Giant magnetoresistance near the magnetostructural transition in $Gd_5(Si_{1.8}Ge_{2.2})$

Cite as: Appl. Phys. Lett. **73**, 3462 (1998); https://doi.org/10.1063/1.122797
Submitted: 17 August 1998 • Accepted: 02 October 1998 • Published Online: 02 December 1998

L. Morellon, J. Stankiewicz, B. García-Landa, et al.

Giant magnetoresistance near the magnetostructural transition in $Gd_5(Si_{1.8}Ge_{2.2})$

L. Morellon,[a] J. Stankiewicz, B. García-Landa, P. A. Algarabel, and M. R. Ibarra
Departamento de Física de la Materia Condensada and Instituto de Ciencia de Materiales de Aragón, Universidad de Zaragoza and Consejo Superior de Investigaciones Científicas, 50009 Zaragoza, Spain

(Received 17 August 1998; accepted for publication 2 October 1998)

Zero-field electrical resistivity over the temperature range of 4–300 K and magnetoresistance in magnetic fields of up to 12 T have been measured in $Gd_5(Si_{1.8}Ge_{2.2})$. This system undergoes a first-order magnetostructural transition at $T_C \cong 240$ K, from a high-temperature paramagnetic to a low-temperature ferromagnetic phase, accompanied by a large drop in the resistivity. The application of an external magnetic field above T_C can induce this transition, and a giant negative magnetoresistance effect ($\Delta\rho/\rho \cong -20\%$) is observed associated with this first-order field-induced transition. © *1998 American Institute of Physics.* [S0003-6951(98)00449-5]

Since the discovery of the giant magnetoresistance effect (GMR) in Fe/Cr multilayers,[1] an enormous experimental and theoretical effort has been focused on these artificially fabricated structures.[2] This active research has been motivated by their prospective use in magnetoresistive read-head technology. The GMR effect is usually attributed to spin-dependent scattering[3] inside the bulk or at the interfaces, the overall resistivity being lower for a parallel than for an antiparallel arrangement of the magnetic moments. In addition to the magnetic multilayers, other systems also show large magnetoresistive effects, namely granular alloys,[4] La-based manganese perovskites,[5] and bulk intermetallic materials.[6] Typical examples of the latter are, for instance, the natural layered $SmMn_2Ge_2$[7] or the FeRh alloy[8] where the GMR effect occurs at a first-order field-induced transition from the antiferromagnetic to the ferromagnetic state. The aim of this letter is to extend the study of the GMR effects in bulk intermetallic systems to the $Gd_5(Si_{1-x}Ge_x)_4$ alloys.

A giant magnetocaloric effect (the highest reported to date) has been recently discovered in the $Gd_5(Si_{1-x}Ge_x)_4$ alloys,[9] making this system a potential candidate for magnetic refrigeration. The composition range $0.24 \leqslant x \leqslant 0.5$ is of special interest. In this region, the giant magnetocaloric effect is related to a first-order magnetic transition from a paramagnetic to a low-temperature ferromagnetic state, at temperatures ranging from 130 ($x=0.24$) to 276 K ($x=0.5$).[10] In a recent study of the $Gd_5(Si_{1.8}Ge_{2.2})$ alloy, we have demonstrated that the transition in question ($T_C \cong 240$ K) is in fact a first-order structural transition from a $P112_1/a$ monoclinic (paramagnetic) to a Pnma orthorhombic (ferromagnetic) structure.[11] This magnetostructural transition can be induced reversibly by applying an external magnetic field, giving rise to strong magnetoelastic effects ($\Delta V/V \cong 0.4\%$). Therefore, these alloys are also attractive in view of their potential technological applications for magnetostrictive transducers. In the present letter we report on the existence of GMR effects in $Gd_5(Si_{1.8}Ge_{2.2})$ at the magnetostructural transition.

The sample used in the present study was taken from a section of the specimen used in our previous magnetostructural investigation. Details on the materials used, preparation techniques, and subsequent characterization can be found elsewhere.[11] The alternating current (ac) initial magnetic susceptibility was measured in the temperature range of 200–300 K using a modified mutual inductance Hartshorn bridge with an excitation field of about 30 mOe of peak value at a frequency of 15 Hz. The zero-field electrical resistivity over 4–300 K and magnetoresistance isotherms up to 12 T between 200 and 300 K were measured by means of a six-probe method on a bar-shaped sample. Before measurements, the sample, quite brittle, was polished and checked for possible cracks. Contact leads were ultrasonically soldered to the sample. The relative error in the resistivity measurements is about 0.05%; absolute values were determined within 5%.

