Electrical resistivity of amorphous $Fe_{82}B_{18-x}Ge_x$ alloys: Coherent electron-magnon scattering contribution

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Detailed analysis of the electrical resistivity (ρ) data taken on amorphous Fe₈₂B_{18-x}Ge_x alloys in the temperature range 4.2 to 320 K reveals that physically meaningful values of the Debye temperature, structure factor, and Fermi wave vector can be deduced from these data only when the existence of an additional contribution to ρ , besides the dominant structural contribution (ρ_{str}), arising from the *coherent* scattering of conduction electrons from the long-wavelength magnons (ρ_{mag}), is duly recognized. Moreover, ρ_{mag} is found to possess values comparable in magnitude to those previously reported for crystalline ferromagnets.

In a recent paper¹ (henceforth referred to as I), a detailed quantitative analysis of the electrical resistivity (ρ) data on amorphous (a-) Fe₈₀B_{20-x}C_x alloys permitted us to draw the following conclusions. (i) Besides a dominant structural contribution ρ_{str} , there exists a significant magnetic contribution ρ_{mag} to ρ . (ii) Quenched disorder does not have any marked influence on the *coherent* scattering of conduction electrons from long-wavelength magnons. (iii) The net contribution to ρ due to the *incoherent* and *elastic* components of electron-magnon scattering is negligibly small. (iv) The resistivity-minimum phenomenon is not, as some of the existing theories claim, purely of structural origin, but is related in some way to the magnetic state of the glassy materials in question.

The principal aim of this paper is to ascertain whether or not the above findings are of more general validity than the study of a single glassy ferromagnetic alloy system would normally indicate. To accomplish this, high-precision resistivity measurements on yet another amorphous ferromagnetic alloy series, namely, a-Fe₈₂B_{18-x}Ge_x, have been performed and the data analyzed along the same lines as described in detail in I. The selection of this particular system for the type of study intended is justified on the grounds that (i) a high Fe content in the present alloy system, as in the previous one, guarantees a sizable magnetic contribution to ρ , which, in turn, ensures easy detection, and (ii) an exactly opposite variation 1,2 of the magnetic moment per alloy atom (μ) with the solute concentration x in the two alloy series is expected to be reflected, particularly in the concentration dependence of those quantities that have a direct bearing on μ . Moreover, the latter prediction provides a consistency check for our earlier data analysis.¹

Amorphous $Fe_{82}B_{18-x}Ge_x$ (x = 2,4,6) alloy ribbons of ~ 1 mm width and $\sim 35 \ \mu$ m thickness were prepared by the melt-spinning technique and procured from the General Electric Company, New York. Highly precise electrical resistivity measurements were performed on these alloys in the temperature range 4.2 to 300 K using a four-probe dc method. The details of the experimental setup and its sensitivity are given in I.

Figure 1 depicts the temperature variation of the normalized resistivity ratio $r(T) = \rho(T)/\rho(T_0)$, where T_0 =273.15 K is the ice point, in the temperature range 4.2 to 320 K for a-Fe₈₂B_{18-x}Ge_x alloys with x = 2, 4, and 6. The total fractional change in resistivity within the investigated temperature range varies from 4% to 6% and increases with increasing x, a concentration dependence just the opposite of that found¹ in a-Fe₈₀B_{20-x}C_x alloys. At low temperatures (10-20 K), ρ as a function of temperature goes through a well-defined minimum at a temperature T_{min} , which increases linearly with x (inset in Fig. 1). Figures 2-4 demonstrate that the variation of resistivity with temperature in the specified temperature ranges is well described by the empirical relations

$$r(T) = \beta_0 + \beta \ln T, \ 4 \lesssim T \lesssim 12 \text{ K} \ (T < T_{\min}) \ ,$$
 (1)

 $r(T) = \alpha_0 + \alpha_2 T^2$, $50 \le T \le 100$ K $(T > T_{\min})$, (2) and

$$r(T) = a_0' + a_1'T + a_2'T^2, \ 200 \lesssim T \le 300 \text{ K}$$
 (3)

Least-squares (LS) fits to the r(T) data based on Eqs. (1)-(3) yield values for the coefficients that are plotted against the Ge concentration x in Fig. 5.



