Negligible influence of domain walls on the magnetocaloric effect in $GdPt_2$

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Resistivity, magnetoresistance, magnetization, and in-field heat capacity measurements were performed of the $GdPt_2$ intermetallic compound. The magnetocaloric parameters ΔT_{ad} and $-\Delta S$ were derived from in-field heat capacity data. Comparison has been made between the magnetocaloric effect $-\Delta S$ and difference in resistivity $-\Delta\rho$ $[=\rho(H)-\rho(0)]$ as a function of temperature. There is distinct difference in the temperature dependence of $-\Delta S$ and $-\Delta\rho$ below the ferromagnetic transition temperature. However, after removing the domain wall contribution from $-\Delta\rho$, the natures of the $-\Delta S$ and $-\Delta\rho$ dependence as a function of temperature are similar. Our observation indicates that the domain wall contribution in the magnetocaloric effect is negligible in spite of the fact that it has a significant contribution in magnetotransport.

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I. INTRODUCTION

The magnetocaloric effect (MCE) is defined as the adiabatic temperature change (ΔT_{ad}) or isothermal entropy change ($-\Delta S$) of magnetic materials with the application of an external magnetic field. The MCE has immense technological importance for magnetic cooling. In recent years studies related to the MCE have gained momentum due to the observation of a giant MCE near room temperature.[1–3] The main focus in the study of the MCE is concentrated on finding new materials with a large MCE. Apart from its technological importance, the MCE can give us valuable information about magnetic materials like the nature of magnetic ordering, metamagnetic transitions etc.[4]

The building block of ferromagnetic materials below the ordering temperature are the magnetic domains which are separated by domain walls. The MCE is related to the thermomagnetic properties of magnetic materials. Therefore magnetic domains as well as domain walls are expected to have an effect on the MCE. However, the contribution of the domain wall on the MCE is not properly highlighted in the literature. The polycrystalline $GdPt_2$ compound crystallizes in a stable cubic $MgCu_2$ structure with ferromagnetic Curie temperature of 31 K.[5] In this present work, the main objective is to find out how strong is the contribution of the domain wall on the MCE in $GdPt_2$. Can it influence the temperature dependence of the MCE so much that it leaves some strong signature in the dependence?

Gadolinium, having $L=0$, has a negligible crystalline electric field in $GdPt_2$ and should reach its full moment value upon ordering and attain its full magnetic entropy value $R\ln(2J+1)$ or 17.3 J/(mol K). Due to the large moment of gadolinium, $GdPt_2$ is expected to show a reasonably large MCE. The magnetic and transport properties of the $GdPt_2$ compound have been studied by various authors.[6–8] It is believed that the magnetic interaction of well-localized $4f$ magnetic moments of Gd are mediated by conduction electrons via RKKY interactions. The critical behavior of the electrical resistivity was studied in the vicinity of the ordering temperature in the framework of molecular field theory.[7] To the best of our knowledge no report on a study of the thermodynamic properties of the $GdPt_2$ compound is available in the literature. We have studied the MCE as well as the magnetotransport properties of $GdPt_2$. Earlier reports[9,10] in the literature suggest that the dependences of the magnetocaloric effect and magnetoresistance can be similar. The comparison of the thermodynamic and magnetotransport data is a method of gaining deeper understanding about magnetic materials. Keeping this context in mind, we have measured and compared the temperature dependence of different quantities $-\Delta\rho$ and $-\Delta S$, one related to transport and the other related to thermodynamic properties.

