Universal collective effect in the low-temperature resistivity of concentrated amorphous spin glasses

J. Kötzler
Institut für Angewandte Physik, Universität Hamburg, D-20355 Hamburg, Germany

G. Thummes
Institut für Angewandte Physik, Universität Hamburg, D-20355 Hamburg, Germany
and Institut für Angewandte Physik, Justus-Liebig-Universität, D-35392 Giessen, Germany

R. Carloff
Institut für Angewandte Physik, Universität Hamburg, D-20355 Hamburg, Germany
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The linear upturn of the electrical resistivity, $\Delta\rho = -C_1T$, first reported below the freezing temperature T_f of $Fe_{2.4}Ni_{77.6}Si_8B_{12}$, is shown to be a rather universal feature of metallic glasses containing localized $3d$ and $4f$ moments in a wide range of concentrations. The normalized amplitudes, C_1T_f, are almost independent of composition and the magnetoresistance also indicates that impurity scattering of the conduction electrons is not effective below T_f. We discuss these results by assuming that the spin-glass order parameter $q \sim (1 - T/T_f)$ increases the resistivity structure factor of the amorphous matrix.

I. INTRODUCTION

The appearance of a resistivity minimum at low temperatures is a widely observed feature of nonmagnetic and magnetic amorphous alloys and of strongly disordered metals. One universal mechanism responsible for the upturn of $\rho(T)$ towards zero temperature is the reduction of the screening of the interaction between the conduction electrons due to their slow diffusion in the disordered structure. This is determined by the thermal length $L_T = (\hbar D/k_BT)^{1/2}$ of the diffusing electrons giving rise to an increase as $\sqrt{T}$,[1]

$$\frac{\Delta\rho_i(T)}{\rho^2} = -\frac{e^2}{4\pi^2\hbar}\frac{1}{L_T} = -C_i\sqrt{T}. \quad (1)$$

The coefficient is a quasiuniversal number, $C_i = 6\,(\Omega\,\mathrm{cm\,K}^{1/2})^{-1}$, which has been realized in an impressive number of disordered alloys[2] including the metallic glasses $Fe_xNi_{80-x}Si_8B_{12}$ (Ref. 3) of interest here.

In this paper, we investigate another, possibly universal origin giving rise to a resistivity minimum, namely, the linear upturn of $\rho(T)$ discovered recently[3] in the conventional spin-glass phase, existing for $x = 2.4$ in the above metallic glass series,

$$\Delta\rho(T \to 0) = -C_1\,T. \quad (2)$$

This linear law replaced the $\sqrt{T}$ law, Eq. (1), which reappeared in the reentrant spin glasses of this series, i.e., for Fe concentrations above the critical concentration for ferromagnetism, $x_c = 4.0$.[4] Based on a theory by Fischer[5] this linear upturn was tentatively related to elastic scattering from the spin-glass order parameter q.[6] Since, in the fluctuationless regime, q may be approximated by a linear (mean field) law,[7,8]

$$q(T) = s^2(1 - \alpha T/T_f), \quad (3)$$

where $\alpha \approx 1$, with $\Delta\rho \sim q$,[5] the linear upturn can be explained.

In order to shed some more light on the mechanism behind the linear upturn of $\rho(T)$ we report here on the magnetoresistance of $Fe_xNi_{80-x}Si_8B_{12}$. We find the low-temperature magnetoresistivity to be completely determined by the magnetization and any contributions from quantum corrections[1] can be ruled out. These data indicate that the coupling of the conduction electrons to the order parameter q cannot be explained by sd scattering from independent impurities as proposed by the current theory,[7,8] but that some collective effect of the spin-glass state in the amorphous structure becomes relevant. This conjecture will be supported by reanalyses of low-temperature resistivities, published for a number of quite different amorphous spin glasses, which previously were unsuccessfully compared to Kondo-like behavior, $\Delta\rho \sim \ln T$. We will show that these materials, containing a wide range of concentrations of magnetic $3d$ and $4f$ moments, also display the linear upturn, Eq. (2).

II. RESULTS FOR $Fe_xNi_{80}Si_8B_{12}$ WITH $x \approx x_c$

The metallic glasses studied here and the measuring technique for ρ have been described in Ref. 3. In order to characterize the magnetic state and to interprete the magnetoresistance data we also investigated the magnetization using a home-made superconducting quantum interference device (SQUID) magnetometer[9,10] at low field and an induction magnetometer[11] in fields up to 5 T.