The temperature dependence of the zero-field electrical resistivity, ρ, in the range of 200–300 K is shown in Fig. 1. A drastic decrease in the resistivity, $\Delta\rho/\rho \approx 20\%$, is clearly observed at 237 K for decreasing temperatures with a thermal hysteresis of about 5 K. This abrupt drop coincides with the onset of the expected first-order magnetostructural phase transition at this temperature ($T_C \cong 240$ K), from a high-temperature paramagnetic to a low-temperature ferromagnetic state.[9–11] To illustrate this point, the temperature dependence of the ac magnetic susceptibility has been included in Fig. 1. As can be also seen, the values of the resistivity in the paramagnetic state depend on the thermal history of the sample. This irreversibility is likely due to the formation of microcracks when cycling the sample through the transition. As reported in a previous work, a large volume contraction, $\Delta V/V \cong 0.4\%$ takes place at the transition,[11] which may introduce stress at the grain boundaries. In the inset of Fig. 1, the resistivity in the whole temperature range of 4–300 K is shown. ρ follows a typical metallic behavior[12] and no other anomalies are observed. The value of the residual resistivity, $\rho_0 = 73\ \mu\Omega$ cm, is relatively high, probably due to a large impurity concentration and static lattice imperfections.

The magnetostructural transition in $Gd_5(Si_{1.8}Ge_{2.2})$ can be induced reversibly by application of an external magnetic field in the paramagnetic state.[9–11] Therefore, according to

[a]Electronic mail: morellon@posta.unizar.es

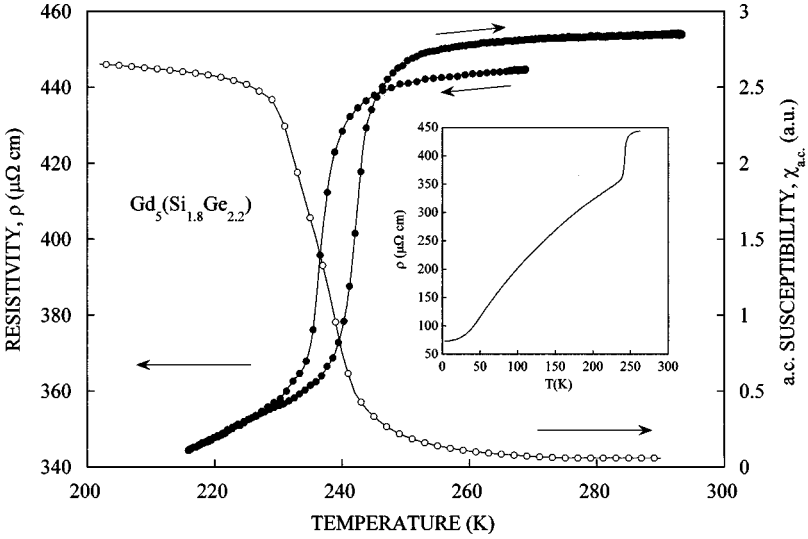

FIG. 1. Zero-field electrical resistivity (ρ) and ac initial magnetic susceptibility (χ_{ac}) of $Gd_5(Si_{1.8}Ge_{2.2})$ as a function of temperature. The inset shows the zero-field ρ over the whole temperature interval of 4–300 K.

the resistivity results presented so far, a large magnetoresistance effect should be expected at this first-order field-induced phase transition. The experimental results for the relative magnetoresistance, defined as $\Delta\rho/\rho = [\rho(H,T)-\rho(0,T)]/\rho(0,T)$, are plotted in Fig. 2 at some selected temperatures. In the ferromagnetic phase, at temperatures below the transition, [see for example Fig. 2(f) at 230 K], the magnetoresistance is small and negative (4% at 12 T) as expected for a ferromagnetic system with localized moments.[12] The behavior of ρ is completely different above T_C [see Figs. 2(a)–2(e)]. Above some critical field H_{CR}, the system undergoes a first-order field-induced phase transition to the ferromagnetic state, and a negative GMR effect is observed, $\Delta\rho/\rho \cong -20\%$. The magnitude of this effect is comparable with that of other intermetallic compounds presenting first-order metamagnetic transitions such as FeRh (50%),[8] $Hf_{1-x}Ta_xFe_2$ (6%),[13] $Ce(FeRu)_2$ (20%),[14] $SmMn_2Ge_2$ (5%),[7] and Gd_2In (10%).[15] Nevertheless, the origin of the transition in $Gd_5(Si_{1.8}Ge_{2.2})$ is of a structural nature[11] although it exhibits similar transport properties to the systems mentioned in which the corresponding transition is of purely magnetic origin. The temperature dependence of the values of H_{CR}, calculated at the maximum slope of the magnetoresistance isotherms, is shown in Fig. 3. H_{CR} varies linearly with temperature with a slope of 4.5 ± 0.1 T/K and extrapolates to zero at the transition temperature. This value compares very well with that previously obtained from magnetostriction measurements (4.8 K/T).[11] A hysteresis of about 1 T has also been observed in the $\rho(H)$ dependence. All of the above results support the existence of a first-order field-induced phase transition from the paramagnetic to the ferromagnetic state. Consequently, a negative GMR effect of similar magnitude as the resistivity change at the transition in zero field (Fig. 1) is observed. It is noteworthy to point out that $\Delta\rho/\rho$ is almost constant for $H > H_{CR}$ in the temperature range studied.

