left[6 - 2\cos q_x - 4\cos\frac{q_x}{2}\cos\frac{\sqrt{3}q_y}{2}\right]. \qquad (16)$$

Taking this into account we obtain the following expression for the spin susceptibility and correlation length:

$$\xi_{2d}(T) = \frac{1}{\sqrt{6}}\left[\frac{J_\parallel S}{T}\right]^{-1/2}\exp\left[\frac{8\pi\sqrt{3}J_\parallel S}{T}\right], \qquad (17)$$

$$\chi_{2d}(T) = \frac{1}{6\sqrt{3}SJ_\parallel}\exp\left[\frac{16\sqrt{3}J_\parallel S^2}{T}\right], \qquad (18)$$

where $S=7/2$. A comparison of the expressions (18) and (15) yields the constants $C=1/6\sqrt{3}SJ_\parallel$, $l=0$, and $C_1 = 16\sqrt{3}J_\parallel S^2$.

Substituting Eq. (18) into Eqs. (14) and (13) we obtain the Curie temperature as

$$k_B T_c \approx \frac{16\sqrt{3}J_\parallel S^2}{\ln\dfrac{6\sqrt{3}J_\parallel S}{J_\perp}}. \qquad (19)$$

This result is quite natural for a quasi-two-dimensional Heisenberg system and reflects the fact that a nonzero transition temperature is caused by the interplane coupling $J_\perp$.

We can estimate the ratio of $J_\perp/J_\parallel$ from measured high-temperature susceptibility and Curie transition temperature. Figure 3 displays the high-temperature reciprocal susceptibility and the inset shows the magnetization of a GdI_2 sample indicating a Curie temperature of 286(2) K (according to Arrotts plots) and a paramagnetic Curie-Weiss temperature Θ of 412(3) K. Using $\Theta = (2/3)S(S+1)\Sigma_i z_i J_i$ where z_i is the number of neighbors interacting with exchange constant J_i we arrive at effective $J_\parallel \approx 6$ K taking into account six in-plane metal-atom neighbors. Furthermore, using Eq. (19) we obtain also $J_\perp/J_\parallel \approx 0.03$.[15]

Let us now turn to the analysis of the damping function in Eq. (13). In general, $\gamma(\mathbf{q},\omega)$ is known only in various limiting cases. Since we are interested mainly in the close vicinity to a Curie transition temperature, we can use it in a form obtained earlier for the critical region,[16]

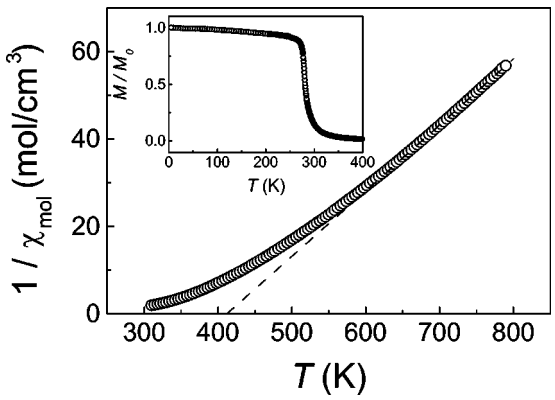

FIG. 3. Temperature dependence of the reciprocal magnetic susceptibility of GdI_2 measured in a field of 0.1 T and of the magnetization in an external field of 0.01 T. The dashed line is a Curie-Weiss fit to the high-temperature data indicating a paramagnetic Curie-Weiss temperature of 412(3) K.

$$\gamma(\mathbf{q},0) \approx \Omega_e \approx T_c(\mathbf{q}a)^{5/2}, \qquad (20)$$

where Ω_e is a characteristic critical-fluctuation energy. The form of the function $\gamma(\mathbf{q},\omega)$ for finite ω is unknown. We only assert that the real and imaginary parts of γ are of the same order of magnitude when $\omega \sim \Omega_e$. The calculation of the static spin-spin correlation function in Eq. (6) can be done using the fluctuation-dissipation theorem,

$$\langle S(\mathbf{q})S(-\mathbf{q})\rangle_0 = \int_0^{\omega_c}\frac{d\omega}{2\pi}\frac{2\hbar V\chi''_{zz}(\mathbf{q},\omega)}{g^2\mu_B^2(1-e^{-\beta\hbar\omega})}, \qquad (21)$$

where $\chi''_{zz}(\mathbf{q},\omega)$ is the imaginary part of the dynamical spin susceptibility of Eq. (13).

The presence of a magnetic field changes the situation significantly. Firstly, the value of $\langle \mathbf{S}\rangle$ in Eq. (6) becomes nonzero even above T_c. Therefore, one would expect a decrease of the magnetic resistivity as has been observed in the experiment. Physically this results from the fact that the applied magnetic field reduces the critical fluctuations around T_c. However, the spin-spin correlation function is quite different in the cases of weak ($g\mu H \ll \Omega_e$) and strong ($g\mu H \gg \Omega_e$) magnetic fields. In the latter case all fluctuations are strongly suppressed, resulting in the disappearance of the magnetic part of the electrical resistivity in accordance with formula (6). Moreover, the three-dimensional magnetic correlation length in the presence of a strong magnetic field changes its form[16] to $\xi(H) = a(k_B T_c/g\mu H)^{2/5}$. As one can see it diverges at $T=T_c$ only when $H=0$. Therefore, there will be no specific changes in the resistivity around T_c. This kind of behavior was indeed observed experimentally. In fact, for the applied magnetic field exceeding 5 T the resistance shows no anomaly around T_c and only a slight change with increasing field. This indicates the nonmagnetic origin of the remaining resistivity.

