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CURIE TEMPERATURES OF AMORPHOUS RFe_2 ALLOYS

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ABSTRACT

Curie temperatures of the series of amorphous rare earth-iron alloys RFe_2 (where $\text{R} = \text{Gd}, \text{Tb}, \text{Dy}, \text{Ho}, \text{Er}, \text{and Y}$) have been determined from Belov-Goryaga plots of the magnetization isotherms. The Curie temperature of GdFe_2 is 500 K, and drops sharply (e.g. ErFe_2 , $T_c = 105 \text{ K}$) as one proceeds to the right in the above series (decreasing R spin). The observed Curie temperatures exhibit a smooth variation with the DeGennes factor of the rare earth ion, with the zero spin limit YFe_2 exhibiting no long-range order. This is in marked contrast to the analogous crystalline Laves phase compounds for which YFe_2 has a 535 K Curie temperature and which show a much weaker dependence on the rare earth spin. These results imply a more significant effect of the structural disorder on the direct Fe-Fe exchange versus the RKKY Fe-R and R-R couplings. A molecular field model describing these interactions has been used to calculate the expected Curie temperature for both the amorphous and crystalline series. Overall agreement with the observed T_c 's was less satisfactory for the amorphous than for the crystalline materials.

INTRODUCTION

This paper presents results for the Curie temperatures of the series of ferrimagnetically ordered heavy rare earth-iron (RFe_2) amorphous alloys prepared by Battelle Northwest Laboratories¹ using rapid dc sputtering. The Gd, Tb, and Ho alloys have been previously studied extensively by neutron² and X-ray scattering³ and have been determined to be amorphous with a structure corresponding to a dense random packing of R and Fe atoms.⁴ Previous magnetization studies⁵ on the Tb, Gd, and Y alloys over the entire range of magnetic order have provided evidence for a strong local random direction anisotropy interaction in amorphous TbFe_2 of magnitude comparable to the R-Fe exchange. This produces a large high field susceptibility and a significant lowering of the measured 0 K spontaneous magnetization due to the non-collinear R spin alignment. At low temperatures coercive fields as large as 32 KOe are observed with a sharp temperature dependence qualitatively explained by an interacting spin-cluster model.⁵ Evidence for this

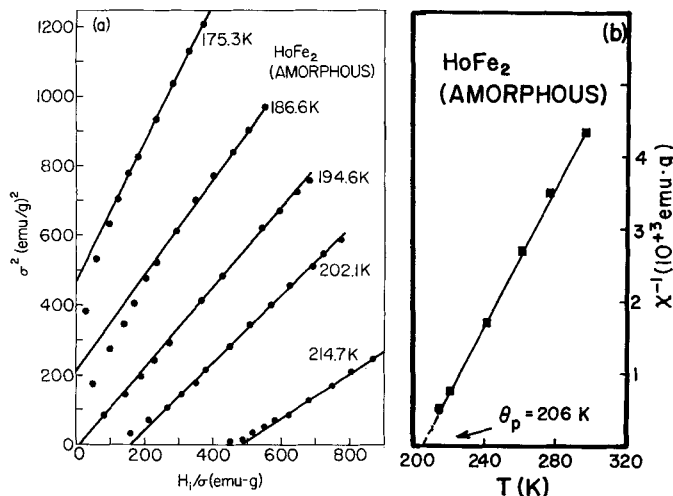


Figure 1. (a) σ^2 vs. H_1/σ plots of the magnetization isotherms for HoFe_2 near T_c . (b) Reciprocal susceptibility obtained from H/σ intercept of (a) versus temperature.

nonhomogeneous or "micro-domain" ordering of the spin system on a scale of order 100\AA is provided also by neutron small angle scattering.²

MAGNETIZATION

Magnetization data as a function of temperature for the RFe_2 series were taken using a PAR vibrating sample magnetometer and an 18 KOe electro-magnet. Isothermal magnetization data were plotted as σ^2 (σ = magnetic moment/g, units of σ : $1 \text{ emu/g} = 1 \text{ erg/(gauss-g)}$) versus H_1/σ (H_1 = applied field corrected for demagnetization) to determine the Curie temperature as shown in Figure 1a for HoFe_2 . This method (Belov-Goryaga⁶ plot) assumes the free energy near T_c can be written as a sum of second and fourth order terms in the magnetization, and it provided the most accurate determination of T_c for these amorphous alloys. Determinations of T_c from the vanishing of the spontaneous moment in the isothermal magnetization curves presents considerable uncertainty due to the field-dependent high field susceptibility near T_c . Attempts to use such extrapolations will produce a higher T_c than the 0 intercept curve on the Belov-Goryaga plot.

From the data of Figure 1a, T_c for amorphous HoFe_2 is 194K. The susceptibility above T_c taken from the H/σ intercept exhibits a linear temperature dependence for $T > 215 \text{ K}$ and extrapolates to a $\theta = 206 \text{ K}$ as shown in Figure 1b. The slope corresponds to an effective moment of $6.7 \mu_B$.