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The temperature variation of the resistivity and of the dc susceptibility in the conventional spin glass ($x = 2.4 < x_c$) are reproduced in Fig. 1. Based on the diffraction model, the T^2 increase has been ascribed[3] to the additional backscattering from phonons,

$$\rho(T) = \frac{\hbar}{e^2 k_F} S(2k_F, T), \tag{4}$$

where the interaction between conduction electrons and the lattice potential has been absorbed in the structure factor S of the amorphous matrix. Immediately below the freezing temperature, $T_f = 6.2$ K, as defined by the cusp of the zero-field susceptibility, $\rho(T)$ starts to rise linearly down to the lowest measured temperature of 0.35 K.

The low-field susceptibility obeys the Curie-Weiss law, $\chi = \lambda_C/(T - 5.0\ \text{K})$, down to temperatures close to T_f; see inset to Fig. 1. The large Curie constant, $\lambda_C = 2.7$ K, signals superparamagnetism and, in fact, taking an experimental estimate for the (technical) saturation magnetization due to the Fe impurities, $M_0 = 0.8$ kOe,[10] we obtain large moments of $160(5)\mu_B$ of concentration $x_s = 5 \times 10^{-4}$. Apparently, these clusters result from the polarization of the Ni matrix by the localized Fe spins which freeze into random directions at T_f. We found that the ferromagnetic alloy with $x = 5.6$ also exhibits the Curie-Weiss law for $T \geq T_c = 48$ K, and by using $M_0 = 2$ kOe,[10] we obtain the same superparamagnetic moments, $160(5)\mu_B$, of concentration $x_s = 1.1 \times 10^{-3}$.

The low-field magnetoresistance (MR) measured in the paramagnetic phase of both materials and also below T_f for $x = 2.4$ is shown in Fig. 2(a). Clearly, the resistance decreases precisely proportional to M^2, and after normalization to the saturation moment, the MR turns out to be proportional to the Fe content,

$$\Delta\rho(T, H) = [-0.10(1)\ \mu\Omega\,\text{cm}]x\ m_z^2(T, H), \tag{5}$$

where $m_z = M/M_0$. This finding suggests a comparison with the theory of Béal-Monod and Weiner[12] on the MR in dilute alloys based on independent sd scattering from single localized moments. More recently, this approach was extended by Fischer[5] to dilute spin glasses. Ignoring any inelastic scattering at the present low fields, these results can be summarized by the form

$$\Delta\rho(T, H \to 0) = c\rho_{sd}\left[\frac{q(T)}{s^2} - 4\ m_z^2(T, H)\right]. \tag{6}$$

Using $\rho_{sd} = (m^*/n_{el}e^2\hbar)(J_{sd}s)^2\ N(\epsilon_F)$ and taking

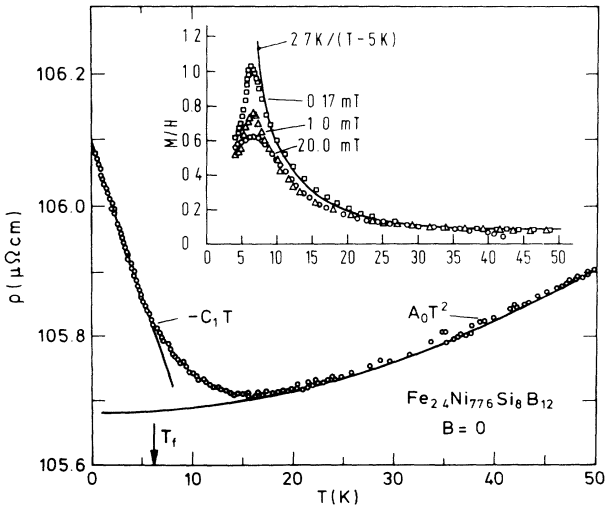

FIG. 1. Zero-field resistivity and low-field susceptibility (inset) about the spin-glass freezing temperature $T_f = 6.2$ K of $Fe_{2.4}Ni_{77.6}Si_8B_{12}$.

