MAGNETIC AND TRANSPORT PROPERTIES OF AMORPHOUS FERRO MAGNETIC Gd-Au, Gd-Ni AND Gd-Co ALLOYS OBTAINED BY SPLAT-COOLING*

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ABSTRACT

We present the results of magnetization and transport measurements on amorphous Gd80Au20, $Gd_{68}Ni_{32}$ and $Gd_{67}Co_{33}$ alloys over a temperature range of 1.8-300°K in fields up to 75 kOe. These ferromagnetic alloys obtained by splat-cooling have Curie temperatures T of 150, 125 and 175°K, respectively. The saturation moment per Gd atom extrapolated to 0 0 K is estimated to be 7 ± 0.1 μ_{B} . The exchange integrals for Gd-Au and Gd-Ni are determined from the value of T and from the temperature dependence of the saturation magnetization. The zero-field resistivity for Gd-Ni and Gd-Co exhibits maxima around ${\bf T}_{\bf C}$. We present some preliminary results of magnetoresistivity measurements with applied field parallel and perpendicular to the foil plane. The anisotropy is in-plane for Gd-Co. For the Gd-Au and Gd-Ni alloys, there is no well-defined easy axis.

INTRODUCTION

Amorphous alloys of gadolinium with noble 1-4 or transition metals, 6 have been the subject of considerable interest in recent years. In particular, magnetic and galvanometric properties of sputtered or coevaporated Gd-Co and Gd-Fe alloys with high transition-metal content have been studied rather extensively. Since it has been shown that the techniques used for sample preparation have some influence on magnetic properties (e.g. anisotropy), 8, a comparison with splat-cooled amorphous alloys seems to be highly desirable. Very few results are available so far on rare-earth noble-(transition) metal amorphous alloys obtained by splat-cooling. These rareearth rich alloys can be quenched into the amorphous state over a rather narrow concentration range, typically over a few atomic per cent around the eutectic composition. We report on magnetic and transport properties measurements performed on amorphous alloys of nominal composition Gd80Au20, Gd68Ni32 and Gd₆₇Co₃₃. (From now on, they will be referred to as Gd₄Au, Gd₂Ni and Gd₂Co). The interest of these alloys is twofold. First, they provide some information about amorphous alloys on the high Gd side, which has received little attention so far. Second, they allow a comparison with results on amorphous Gd alloys obtained by other preparation techniques. A preliminary report on Gd₄Au alloys was presented elsewhere.10

EXPERIMENTAL PROCEDURES

The samples were quenched from the melt by the piston and anvil technique. 11 Samples were in the form of foils with thickness of about $40\,\mu m$. The structure of each foil was checked by X-ray scanning with a Norelco diffractometer. Spontaneous crystallization is observed at temperatures of about 200 to 250°C.

Magnetization measurements were carried out by the Faraday method using a superconducting coil (maximum field 75 kOe) over temperatures ranging from 1.8 to 300 $^{\rm O}$ K. The Curie temperatures T $_{\rm C}$ were observed using an ac inductance bridge technique. T was defined as the inflexion point of the Curie transition (transition width of about 5°K). For the

Gd₄Au sample, this determination was found to be in good agreement with that obtained from detail magnetization measurements over the critical region using the thermodynamical parameters method. Resistivity at zero field and magnetoresistivity (in fields up to 75 kOe) measurements were performed by a standard four-probe technique between 1.8 and 300°K. We measured the transverse and longitudinal magnetoresistivities with applied field parallel to the foil plane, and the transverse magnetoresistivity with applied field perpendicular to the plane.