II. EXPERIMENTAL DETAILS

The binary polycrystalline sample was prepared by arc melting of constituent elements of purity better than 99.9% in an argon atmosphere. The x-ray diffraction pattern confirms the single-phase nature of the compound which crystallizes in cubic $MgCu_2$ structure. Structural refinement was carried out using the Rietveld profile refinement method, and the lattice parameter is found to be 7.626 Å. Scanning electron microscope images have been taken at different positions of the sample and energy-dispersive x-ray (EDX) analysis has confirmed the compositional stoichiometry and homogeneity of the compound $GdPt_2$. Specific heat (C) measurements were performed using the semiadiabatic heat-pulse method in the temperature interval 4–60 K in the presence of 10- and 70-kOe magnetic fields. The temperature interval of the zero-field C measurement was 4–130 K. In the presence of a 5-kOe magnetic field a C measurement was performed in the temperature interval 4–40 K. The temperature dependence of the resistivity (ρ) in the absence of a field as well as in the presence of 5-, 10-, and 70-kOe magnetic fields was measured by the conventional four-probe method. The longitudinal magnetoresistance (MR) $[\Delta\rho/\rho=\{\rho(H)-\rho(0)\}/\rho(0)]$ measurements at 4, 10, 20, 40, and 80 K were carried out in magnetic fields up to 75 kOe. dc magnetization measurements were performed using a superconducting quantum interference device (SQUID) magnetometer.

III. RESULTS AND DISCUSSION

The temperature dependence of the dc susceptibility (χ) of the samples measured during heating (zero-field-cooled

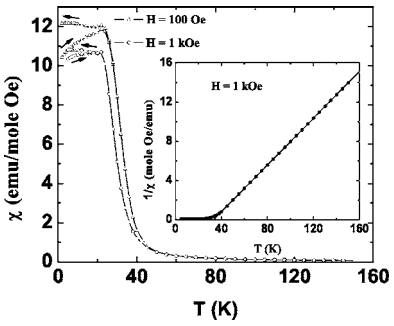

FIG. 1. Temperature dependence of zero-field-cooled (ZFC) and field-cooled (FC) susceptibility (χ) for 100-Oe and 1-kOe magnetic fields, respectively. Inset: inverse of susceptibility ($1/\chi$) as a function of temperature for a 1-kOe magnetic field. The solid line through the data points is the linear fit in the temperature interval 40–150 K.

susceptibility χ_{ZFC}) and cooling (field-cooled susceptibility χ_{FC}) in the presence of 100-Oe as well as 1-kOe magnetic fields is shown in Fig. 1. The temperature at which the change of χ as a function of temperature in the presence of a 1-kOe magnetic field is maximum was considered as the magnetic transition temperature which comes out to be ($T_C \sim$) 30 K. Below the transition temperature, there are clear bifurcations in the temperature dependence of χ_{ZFC} and χ_{FC}. The difference between the χ_{ZFC} and χ_{FC} curves as a function of temperature is large in a lower (100 Oe) magnetic field. The bifurcation in χ_{ZFC} and χ_{FC} below the magnetic ordering temperature of ferromagnetic materials can occur due to domain wall effects. In the case of χ_{ZFC}, the thermal fluctuation is reduced with decreasing T and thereby increases the zero-field-cooled magnetization (M_{ZFC}). At the same time, the movement of the domain walls also slows down, which results in decreases in M_{ZFC}. Due to these two competing effects, the χ_{ZFC} curve shows a maximum and then decreases with decreasing temperature. On the other hand, the χ_{FC} curve exhibits a tendency to saturate at low temperature rather than a maximum, indicating the absence of domain wall effects which implies that even a small magnetic field could cause domain wall movement at T_C.

The inverse susceptibility ($1/\chi$) as a function of temperature for $GdPt_2$ is plotted in the inset of Fig. 1 for an applied magnetic field of 1 kOe. The effective magnetic moment calculated from the slope of the $1/\chi$ curve in the paramagnetic region is found to be $8.17\mu_B$/Gd atom with positive paramagnetic Curie temperature 33 K for a 1-kOe magnetic field.

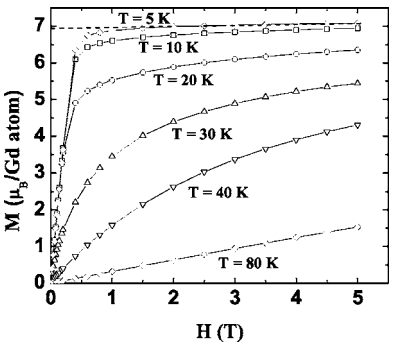

FIG. 2. Magnetization (M) as a function of magnetic field at different constant temperatures.