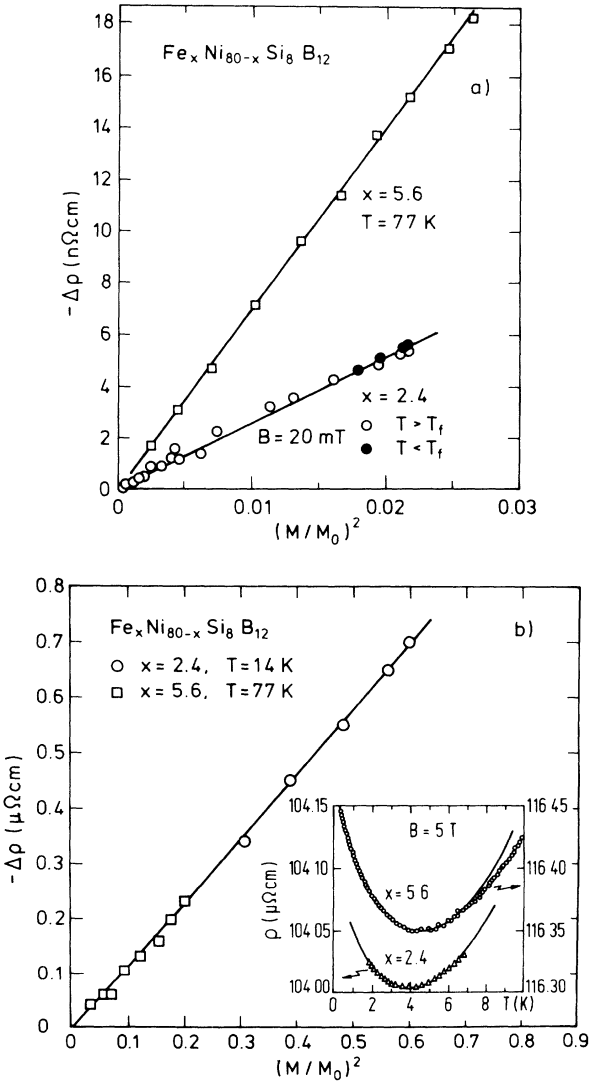

FIG. 2. Magnetoresistance of $Fe_xNi_{80-x}Si_8B_{12}$ in the paramagnetic states of the conventional ($x = 2.4$) and the reentrant ($x = 5.6$) spin glass versus squared reduced magnetization: (a) at low fields, including some data for $T < T_f$; (b) at high fields: Inset shows for $x = 2.4$ the recovery of the universal $\sqrt{T}$ law at high fields which is present in $x = 5.6$ at all fields; solid lines are fit to Eq. (7).

$c = x/80$, $N(\epsilon_F) = 4.5$ states/(atom eV) and for the effective mass of the conduction electrons in the present alloy $m^* = 10m_e$ from Ref. 3, we find a rather reasonable value for the *sd* exchange, $J_{sd}s = 0.1$ eV. As a rather remarkable feature we note that the second term in Eq. (6) also accounts for the low-field MR at temperatures slightly below T_f, where the magnetization decreases at constant field (see inset to Fig. 1). Apparently, the small field does not yet influence the spin-glass order parameter q itself; however, the finite q reduces the magnetization and hence increases the disorder scattering.

Another consequence following from the impurity model is the relation between the coefficients of the measured MR, Eq. (5), and of the linear upturn C_1 [Eq. 2]. After inserting $q/s^2 = 1 - \alpha\, T/T_f$ [Eq. 3] into Eq. (6), we obtain $C_1 = -(\alpha/4T_f)d\rho/dm_z^2 = 0.010\ \mu\Omega$ cm/K, to be compared to the experimental value, $C_1 = 0.046\ \mu\Omega$ cm/K. Since the present theoretical evidence is $\alpha \sim 1$,[7,8] the impurity model significantly underestimates the linear upturn by a factor of almost 5. We take this a hint that the disorder scattering in the spin-glass phase is significantly stronger than expected from the model of independent impurities.

This impurity model also breaks down at high fields even in the paramagnetic state, where the MR becomes independent of the Fe concentration. According to Fig. 2(b), the reduction of ρ is again proportional to the square of the reduced magnetization, $\Delta\rho(T,H) = -1.25(20)m_z^2\ \mu\Omega$ cm but independent of x, which we ascribe to a reduction of the disorder *sd* scattering from the superparamagnetic moments. In fields up to 5 T, the Fe moments are fully aligned, and the Ni matrix is partially polarized. In the spin-glass phases of both materials, i.e., the conventional one for $x = 2.4$ and the reentrant one for $x = 5.6$, the resistivity around the minima, displayed in the inset to Fig. 2(b), can be fitted to

$$\Delta\rho(T, H = 50\ \text{kOe}) = A\ T^2 - C\ \rho^2(T_f)T^{1/2}, \qquad (7)$$

with concentration independent coefficients for the T^2 law, $A = 2.3(1)$ nΩ cm/K^2 and for the interaction term, Eq. (1), $C_i = 6.1(3)$ (Ω cm K$^{1/2}$)$^{-1}$, the latter agreeing with the quasiuniversal value.[2] These high-field data imply that the spin-glass states have been destroyed and that only the scattering from the disordered Ni background survived.