RESULTS AND DISCUSSION

1. Magnetic properties.
Curie temperatures. The three amorphous alloys we studied were found to be good ferromagnets. The effect of amorphousness cannot be evaluated in a direct way, for the crystalline counterparts do not exist. In the Gd-Au system, $(T_c = 149.50^{\circ} \text{K})$ the closest compound is GdAu (antiferromagnetic, $T_N = 37^{\circ} K$). 13 In the concentration range we are interested in, amorphous $Gd_2Ni(T_c = 125^{\circ}K)$ can be compared with crystalline Gd₃Ni (antiferromagnetic, $T_N = 100^{\circ} \text{K}$)¹⁴ and GdNi (ferromagnetic; $T_C = 71^{\circ} \text{K}$)¹⁵, and amorphous Gd₂Co($T_C = 175^{\circ} \text{K}$) lies between the compounds Gd₃Co (antiferromagnetic; $T_N = 127^{\circ} \text{K}$)¹⁶ and Gd₄Co₃ (ferromagnetic; $T_C = 230^{\circ} \text{K}$).¹⁷ This rough comparison suggests that the amorphous structure favors in some way the ferromagnetic order as noticed before for amorphous $\operatorname{Gd}_{56}^{\operatorname{Ag}}_{44}$. The relatively high value of T for amorphous Gd2Co as compared with that of amorphous Gd2Ni may be explained by the strong Co-Co interactions. A molecular field analysis 2,18 of Co rich amorphous R.E.-Co alloys provided the following order of magnitude for the exchange constants: $J_{CoCo} \approx 200$; $J_{CoGd} = -20$; $J_{GdGd} = 2$ (in 10^{-6} ergs). Thus, the influence of the strong Co-Co interactions seems to persist in the Gd dominated amorphous Gd2Co. The values of T_c for amorphous Gd₄Au and Gd₂Ni may be correlated to the exchange interaction (assuming that Au and Ni play only as diluents). First, a value of the mean exchange constant may be determined from the Rushbrooke-Wood formula 19:

 $k_B^T_c/J = (5/96) (z - 1) [11S(S + 1) - 1]$ where z is the number of nearest neighbors. For Gd_AAu , we determined $J = 2.28^{\circ}K$ taking $z = 8.20^{\circ}K$ Similar value is obtained for Gd_2Ni taking $z \simeq 7$ and T_c = 125°K. The value of S is taken to be 7/2 in both cases. Using de Gennes²¹ approach, the exchange constant J_{sf} of the indirect RKKY interaction in rareearths can be evaluated:

$$k_{\rm B}T_{\rm c} = (3\pi n^2/4E_{\rm F})_{\rm sf}^2(g-1)^2S(S+1)\sum_{\rm j\neq i} F(2k_{\rm F}R_{\rm ij})$$
 (2)

where the notation follows that in reference 21. In $\mathrm{Gd}_4\mathrm{Au}$, J_{sf} was determined to be 0.19 eV. Assuming similar values for the electronic parameters n, EF and S for Gd_2Ni , we also obtain $J_{sf} \simeq 0.19$ eV by including a reduction factor of 0.8 in the summation ΣF. The latter comes from a reduction in the Gd-Gd

coordination number.

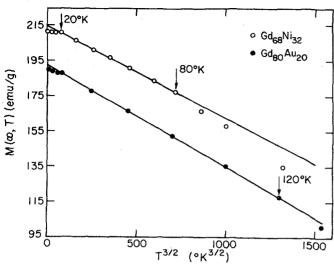


Fig. 1. $M(\infty, T)$ versus $T^{3/2}$ for amorphous Gd_2Ni and Gd_4Au alloys. Arrows indicate departures from the $T^{3/2}$ Bloch law.

Saturation magnetization. From the saturation magnetization extrapolated at $0\,^{\rm O}K$, it is not possible to determine a reliable value for the small moment carried by the Ni or Co atoms. The moment per Gd atom can be estimated to be $7\pm0.1\mu_{\rm B}$. The temperature dependence of the saturation magnetization for Gd_Au and Gd_Ni is shown on figure 1. The exchange constant $\rm J_{\rm Gd-Gd}$ can then be evaluated from the spin-wave theory $\rm ^{\rm 22}$ using the relation

$$M(\infty, T) = M(\infty, 0) (1 - bT^{3/2})$$

with

$$b = \frac{0.0587}{QS} \left\{ \frac{k_B}{2JS} \right\}^{3/2}$$
 (3)

where Q depends on the Gd-Gd coordination number. We determined $J=1.8^{\rm O}{\rm K}$ for Gd₄Au by taking Q=2. In Gd₂Ni, Q is reduced to about 1.84 by considering the coordination number mentioned beforehand, and J is estimated to be $\sim 2.18^{\rm O}{\rm K}$. By comparing the J_{Gd-Gd} values determined by the two methods above, one sees an appreciable discrepancy between the two J values in Gd₄Au. In crystalline Gd, this was accounted for by Goodings²³ in terms of RKKY coupling beyond the Gd nearest neighbors. Apparently, this type of coupling is less significant in amorphous Gd₂Ni as compared with amorphous Gd₄Au.