FIG. 1. Temperature of r(T) for $a - \text{Fe}_{82}B_{18-x}\text{Ge}_x$ alloys. Inset depicts the concentration dependence of T_{min} .

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 $[\rho(T) - \rho(T_{\min})]/\rho(T_{\min})$ FIG. 2. vs $\ln T$ for a- $Fe_{82}B_{18-x}Ge_x$ alloys. Straight lines through the data points serve as a guide to the eye.

With a view to unraveling the scattering mechanisms basically responsible for the observed resistivity behavior particularly for $T \gg T_{\min}$ (i.e., $50 \le T \le 300$ K), we employ two distinctly different approaches to interpret the r(T) data. In the first approach, the normalized resistivity data are fitted to the expression¹

$$r(T) = a_1 + a_2 \exp\left[-8W(0)\left(\frac{T}{\Theta_D}\right)^2 \int_0^{\Theta_D/T} \frac{zdz}{e^z - 1}\right]$$
$$\equiv f_1(T) \quad , \tag{4a}$$

where

$$W(0) = 3\hbar^2 k_F^2 / 2M k_B \Theta_D \tag{4b}$$

is the Debye-Waller factor at 0 K, M is the atomic mass, Θ_D is the Debye temperature, and the remaining symbols have their usual meaning, given by the diffraction model,³



T ² $[\rho(T) - \rho(T_{\min})] / \rho(T_{\min})$ FIG. 3. for vs a - $Fe_{82}B_{18-x}Ge_x$ alloys. Continuous straight lines through the data points are intended to serve as a guide to the eye.



FIG. 4. r'(T) vs T for a-Fe₈₂B_{18-x}Ge_x alloys. Inset displays the concentration dependence of Θ_D ; open circles, values of Θ_D obtained from $f_1(T)$ fits; filled circles, Θ_D values deduced from $f_2(T)$ fits; crosses, values derived from the asymptotic fits [Eq. (7) of the text].



FIG. 5. α'_1 , α'_2 , α_2 , β , α_3 and μ as functions of Ge concentration.

which completely ignores the magnetic contribution to ρ . In the second approach the expressions for $\rho_{str}(T)$ and $\rho_{mag}(T)$, derived from the theories based on the diffraction model^{1,3} and the spin-disorder (SD) model, ^{1,4} respectively, are used in the relation (Matthiessen's rule)

$$\rho(T) = \rho_{\text{str}}(T) + \rho_{\text{mag}}(T) \quad , \tag{5}$$

whose validity for amorphous ferromagnets is claimed by the theory,⁴ in order to take into account both structural (ρ_{str}) and magnetic (ρ_{mag}) contributions to the total resistivity, and theoretical fits to the data of the type

$$r(T) = f_1(T) + a_3 T^2 \equiv f_2(T)$$
, (6a)
and

$$r(T) = f_2(T) + a_4 T^{3/2} , \qquad (6b)$$

where $f_1(T)$ is given by Eq. (4a), are attempted. Justification for the existence of T^2 and $T^{3/2}$ terms in Eqs. (6a) and (6b) is provided by the theoretical calculations,⁴ based on the SD model, which yield an expression for $\rho_{mag}(T)$ in amorphous ferromagnets at low temperatures that contain two terms of significant magnitude:¹ One of them denotes the net outcome of two competing contributions, both varying as $T^{3/2}$, due to incoherent (momentum-nonconserving) and elastic components of electron-magnon scattering, and the other represents the *coherent* spin-wave contribution, which varies as T^2 . This model also predicts that the $T^{3/2}$ term is at least two orders of magnitude greater than the T^2 term so that $\rho(T) \propto T^{3/2}$ in amorphous ferromagnets at low temperatures. By contrast, we find a T^2 dependence at low temperatures (Fig. 3) in the glasses under consideration. This observation, coupled with the fact that the LS fits based on Eq. (6b) yield unphysically large values for k_F and Θ_D and a *negative* value for a_4 , strongly suggests that the $T^{3/2}$ term, if present, must be much smaller in magnitude compared to the T^2 term. We now concentrate on the relative merits of the LS fits based on Eqs. (4a) and (6a). In order to make such an assessment possible, the two sets of values of the fit parameters $a_1, a_2, W(0)$, and Θ_D , and the values of a_3 , determined from the nonlinear LS fits involving Eqs. (4a) and (6a), and the corresponding values of