In order to correlate the behavior of the magnetoresistance with the magnetic properties, we tried to use the so-called ''s-f model.'' [16] Although this model accounts for the GMR in granular systems[17] and dilute magnetic alloys,[18] it does not explain the GMR effect observed in the $Gd_5(Si_{1.8}Ge_{2.2})$ alloy. The spin-dependent scattering at a paramagnetic–ferromagnetic transition in a system with localized moments system leads to a quite small variation in resistivity. The application of a magnetic field in this case would give a magnetoresistance value typically one order of magnitude smaller than the one observed.[12] Therefore, other

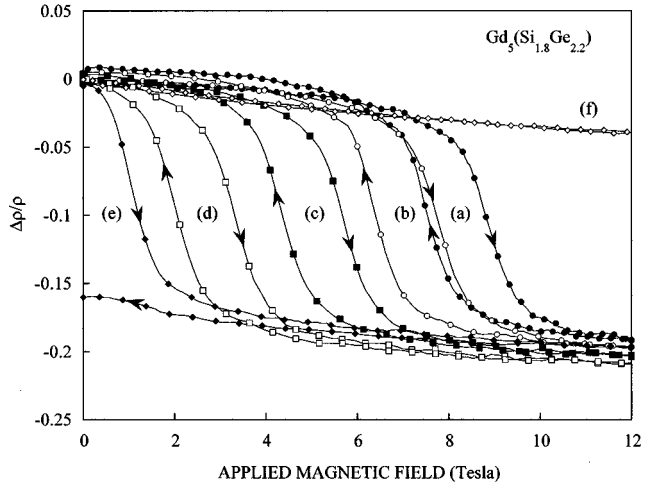

FIG. 2. Magnetoresistance ratio $\Delta\rho/\rho = [\rho(H,T)-\rho(0,T)]/\rho(0,T)$ as a function of the applied magnetic field at some selected temperatures of (a) 275, (b) 270, (c) 260, (d) 250, (e) 240, and (f) 230 K.

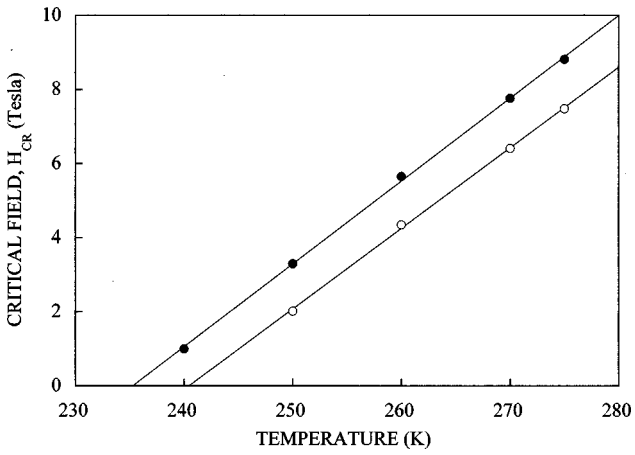

FIG. 3. Temperature dependence of the critical field, H_{CR}, obtained from the magnetoresistance isotherms for the $Gd_5(Si_{1.8}Ge_{2.2})$ compound. The symbols indicate increasing (●) and decreasing (○) magnetic field.

mechanisms, apart from spin-dependent scattering, such as formation of superzone boundary gaps at the transition temperature may be important for the GMR of the alloys studied, as in the case of UNiGa.[19] However, an increase in resistivity should be expected, which, obviously, is not our case. One may also consider variations in the band structure of $Gd_5(Si_{1.8}Ge_{2.2})$, since the crystal symmetry changes through the transition.[11] This would lead to different density of states at the Fermi level, which may explain a decrease in resistivity. Detailed band structure calculations should be carried out to confirm this point.