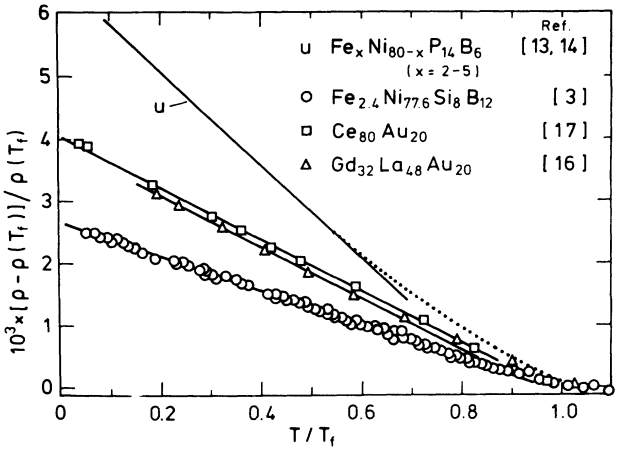

FIG. 3. Low-temperature resistivity below the freezing temperatures T_f of various amorphous spin glasses.

III. UNIVERSALITY OF THE LINEAR UPTURN

Resistivity minima have been reported for a number of spin glasses hosted by amorphous transition metals like Ni,[13,14] Pd,[15] and rare earth Au alloys.[16,17] In the literature, these data were depicted vs ln$T$ in order to search for a temperature range below the minimum, where the Kondo effect is operative. None of these analyses, however, revealed clear evidence for this effect. On the other hand, if one replots these data on a linear temperature scale versus T/T_f as illustrated by Fig. 3, well defined "linear" regions are realized at low temperatures, which in some examples extend almost up to T_f. Deviations from the linear temperature dependence are seen in spin glasses with high T_f's, in which the high-temperature increase of ρ, AT^2, gains more importance. We made no attempt to correct the data for this effect, because fluctuations may also modify the linear T dependence of q, Eq. (2), near T_f.

Considering the fairly wide range of freezing temperatures, the quite different compositions of these alloys and the common property of all, i.e., the spin-glass phase, it is rather suggestive to associate the linear upturn with the spin-glass order parameter. The relevant parameters

TABLE I. Freezing temperatures, resistivities, and slopes of the linear upturn for various amorphous spin glasses with magnetic-ion concentrations x related to the critical concentration for ferromagnetism x_c.

Sample	$\frac{x}{x_c}$	T_f (K)	$\rho(T_f)$ ($\mu\Omega$ cm)	C_1 (nΩ cm/K)	C_1T_f ($\mu\Omega$ cm)	Ref.
$Fe_{2.4}Ni_{77.6}Si_8B_{12}$	0.60	6.2	106	46.6	0.29	3
$Fe_{5.6}Ni_{74.4}Si_8B_{12}$	1.4	19.4	118	13.4	0.26	3
$Fe_2Ni_{78}P_{14}B_6$	0.26	4.0	$\simeq 130$	245	0.98	13,14
$Fe_5Ni_{75}P_{14}B_6$	0.64	10.0	$\simeq 130$	98	0.98	13,14
$Fe_5Pd_{72.5}Si_{16.5}Cu_6$	0.7(1)	15.0	113	43.8	0.66	15
$Gd_{32}La_{48}Au_{20}$	0.6	21.5	277	54.0	1.16	16
$Ce_{80}Au_{20}$	-	1.7	153	376.0	0.64	17

T_f, $\rho(T_f)$, and C_1 are listed in Table I. One striking feature of these data is the discovery of the scaling relation $\Delta\rho(T)/\rho(T_f) = f(T/T_f)$ for the alloy $Fe_xNi_{80-x}P_{14}B_6$ by Rao and co-workers[13,14] holding for $x \leq 5$. This implies that the amplitude of the linear law, valid at low temperatures, $\Delta\rho(T \to 0) = -(C_1T_f)T/T_f$, is independent of the Fe concentration. This fact strongly corroborates our conjecture based on the MR of the related spin-glass alloy, $Fe_{2.4}Ni_{77.6}Si_8B_{12}$, that the linear upturn cannot be ascribed to impurity scattering of the conduction electrons. It should rather be related to the spin-glass phase in the amorphous matrix as a whole. This point of view is further substantiated by the fact that the products C_1T_f obtained for all these alloys (see Table I) vary within a rather narrow interval from 0.3 $\mu\Omega$ cm to 1.1 $\mu\Omega$ cm with respect to the wide range of $3d$ and $4f$ concentrations and compositions.