 k_F and the structure factor at 0 K, $S_0(2k_F)$, deduced from them using Eq. (4b) and the relation¹ $S_0(2k_F) = 1 + (a_2/a_1)$, respectively (Table I), are then used to compute the theoretical values of the fractional change of resistivity at 300 K, $\Delta \rho / \rho = [\rho (300 \text{ K}) - \rho (0 \text{ K})]$ K)]/ $\rho(0$ K), the normalized resistivity at 300 K, r(300K), and the temperature coefficient of resistivity (TCR) α'_1 from the expressions given in I. Theoretical values so obtained are compared with the observed ones. Such a comparison demonstrates that the inclusion of the magnetic contribution [the coherent electron-magnon scattering term a_3T^2 in Eq. (6a)] in the expression for total resistivity does not significantly change the values of a_1, a_2 [and hence of $S_0(2k_F)$], $\Delta \rho / \rho$, and r(300 K) but considerably improves the quality of the fits as inferred from the reduced (by an order of magnitude) value of the sum of squares (χ^2) . It completely removes the large discrepancy between the observed values of α'_1 and those deduced from the $f_1(T)$ fits, and gets rid of the unphysical magnitudes of k_F and Θ_D , so that, in accordance with the predictions of the diffraction model, ^{1,3} positive TCR is now associated with $2k_F$ values that are either well below or well above $k_p \approx 3.1$ Å⁻¹ [the value of k at which the first peak in the S(k) vs k curves occurs¹]. Θ_D now possesses values reasonably close to $T^* \simeq 200$ K, the temperature above which a linear term lignoring for the moment the small T^2 term in Eq. (3)] in r(T) is observed. Foregoing observations thus point to the existence of a small but finite magnetic contribution, besides a dominant structural contribution, to ρ in the glasses in question.

Based on the theoretical prediction⁴ that Matthiessen's rule [Eq. (5)] holds for amorphous ferromagnets, we contend that the magnetic contribution to ρ escapes detection at low temperatures because $\rho_{str}(T)$ and $\rho_{mag}(T)$ both follow a T^2 power law in that temperature range, but shows up very clearly at high temperatures (i.e., $T \gtrsim T^*$) where $\rho_{str}(T)$ exhibits a transition from a quadratic to a linear temperature dependence while $\rho_{mag}(T)$ continues to vary as T^2 . This implies that we ascribe the terms $\alpha_2 T^2$, $\alpha'_1 T$, and $\alpha'_2 T^2$ of the low- and high-temperature fits, Eqs. (2) and (3), to $\rho_{str}(T) + \rho_{mag}(T)$, $\rho_{str}(T)$, and

TABLE I. Parameter values for amorphous $Fe_{82}B_{18-x}Ge_x$ alloys obtained by fitting the resistivity data to Eqs. (4a) and (6a) of the text in the temperature interval $50 \le T \le 300$ K. The least-squares fits based on Eq. (6a) yield the parameter values given within the square brackets whereas the numbers in the parentheses denote estimated uncertainty in the least significant figure.