In conclusion, the long-range structural disorder results in a lowering of the magnetic moment per Gd atom. However, by considering the value of $J_{\rm sf}$ obtained in dilute La-Gd-Au alloys²¹, it is seen that the reduction in the RKKY interaction is surprisingly small as compared with what one could expect from the very short electronic mean free path in these amorphous alloys.

2. Transport properties.

Zero-field resistivity. The temperature dependence of the zero-field resistivity for the three samples is shown in figure 2. Resistivity minima occur at 15, 2 and 8°K, for amorphous Gd Au, Gd Ni and Gd Co, respectively. These minima are presumably magnetic in origin. Detail magnetoresistivity studies at temperatures below the minima are in progress. Above the minimum, the resistivity for amorphous Gd4Au increases monotonically with

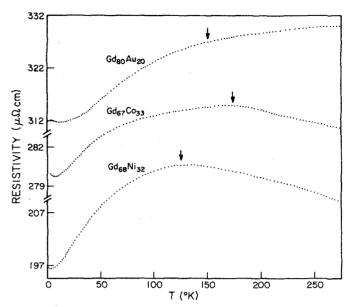


Fig. 2. Zero field resistivity versus temperature for amorphous $\mathrm{Gd}_4\mathrm{Au}$, $\mathrm{Gd}_2\mathrm{Co}$ and $\mathrm{Gd}_2\mathrm{Ni}$ alloys. Arrows indicate the Curie temperatures.

temperature without exhibiting any noticeable singularity at T_c (except possibly a gradual change in dp/dT around T_c). In contrast, the $\operatorname{Gd}_2\operatorname{Ni}$ and $\operatorname{Gd}_2\operatorname{Co}$ samples exhibit a broad maximum at temperatures close to their respective Curie temperature. This behavior is not clearly understood yet.

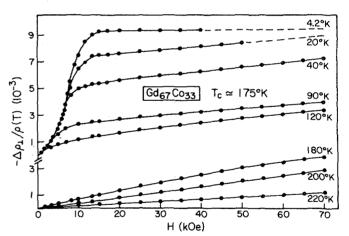


Fig. 3. Differential transverse magnetoresistivity (H// plane) versus applied field for amorphous Gd₂Co alloy at temperatures below and above the Curie temperature (175^OK).

Magnetoresistivity. Field parallel to the sample plane. Figure 3 shows the transverse magneto-

resistivity for an amorphous Gd_2Co sample. The magnetoresistivity isotherms strongly resemble the magnetization isotherms at corresponding temperatures below and above T_c . In the ferromagnetic region, the spontaneous magnetoresistivity (defined by extrapolating $\Delta\rho$ at $B=H+4\pi M_s+H_D=0$) decreases as a function of temperature much faster than the spontaneous magnetization for $4 \le T \le 100^{\circ} K$. Over the temperature range $100\text{-}170^{\circ} K$, the magnetoresistivity at high field is weakly temperature dependent. Above T_c , the magnetoresistivity gradient obeys roughly a Curie-Weiss law. The magnetoresistivity isotherms

for amorphous Gd₄Au are very similar to those obtained for Gd, Co, except the fact that the saturation at low temperature is harder than for the Gd2Co sample (figure 4). In both cases, the negative saturated magnetoresistivity 25 has the same order of magnitude at 4.2°K. On the other hand, comparing the longitudinal and transverse magnetoresistivity as measured with applied field parallel to sample plane, the anisotropy at 4.2°K, if any, is extremely weak.26 This temperature dependence of the gradient dΔρ/dH in the saturated field region resembles that observed for amorphous ferromagnetic Ni-Au. 27 Preliminary measurements on the Gd2Ni sample did not exhibit any trend of saturation at 1.8°K and 75 kOe, as shown in figure 4.

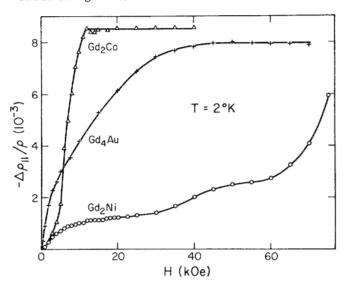


Fig. 4. Differential longitudinal magnetoresistivity (H// plane) at 20K for amorphous Gd4Au, Gd2Co and Gd2Ni alloys.