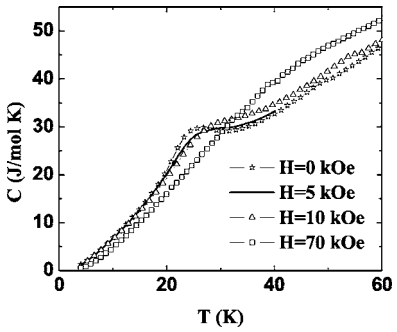

FIG. 3. Heat capacity (C) as a function of temperature for $GdPt_2$ at different constant magnetic fields.

The isothermal magnetization as a function of magnetic field at different temperatures is plotted in Fig. 2. The field dependence of the magnetization below the transition temperature clearly shows the ferromagnetic nature of the magnetic ordering in the $GdPt_2$ compound. Well above the ferromagnetic transition temperature—i.e., at 80 K—the nature of the magnetization versus temperature curve is linear. The saturation magnetization value is $6.94\mu_B$/Gd atom which was obtained by extrapolating high-field magnetization data to zero field at 5 K which is shown in Fig. 2 by a dashed line. At low temperature the magnetization increases rapidly up to 5 kOe; above that, it tends to saturate.

The specific heat of $GdPt_2$ as a function of temperature at various constant magnetic fields is plotted in Fig. 3. In the presence of a small external magnetic field (~5 kOe) the peak position of C shifts to higher temperature, indicating the ferromagnetic nature of magnetic ordering. At 70 kOe field, the specific heat peak disappears completely.

To find out the magnetic contribution of the specific heat we have fitted the zero-field C data using a Debye integral along with a linear contribution within the temperature interval 80–130 K and extrapolated the fitted data down to low temperature which is shown in Fig. 4. The total specific heat C can be expressed as

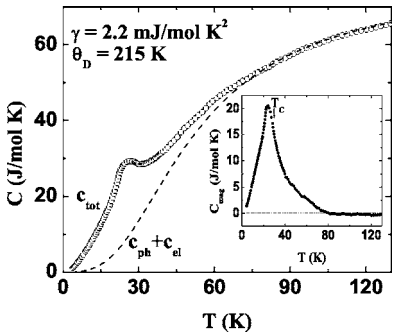

FIG. 4. Zero-field specific heat data as a function of temperature. The dashed line represents the lattice contribution of the specific heat. Inset: magnetic contribution of the specific heat as a function of temperature.

$$C = C_{el} + C_{ph} + C_{mag},$$

where C_{mag} is the magnetic contribution of the specific heat and C_{el} and C_{ph} are, respectively, the electronic and phonon contributions of the specific heat. The electronic part is of the form $C_{el}=\gamma T$, where γ is the electronic heat capacity coefficient. The phonon part, approximated as the Debye model, is of the form $C_{ph}=\mathcal{D}(\theta_D/T)$, where $\mathcal{D}(\theta_D/T)$ is the Debye function and θ_D is the Debye temperature. The C data were fitted using

$$C_{el} + C_{ph} = \gamma T + \mathcal{D}(\theta_D/T)$$

in the temperature interval 80–130 K under the approximation that well above the transition temperature the magnetic contribution is negligibly small. From the fitting, the value of γ and θ_D turns out to be 2.2 mJ/(mol K^2) and 215 K, respectively. The magnetic contribution of the specific heat was obtained by subtracting the regenerated nonmagnetic contribution in the temperature range 4–130 K using the above-mentioned γ and θ_D values. The temperature dependence of C_{mag} is shown in the inset of Fig. 4. From the inflection point of the C_{mag} data, we have obtained the ferromagnetic ordering temperature $T_C \sim 29$ K which is close to the T_C obtained from magnetization measurements. The maximum value of C_{mag} reaches 20.54 J/(mol K). The magnetic contribution to the specific heat for equal-moment (EM) magnetic structure in Gd intermetallic compounds is expressed as[11]

$$C_{EM} = \frac{5J(J+1)}{(2J^2+2J+1)}R.$$

Gadolinium, having $J=7/2$, yields $C_{EM}=20.15$ J/(mol K). The experimentally observed value of C_{mag} which is very close to the C_{EM} value indicates that the magnetic configuration in $GdPt_2$ is of equal-moment nature. Moreover, a noticeable magnetic contribution persists well above the transition temperature. The magnetic entropy of Gd intermetallic compounds attains its full value $R\ln(2J+1)$ or 17.3 J/(mol K) just above the ordering temperature.[12] The calculated magnetic entropy of our sample is 17.6 J/(mol K) at the ordering temperature which is in agreement with the above-mentioned value. This indicates that the Gd ions are ordered with full moments within $GdPt_2$.