x (at. %)	<i>a</i> 1	<i>a</i> 2	W(0)	θ _D (K)	$S_0(2k_F)$	<i>k_F</i> (Å ⁻¹)	χ^2 (10 ⁻⁹)	(10^{-8} K^{-2})	d (g/cm ³)	μ /alloy atom at 77 K Units of μ_B
2	1.307(6) [1.305(16)]	-0.341(6) [-0.339(16)]	0.0383(9) [0.0200(10)]	503(4) [362(3)]	0.739(17) [0.740(50)]	3.60(5) [2.21(6)]	11.758 [1.037]	[8.49(21)]	7.52ª	1.6974(100) ^a
4	1.399(10) [1.3980(8)]	-0.438(10) [-0.4380(8)]	0.0316(11) [0.0085(2)]	481(6) [223(4)]	0.687(25) [0.687(2)]	3.24(6) [1.14(2)]	43.971 [2.542]	[15.25(16)]	7.61 ª	1.7220(100) ^a
6	1.821(9) [1.822(94)]	-0.866(9) [-0.868(94)]	0.0176(3) [0.0073(8)]	467(4) [293(6)]	0.524(11) [0.523(120)]	2.41(2) [1.23(7)]	32.755 [6.533]	[14.10(43)]	7.70ª	1.7730(100)ª

^aReference 2.

 $\rho_{mag}(T)$, respectively. Such an identification of various terms in Eqs. (2) and (3) allows calculation of the Debye temperature Θ_D via the relation¹

$$\Theta_D \simeq (\pi^2/6) [\alpha_1'/(\alpha_2 - \alpha_2')] \quad . \tag{7}$$

Additional findings that vindicate our claim of a significant magnetic contribution to ρ are (i) the inset of Fig. 4 demonstrates that the Θ_D values derived from the asymptotic fits, based on Eqs. (2) and (3), through Eq. (7) are in reasonably good agreement with those deduced from the fits based on Eq. (6a), and (ii) the high-temperature and $f_2(T)$ fits yield roughly the same value (and hence the same concentration dependence, see Fig. 5) for the coefficient of the magnetic term [the T^2 term in Eqs. (3) and (6a)]. Moreover, the values of this coefficient in the expression of total resistivity, i.e., $a' = \rho(T_0)a_3$ or $a'' = \rho(T_0)a'_2$, for the present glassy alloys, as for a- $Fe_{80}B_{20-x}C_x$ alloys, conform well with those found in crystalline ferromagnets (Fe, Co, Ni, and their alloys),^{5,6} e.g., the value $a' = (1.02 \pm 0.03) \times 10^{-11} \Omega \text{ cm K}^{-2}$ found in this work for $a - Fe_{82}B_{16}Ge_2$ is in excellent agreement with the value $0.96 \times 10^{-11} \ \Omega \ \text{cm K}^{-2}$ previously reported⁵ for crystalline Fe. From this agreement we conclude that the quenched disorder has practically no influence on the coherent electron-magnon scattering contribution to ρ .

Finally, we focus our attention on the concentration

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dependence of α'_1 , α'_2 , α_2 , β , α_3 , and μ (Fig. 5). α_3 and α'_2 are noted to have a strikingly similar x dependence which completely differs from that of α'_1 and α_2 . This strongly suggests a common origin (presumably magnetic in nature) for the terms in r(T) with coefficients a_3 and a'_2 . At variance with our earlier results, ¹ the coefficients a_3 and α'_2 do not scale with μ and the coefficient β exhibits a variation with x that is the reverse of that of $\mu(x)$. This probably reflects the different nature of the substitution processes in the two alloy systems, i.e., the replacement of B by C in a-Fe₈₀B_{20-x}C_x alloys occurs at the interstitial position, whereas that by Ge in the present alloy system takes place at the substitutional position.² Another important result which deserves attention is that all the above-mentioned quantities for the glassy alloys under consideration exhibit a concentration dependence which is just the opposite of that found by us in a-Fe₈₀B_{20-x}C_x alloys. In view of the arguments forwarded in the beginning of this paper, this observation lends firm support to our earlier data analysis. To summarize, all our previously drawn conclusions¹ are found to be characteristic of a wide class of glassy ferromagnetic alloys.

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