Field perpendicular to the sample plane. We have measured the magnetoresistivity with applied field perpendicular to the foil plane. The splat-cooled amorphous Gd2Co was found to have a magnetically hard axis perpendicular to the sample plane, as reported⁸ for thermally evaporated Gd-Co films when Co concentration is less than 91 at. %. The anisotropy is believed to be in-plane also for Gd₂Ni and Gd₄Au, although the easy axis is poorly defined in these latter alloys. The magnetoresistivity for Gd2Ni exhibits some particular features. The magnetoresistivity reaches a plateau (15 \leq H \leq 25 kOe and 35 \leq H \leq 45 kOe for the in-plane and off-plane magnetoresistivity, respectively). Then, for higher fields, the magnetoresistivity does not saturate. In addition, irrespectively of the field direction, the magnetoresistivity for Gd2Ni seems to exhibit an oscillatory behavior.

A more detailed study of the galvanometric properties of Gd Ni is being planned. In conclusion, the effects due to the Au, Co, Ni

contents, respectively, seem to be very important for the temperature and field dependence of the galvanometric properties of amorphous Gd-based alloys.

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REFERENCES

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J. J. Hauser, Phys. Rev. B 12, 5160 (1975).

- N. Heiman and K. Lee, AIP Conf. Proc. 34, 319 (1976).
- B. Boucher, J. Phys, (Paris) Lett. 37, L345 (1976).
- T. Mizoguchi, T. R. McGuire, R. J. Gambino, and S. Kirkpatrick, Physica 86-88B, 783 (1977). R. J. Gambino, P. Chaudhari, and J. J. Cuomo,
- AIP Conf. Proc. 18, 578 (1973).
- K. Lee and N. Heiman, AIP Conf. Proc. 24, 108 (1974).
- P. Chaudhari, J. J. Cuomo, and R. J. Gambino, IBM J. Res. Develop. 17, 66 (1973).
- R. C. Taylor and A. Gangulee, J. Appl. Phys.
- 47, 4666 (1976).

 W. L. Johnson, S. J. Poon, and P. Duwez, Phys. Rev. B<u>11</u>, 150 (1975).
- 10. S. J. Poon and J. Durand, in Amorphous Magnetism II, Troy, New York, 1976, edit. R. Hasegawa and R. A. Levy (Plenum Press, New York, 1977, to be published).
- P. Duwez, in Progress in Solid State Chemistry
- (Pergamon Press, Oxford, 1966), vol. 3, p. 377. S. J. Poon and J. Durand, Phys. Rev. B16, (1977, to be published).
- 13. F. Kissel and W. E. Wallace, J. Less Common Met. 11, 417 (1966).
 14. J. L. Feron, R. Lemaire, D. Paccard, and R.
- Pauthenet, C. R. Acad. Sci. (Paris) 267B, 371, (1968).
- 15. R. E. Walline and W. E. Wallace, J. Chem. Phys. 44, 4651 (1966).
- J. L. Feron, D. Gignoux, R. Lemaire, and D.
- Paccard, Colloq. CNRS 180, vol. 2. p. 75 (1970).
 17. C. Berthet-Colomiras, J. Laforest, R. Lemaire, R. Pauthenet, and J. Schweitzer, Cobalt 39, 97 (1968).
- 18. R. Hasegawa, B. E. Argyle, and L. J. Tao. AIP Conf. Proc. 24, 110 (1974).
- G. S. Rushbrooke and P. J. Wood, Mol. Phys. 1, 258 (1958).
- 20. J. Logan, Scripta Met. 9, 379 (1970).
- 21. P. G. de Gennes, J. Phys. Radium 23, 510 (1962); S. J. Poon and J. Durand, Solid State Commun.,
- 21, 793 (1977); 21, 999 (1977). 22. J. A. Hoffmann et al., J. Phys. Chem. Sol. 1, 45 (1956).
- D. A. Goodings, Phys. Rev. <u>127</u>, 1532 (1962).
 S. J. Poon, J. Durand and M. Yung, Sol. State Commun. 22, 475 (1977).
- J. Smit, Physica 26, 612 (1951); J. Kondo, Prog. Theoret. Phys. 27, 772 (1962).
 A. Fert and A. Friederich, AIP Conf. Proc. 24,
- 466 (1974); R. Asomoza, G. Creuzet, A. Fert, and R. Reich, Solid State Commun. 18, 905 (1976).
- 27. G. Bergman, Phys. Rev. B15, 1514 (1977).