The isothermal entropy change ($-\Delta S$) and adiabatic temperature change (ΔT_{ad}) were obtained from the total entropy, which was calculated from the experimental C data as a function of temperature at various constant magnetic fields. To calculate the entropy contribution for 0–4 K, linear variation of the C data was considered. The difference between the two entropy curves from zero field to in field for isothermal translation results in $-\Delta S$ and isentropic subtraction gives ΔT_{ad}. The temperature dependence of $-\Delta S$ and ΔT_{ad} for 5-, 10-, and 70-kOe magnetic fields is plotted in Figs. 5(a) and 5(b), respectively. The plot of ΔT_{ad} as a function of temperature shows a positive caretlike shape with maxima around the magnetic ordering temperature and ΔT_{ad} positive in the entire temperature range for all magnetic fields. The positive ΔT_{ad} even for 5-kOe magnetic fields is an indication of ferromagnetism in $GdPt_2$. The temperature dependences

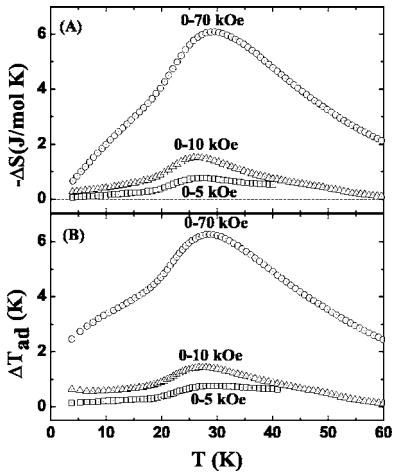

FIG. 5. (A) Isothermal entropy change $-\Delta S$ as a function of temperature calculated from the heat capacity data at constant magnetic fields. (B) Adiabatic temperature change ΔT_{ad} as a function of temperature calculated from the heat capacity data at constant magnetic fields.

of both ΔT_{ad} and $-\Delta S$ are almost similar to each other. The values of ΔT_{ad} around the magnetic ordering temperature for 5-, 10-, and 70-kOe magnetic fields are, respectively, 0.8, 1.4, and 6.3 K; i.e., the rate of change of ΔT_{ad} as a function of magnetic field decreases with increasing fields. This feature also indicates the ferromagnetic nature of $GdPt_2$ compounds.

The temperature dependence of resistivity (ρ) at various constant magnetic fields is shown in Fig. 6(a). The absolute values of the zero-field resistivity are, respectively, 57 $\mu\Omega$ cm at 4.2 K and 127 $\mu\Omega$ cm at 300 K. Our experimentally observed value of the ratio $\rho_{300}/\rho_{4.2}$ is approximately 2.2 which is in agreement with the reported value.[6] The temperature dependence of $-\Delta\rho$ is shown in Fig. 6(b) which was calculated from experimental resistivity data from 4 to 60 K at various constant magnetic fields. Below the

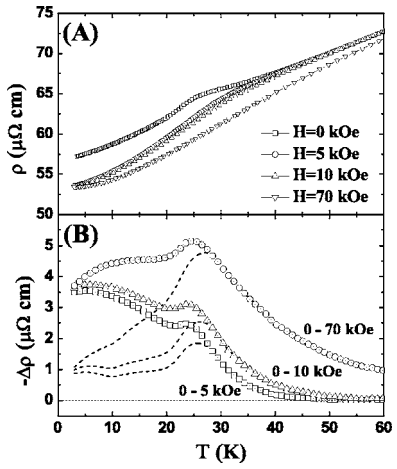

FIG. 6. (A) Temperature dependence of the resistivity at various constant magnetic fields. (B) Differences in resistivity $-\Delta\rho$ are plotted as a function of temperature with different symbols. The dashed line curves are the $-\Delta\rho$ vs temperature curve after subtracting the domain wall contribution in magnetoresistance.