In conclusion, a negative GMR effect ($\Delta\rho/\rho \cong -20\%$) has been found in the $Gd_5(Si_{1.8}Ge_{2.2})$ alloy at temperatures above the magnetostructural transition ($T_C \cong 240$ K). The results presented are consistent with a first-order field-induced phase transition from the paramagnetic to the ferromagnetic state. Nevertheless, the potential technological applications of this material are still limited by the relatively large critical fields at room temperature. Work is in progress in order to reduce the required applied magnetic field.

The authors acknowledge the financial support of the Spanish CICYT under Grant No. MAT96-0826.

[1] M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friederich, and J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988).
[2] S. S. P. Parkin, in *Ultrathin Magnetic Structures II*, edited by B. Heinrich and J. A. C. Bland (Springer, Berlin, 1994), p. 148.
[3] P. M. Levy, S. Zhang, and A. Fert, Phys. Rev. Lett. **65**, 1643 (1990).
[4] A. E. Berkowitz, J. R. Mitchell, M. J. Carey, A. P. Young, S. Zhang, F. E. Spada, F. T. Parker, A. Hutten, and G. Thomas, Phys. Rev. Lett. **68**, 3745 (1992); J. Q. Xiao, J. S. Jiang, and C. L. Chien, Phys. Rev. Lett. **68**, 3749 (1992).
[5] R. von Helmolt, J. Wecker, B. Holzapfel, L. Shultz, and K. Samwer, Phys. Rev. Lett. **71**, 2331 (1993); K. Chahara, T. Ohno, M. Kasai, and Y. Kozono, Appl. Phys. Lett. **63**, 1990 (1993); S. Jin, T. H. Tiefel, M. McCormack, R. A. Fastnacht, R. Ramesh, and J. H. Chen, Science **264**, 413 (1994).
[6] V. Sechovský, L. Havela, K. Prokeš, H. Nakotte, F. R. de Boer, and E. Brück, J. Appl. Phys. **76**, 6913 (1994); V. Sechovský, L. Havela, H. Nakotte, and E. Brück, IEEE Trans. Magn. **32**, 4687 (1996).
[7] R. B. van Dover, E. M. Gyorgi, R. J. Cava, J. J. Krajewski, R. J. Felder, and W. F. Peck, Phys. Rev. B **47**, 6134 (1993); E. V. Sampathkumaran, P. L. Paulose, and R. Mallik, Phys. Rev. B **54**, R3710 (1996).
[8] P. A. Algarabel, M. R. Ibarra, C. Marquina, A. del Moral, J. Galibert, M. Iqbal, and S. Askenazy, Appl. Phys. Lett. **66**, 3062 (1995).
[9] V. K. Pecharsky and K. A. Gschneidner, Jr., Phys. Rev. Lett. **78**, 4494 (1997).
[10] V. K. Pecharsky and K. A. Gschneidner, Jr., Appl. Phys. Lett. **70**, 3299 (1997).
[11] L. Morellon, P. A. Algarabel, M. R. Ibarra, J. Blasco, B. Garcia-Landa, Z. Arnold, and F. Albertini, Phys. Rev. B (to be published).
[12] J. M. Fournier and E. Gratz, in *Handbook of the Physics and Chemistry of Rare Earths*, edited by K. A. Gschneidner, Jr., L. Eyring, G. H. Lander, and G. R. Choppin (North–Holland, Amsterdam, 1993), Vol. 17, pp. 409–537.
[13] H. G. M. Duijn, E. Brück, A. A. Menovsky, K. H. J. Buschow, F. R. de Boer, R. Coehoorn, M. Winkelmann, and K. Siemensmeyer, J. Appl. Phys. **81**, 4218 (1997).
[14] H. P. Kunkel, X. Z. Zhou, P. A. Stampe, J. A. Cowen, and Gwyn Williams, Phys. Rev. B **53**, 15099 (1996).
[15] P. A. Stampe, X. Z. Zhou, H. P. Kunkel, J. A. Cowen, and Gwyn Williams, J. Phys.: Condens. Matter **9**, 3763 (1997).
[16] K. Yosida, Phys. Rev. **107**, 396 (1957); T. Van Peski-Tinbergen and A. J. Dekker, Physica **29**, 917 (1963).
[17] J. Q. Wang, P. Xiong, and G. Xiao, Phys. Rev. B **47**, 8341 (1993).
[18] J. Bews, A. W. Sheikh, and G. Williams, J. Phys. F **16**, 1537 (1986).
[19] V. N. Antonov, A. Ya. Perlov, P. M. Oppeneer, A. N. Yaresko, and S. V. Halilov, Phys. Rev. Lett. **77**, 5253 (1996).