IV. DISCUSSION AND CONCLUSIONS

The results presented here for resistivity and magnetoresistance in the spin-glass phase of various transition metal based amorphous alloys provide fairly convincing evidence that the spin-glass order parameter q determines the scattering of the conduction electrons at low temperatures. To the best of our knowledge, the existing theory[5,8] rests on independent *sd* scattering from localized impurities which acquire spin-glass order. Since this model appears to be inappropriate for the present amorphous and rather concentrated materials, we propose a crude alternative. We empirically assume that the appearance of q modifies the diffraction scattering of the conduction electrons from the amorphous structure. If the exchange coupling of the localized spins to the lattice is weak compared to the Coulomb interaction, we may expand the structure factor, Eq. (4), with respect to q to obtain in first order

$$S(2k_F, q(T)) = S(2k_F, 0) + S'(2k_F, 0)q(T), \qquad T < T_f. \tag{8}$$

In conjunction with Eqs. (3) and (4), this ansatz can explain the linear upturn, the quasiuniversal coefficient of which is then given by $C_1T_f = (s^2\hbar/e^2k_F)S'$, where we set again $\alpha = 1$. Furthermore, this ansatz implies that this coefficient is independent of the concentrations of the localized spins. We propose to associate this collective effect with the superparamagnetic clouds, present for concentrations near the critical one (or 80% Ce in the case of $Ce_{80}Au_{20}$), which in the frozen state transfer the disorder to the whole matrix. The observed slight variation of C_1 T_f between the different materials (see Table I) would then arise from the different coupling constants between the frozen spin and the amorphous lattice. The upturn of $\rho(T)$ demands that the resistivity structure factor increase at the onset of the spin disorder at T_f, i.e., $S' > 0$, which is not unplausible because the thermal disorder also increases S' giving rise to the T^2 law above the resistivity minimum.[3] Of course, these entirely qualitative arguments require more detailed considerations.

Using this model, we now ascribe the observed suppression of the linear upturn in high magnetic fields [inset to Fig. 2(b)] to the full alignment of the localized moments, i.e., $q = 0$. Then the diffusing conduction electrons do not feel any additional disorder near the dominant wave number $k = 2k_F$ and the universal $\sqrt{T}$ law is recovered. Then the question arises why this interaction mechanism, Eq. (1), does not work at low fields: The data for the conventional spin glass $Fe_xNi_{80-x}Si_8B_{12}$ with $x = 2.4$ exclude this contribution. We tentatively associate this interesting phenomenon with the strong random exchange fields $J_{ds}s$ acting on the conduction electrons by the freezing moments. If these fields fluctuate within the thermal diffusion length, which for $x = 2.4$ is limited from below by $L_T(T_f) = 43$ Å, then the increase of L_T towards lower temperature loses its significance since, roughly speaking, the thermal disorder is overruled by the magnetic disorder.

This picture can also be used to discuss the case of the reentrant spin glass, $x = 5.6$, where the $\sqrt{T}$ law dominates the low-temperature resistance.[3] In this material it is very likely that locally the ferromagnetic order remains on length scales larger than $L_T(T_f) = 24$ Å, so that the electron feels a fairly homogeneous exchange field within the thermal length and hence there is no cutoff of the interaction effect. With decreasing temperature L_T rises, and we expect the linear upturn to gain importance if L_T reaches the extension of the residual ferromagnetic regions. In fact, there is some indication of such a thermal crossover for $Fe_{5.6}Ni_{74.4}Si_8B_{12}$: In Ref. 3, the fit of $\rho(T)$ about the minimum, which considered only the interaction term, produced $C_i = 8.4$ $(\Omega$ cm $K^{1/2})^{-1}$ which is larger than the mean $C_i = 6.3$ $(\Omega$ cm $K^{1/2})^{-1}$ for the ferromagnetic concentrations of the $Fe_xNi_{80-x}Si_8B_{12}$ alloy. Including now a linear term into the fit of the data, we find $C_i = 6.4$ $(\Omega$ cm $K^{1/2})^{-1}$ and $C_1T_f = 0.26$ $\mu\Omega$ cm in very good agreement with the 0.29 $\mu\Omega$ cm obtained for $x = 2.4$. In the present model this agreement implies that the coupling parameter S' between the Fe spins and the Ni matrix as defined by our basic ansatz, Eq. (8), is essentially the same in the conventional and reentrant spin glass. We believe this further supports our central suggestion that the linear upturn arises from a change of the electron diffraction scattering in the amorphous structure driven by the spin-glass order parameter.

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