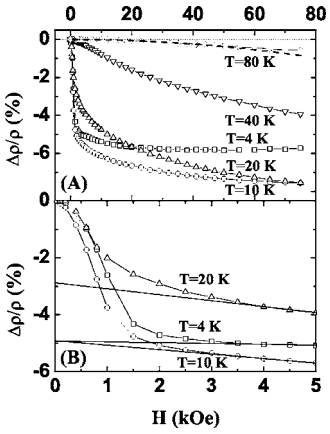

FIG. 7. (A) Magnetoresistance as a function field at various constant temperatures. (B) LFMR at different temperatures calculated by extrapolating 5-kOe data to zero field.

magnetic ordering temperature the variation of $-\Delta\rho$ and ΔT_{ad} or $-\Delta S$ with temperature is distinctly different for all three magnetic fields of 5, 10, and 70 kOe. It has been shown earlier that the temperature dependence of $-\Delta\rho$ and $-\Delta S$ can be similar.[9,10] It implies that for a ferromagnetic compound with increasing (decreasing) magnitude of $-\Delta S$ the magnitude of $-\Delta\rho$ is expected to increase (decrease) as a function of temperature. As a result one can expect that $-\Delta\rho$ decreases gradually as does $-\Delta S$ with decreasing temperature after showing a maximum around the ferromagnetic transition temperature of $GdPt_2$. In contrast to the expectation, $-\Delta\rho$ shows a broad hump at lower temperature. To find out the main cause behind the dissimilar behavior between $-\Delta\rho$ and the MCE we have performed MR measurements as a function of field at different constant temperatures, which is shown in Fig. 7(a). The MR curves at constant temperature clearly demonstrate the existence of significant low-field magnetoresistance (LFMR) originating from magnetic domain walls at low temperature. In the paramagnetic state the low-field MR vanishes and at higher temperature—i.e., at 80 K—MR follows a $-H^2$ magnetic field dependence as indicated by the dashed line in Fig. 7(a), which is an indication of enhanced spin fluctuation even at this high temperature. The values of LFMR at different constant temperatures (4, 10, and 20 K) were obtained by linear extrapolation of MR data around 5 kOe to zero field which is shown in Fig. 7(b). The LFMR below the ferromagnetic ordering temperature of polycrystalline compounds originates due to the suppression of domain wall scattering of conduction electrons with the application of magnetic fields and the value of LFMR was considered as a domain wall contribution in MR of $GdPt_2$. At the ferromagnetic transition temperature (~30 K) the domain wall contribution in MR was assumed to be zero. For the determination of the domain wall contribution in MR at the temperature intermediate to the above-mentioned temperatures (4, 10, 20, and 30 K), linear interpolation was performed. The domain wall contribution in $-\Delta\rho$ as a function of temperature is defined as $-\Delta\rho_d$ which has been calculated by multiplying the LFMR value with zero-field resistivity at the corresponding temperatures. The contributions of $-\Delta\rho_d$ at different temperatures for 5-, 10-, and 70-kOe magnetic fields were considered to be same. The evaluated domain contributions $-\Delta\rho_d$ at different temperatures have been subtracted from the temperature dependence of $-\Delta\rho$ for magnetic field values of 5, 10, and 70 kOe. After removing the domain wall contribution, the broad hump in $-\Delta\rho$ vanishes which is shown in Fig. 6(b) by the dashed line and the natures of the $-\Delta\rho$ and $-\Delta S$ curves as a function of temperature turn out to be similar. These observations indicate that the dissimilar temperature dependence of $-\Delta\rho$ and $-\Delta S$ in $GdPt_2$ is originating from the fact that the magnetic domain wall has a significant contribution in $\Delta\rho$ but negligible influence on the MCE.

IV. SUMMARY

The MCE along with the transport properties has been studied in the $GdPt_2$ compound. We have observed distinct differences in the temperature dependence of $-\Delta\rho$ and $-\Delta S$ below the ferromagnetic ordering temperature. However, if we remove the domain wall contribution from $-\Delta\rho$, then the natures of the $-\Delta\rho$ and $-\Delta S$ curves as a function of temperature are similar. It highlights the fact that the domain wall contribution in the magnetocaloric effect is negligible in spite of the fact that it has a significant contribution in transport in $GdPt_2$.

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