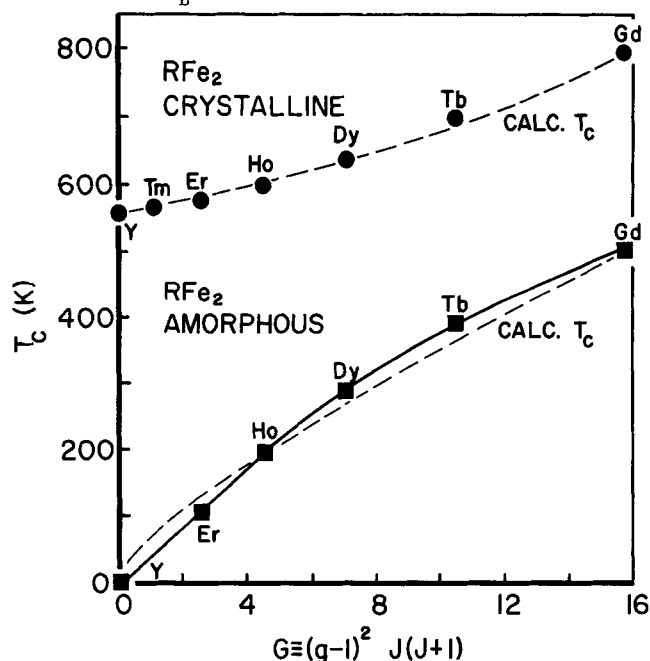


Figure 2. Curie temperatures of crystalline RFe_2 compounds and amorphous RFe_2 alloys versus DeGennes factor. The dashed line is a molecular field calculation (see text).

CURIE TEMPERATURES

The transition temperatures for the RFe_2 series ($\text{R} = \text{Gd}, \text{Tb}, \text{Dy}, \text{Ho}, \text{Er}$ and Y) in both crystalline and amorphous forms are shown in Figure 2 as a function of the effective rare earth spin squared given by the DeGennes factor $G = (g-1)^2 J(J+1)$. The crystalline compound data taken from Burzo⁷ and Buchow and Van Stapele⁸ show a large 535 K Curie temperature for

YFe_2 ($G=0$) which increases monotonically as the rare earth spin moment increases to 793 K for GdFe_2 . In contrast, the amorphous alloys show a much stronger dependence of T_c on rare earth spin, and anomalously show no long range magnetic order for YFe_2 . The ordering temperature of amorphous TmFe_2 could only be determined to be less than 50 K due to the complicating large coercive field behavior seen in these alloys at low temperatures. The strong dependence of T_c on rare earth spin and the anomalous vanishing of magnetic order as the rare earth spin goes to zero is quite puzzling, and suggests that the background Fe-Fe exchange (in YFe_2) is strongly quenched by the structural disorder, leaving only the R-Fe and R-R exchange to affect magnetic order. These latter two are of a long range electron polarization wave indirect exchange type which may be less effected by disorder than the Fe-Fe exchange interaction which is probably of much shorter range.

The dashed lines in Figure 2 show the results of applying a two sublattice molecular field calculation by Cullen⁹ to the crystalline and amorphous systems. Due to its intrinsic nature, the molecular field model applied to determining T_c is not expected to give accurate results for the exchange constants; however, it is useful in making comparisons between crystalline and amorphous systems. In this model the rare earth and iron sublattice magnetizations are written respectively as

$$M_R = N_R \mu_B J_B \left[\frac{J_{R-R} (g_J - 1)^2 J_{R-Fe} / N_R + J_{R-Fe} (g_J - 1) J_{Fe-Fe} / N_{Fe}}{kT} \right] \quad (1)$$

and

$$M_{Fe-Fe} = N_{Fe} \mu_B S B_S \left[\frac{J_{Fe-Fe} S M_{Fe-Fe} / N_{Fe} + J_{R-Fe} S (g_J - 1) M_R / N_R}{kT} \right] \quad (2)$$

where B_J and B_S are the Brillouin functions for rare earth total angular momentum J and for iron spin S ; J_{Fe-Fe} , J_{R-Fe} and J_{R-R} are the exchange couplings, μ_B is the Bohr magneton and N_R and N_{Fe} are the number of rare earth and iron atoms per formula unit.

Solving expressions (1) and (2) for the transition temperature gives

$$\theta_c = \frac{1}{2} \theta_c^{Fe} + J_{R-R} G / 6k + \sqrt{\left(\frac{\theta_c^{Fe}}{2} - \frac{J_{R-R} G}{6k} \right)^2 + \frac{J_{Fe-Fe}^2 S(S+1) G}{9k^2}} \quad (3)$$

where G is the DeGennes factor and $\theta_c^{Fe} = \frac{S(S+1)}{3k} J_{Fe-Fe}$

is the ordering temperature for the compound with no rare earth spin (YFe_2). Expression (3) has been fit to the observed transition temperatures of Figure 2 and the results are given by the dashed lines. The exchange constants obtained for the crystalline case were $J_{Fe-Fe} = 832\text{K}$, $J_{R-Fe} = -137\text{K}$ and $J_{R-R} = 98\text{K}$ which represents

the data very adequately. The fit for the amorphous case is less satisfactory, and yielded exchange constants of $J_{Fe-Fe} = 0$ (non-magnetic YFe_2) $J_{R-Fe} = -129\text{K}$ and $J_{R-R} = 73\text{K}$, the latter two being only slightly reduced from their crystalline counterpart values. The departures from the molecular field calculation, and the rapid rise in T_c for the amorphous alloys compared with the crystalline compounds, suggests that the Fe-Fe exchange energy may not vanish uniformly, particularly for the higher spin rare earth alloys (e.g. GdFe_2 for which $T_c = 500\text{K}$), but may be dependent on the presence of rare earth exchange to become effective.

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