On the other hand, the effect induced by a weak magnetic field can be analyzed using an expansion of the static spin susceptibility χ, and damping function γ, in powers of $(g\mu H/\Omega_e)$ on the basis of the generalized dynamical scaling

hypothesis.[17] In the critical region, i.e., $\mathbf{q} \gg \xi^{-1}$ the expansion was first obtained by Lazuta and co-workers:[16]

$$\chi(\mathbf{q},H)=\chi(\mathbf{q})\left[1+\left\{\frac{1}{\xi\mathbf{q}}\right\}^{3/2}\left(\frac{g\mu H}{\Omega_e}\right)^2\right], \tag{22}$$

$$\gamma(\mathbf{q},\omega,H)=\gamma(\mathbf{q},\omega)\left[1+\frac{ig\mu H}{\Omega_e}\left(\frac{1}{\xi\mathbf{q}}\right)^{3/2}\right], \tag{23}$$

$$k_B T_c(H)=k_B T_c\left[1+\left\{\frac{1}{\xi\mathbf{q}}\right\}^{3/2}\left(\frac{g\mu H}{\Omega_e}\right)^2\right]. \tag{24}$$

This expansion provides a natural explanation for the suppression of the resistivity due to the external magnetic field as a result of an increase of the damping of the critical spin fluctuations. On the other hand, the magnetic field also slightly shifts T_c. This effect is quite clear if one remembers that even in MFA T_c will increase by applying a magnetic field. Therefore the peak in the resistivity will also shift towards higher temperatures.

In Fig. 4 we present the calculated normalized magnetic resistivity versus temperature together with experimental data obtained earlier[2] for various small magnetic fields up to $1T$. The results were obtained with the help of Eqs. (6) and (13). For $H=0$ the resisitivity has an anomalous peak centered around T_c due to the strong scattering of $5d$ conduction electrons on localized $4f^7$ spins. In particular, the three-dimensional magnetic correlation length of localized spins diverges at $T=T_c$ and therefore leads to an enhancement of the scattering of conduction electrons. With applied magnetic field the resistivity decreases and the anomalous peak is washed out due to nonzero $\langle S\rangle$ induced by magnetic field above T_c and quadratic correction to the damping, respectively. In the vicinity of T_c where the critical spin fluctuations play an important role and our analysis is applicable, the agreement between theory and normalized experimental data is quite remarkable.[18] Beyond this region the expansion (24) does not hold and the effect of critical fluctuations is overestimated. In the inset we also present the calculated curves in an extended temperature region. Note that the broad maximum of the resistivity curve around T_c observed in the experiment can be caused due to a presence of the short-range order above Curie temperature[19] and inhomogeneities of the sample.

In summary, we have analyzed the temperature dependence of the magnetic resistivity for GdI_2 and its dependence on an applied magnetic field. The anomalous peak of the resistivity near the Curie temperature is the result of a strong scattering of $5d_{z^2}$ conduction electrons by the localized $4f^7$ spin multiplets with $S=7/2$. It is due to a specific topology of the Fermi surface and the layered character of the system. We have investigated the influence of the external magnetic field in two limiting cases. A weak magnetic field causes a slight increase of the damping of the critical spin fluctuations and a nonzero magnetization even above T_c. That produces both, the broadening of the peak as well as its suppression. On the other hand, for a strong magnetic field, the spins align along the field direction and their fluctuations are reduced. This results in a suppression of the scattering channel for the conduction electrons by localized spins.

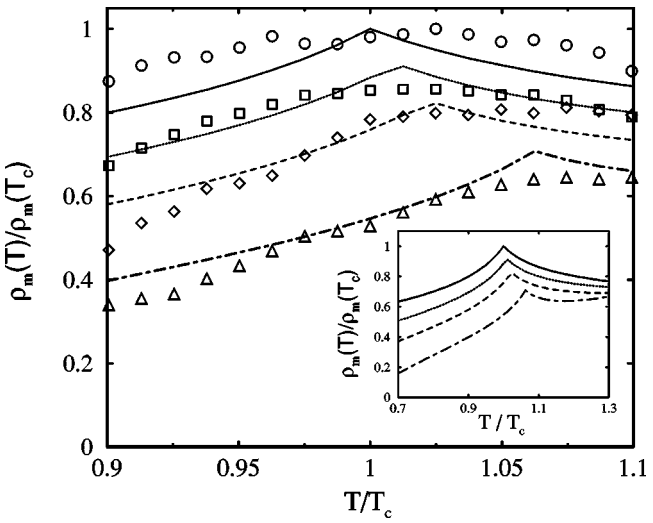

FIG. 4. Normalized magnetic part of the resisistivity with respect to its value at T_c and zero magnetic field versus temperature in the critical region for various magnetic fields. ($H=0.01$ T, solid curve; $H=0.03$ T, dotted curve; $H=0.06$ T, dashed curve; $H=1$ T, dashed-dotted curve). The corresponding experimental data are taken from Ref. 2 and shown as circles, squares, rhombs, and triangles, respectively. The inset shows the calculated curves for broader temperature region.

It is a pleasure to thank A. Zvyagin, V. Yushankhai, D. Manske, and K. H. Bennemann for stimulating discussions. One of us (I.E.) would like to thank the financial support of CRDF Grant No. REC. 007.

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