Absolute thermoelectric power in amorphous $Fe_x Ni_{80-x} B_{19}Si_1$ alloys

W. Kettler, S. N. Kaul, and M. Rosenberg

Institut für Experimentalphysik VI, Ruhr-Universität Bochum, D-4630 Bochum, West Germany

The absolute thermoelectric power S of glassy $Fe_x Ni_{80-x} B_{19}Si_1$ ($x = 10,13,16$) alloys has been measured from 4.2 to 300 K. Of all the available transport theories, only the Mott s-d scattering and Kasuya models reproduce reasonably well the experimental results on composition and temperature dependence of S for temperatures below T_C, the Curie temperature. Both these models, however, fail to account for the observed behavior of S for $T \geq T_C$. Arguments are presented to show that the nonlinear thermoelectric power (TEP) in magnetic glasses is basically due to the dominant contribution to TEP arising from the magnetic scattering from magnetic atoms for $T \lesssim T_C$.

I. INTRODUCTION

Several theoretical models that include (1) the extended Faber-Ziman diffraction model,[1] (2) the Mott s-d scattering model,[2] (3) the pseudo-Kondo-model based on the Kondo-type scattering from a two-level tunneling system,[3] and (4) the resurrected Kondo model[4] (applicable only to the magnetic glasses) have been proposed to understand the electronic transport in metallic glasses. While these models have all been able to reproduce the observed features of the electrical resistivity (ρ) data within some range of temperatures for certain sets of parameters in the theory, they predict entirely different behavior as far as the thermoelectric power (TEP) is concerned.[5] A study of TEP, therefore, provides a powerful method of finding out which of the above-mentioned theories adequately describes the transport properties of metallic glasses. For this reason, the experimental interest in the recent years has shifted from the study of ρ to that of TEP in such materials.

A quantitative comparison between the predictions of the available transport theories and the experimental results on composition and temperature dependence of TEP in nonmagnetic glasses[6,7] has revealed that (i) the agreement between the experiment and theory is reasonably good for both Mott and Faber-Ziman models (in the latter case, particularly when the structure factor is allowed to vary with temperature[8]), and (ii) TEP at low temperatures ($T < 100$ K) can be understood in terms of the electron-phonon mass enhancement.[6] By contrast, the situation in the case of magnetic glasses is not at all clear, primarily because most of the presently available TEP data have been taken on alloys with completely different constituents.[9,10] Such a heterogeneous collection of TEP data has found a tentative explanation in terms of a resurrected Kondo model[10] based on the Kondo scattering of conduction electrons by spins situated in the tail of the effective internal field distribution near zero effective field. In spite of the fact that a TEP behavior similar to that found in crystalline ferromagnets Fe, Co, and Ni has been observed in these glassy ferromagnetic alloys, no account has been taken of the contribution to TEP arising from the magnetic scattering. More recently, Korenblit[11] has pro-

posed a theory for TEP in amorphous ferromagnets which leads qualitatively to the TEP behavior generally observed in such materials, but the major predictions of this theory remain yet to be verified.

In this paper, the results of the TEP measurements performed on amorphous $Fe_x Ni_{80-x} B_{19}Si_1$ ($x = 10,13$, and 16) (x in units of at. % throughout) alloys in the temperature range 4.2—300 K are reported and discussed in the light of the available transport theories. The success and limitations of various models to explain different aspects of our results have been pointed out. In addition, various contributions to TEP, in such magnetic glasses, due to different scattering mechanisms have been identified.

II. EXPERIMENTAL DETAILS

Amorphous $Fe_x Ni_{80-x} B_{19}Si_1$ ($x = 10,13$, and 16) alloy ribbons having a cross section of about 0.03×2 mm^2 were prepared by the rotating-drum technique and procured from the General Electric Co. (New York). TEP measurements were performed in the temperature range 4.2—300 K using a differential ac method. Both hot and cold thermocouple junctions were formed by soldering 0.1-mm-diam chromel and constantan wires (placed side by side in contact with each other) onto the sample (length $\simeq 4$ cm), and using as small a quantity of indium soldering material as possible, so that the diameter of the thermocouple bead hardly exceeded that of the thermocouple wires. Use of soldered junctions ensures not only a good thermal but also a pure Ohmic contact with the sample, and hence a measuring signal that is free from capacitive effects. With the use of the new Roberts[12] scale of thermoelectricity, the chromel and constantan wires were calibrated against lead (99.999% pure). To provide a continuous $S = f(T)$ relationship for each thermocouple, the experimental data were approximated by a series of Chebyshev polynomials in the L_2 norm. An ac temperature gradient (5—50 mK) was produced along the sample by heating the hot junction periodically ($f \simeq 4$ Hz) by means of a small strain gauge (heating area ~ 6 mm^2), which was glued to the sample surface with General Electric 7031 varnish. ac voltages resulting from the two parallel thermocouples were phase-sensitively detected, and acquired and pro-

cessed by a microcomputer. The relative resolution of the experimental setup reached 0.02 μV/K, whereas the absolute accuracy of the data was as low as 10% throughout the investigated temperature range. The sample temperature was monitored by calibrated germanium ($T < 40$ K) and platinum ($T > 40$ K) resistance thermometers. The data presented here are reproducible to within 1% when a fresh sample from the same alloy batch is used for the measurements.

III. RESULTS AND DISCUSSION

The temperature dependence of the absolute TEP, S, for the amorphous Fe$_x$Ni$_{80-x}$B$_{19}$Si$_1$ alloys with $x = 10$, 13, and 16 in the temperature range 4.2—300 K, is shown in Fig. 1. The important features of the TEP data[13] are (i) S is negative throughout the investigated temperature range, (ii) at low temperatures, S exhibits roughly a linear dependence on temperature and this linear variation when extrapolated to 0 K gives a slope value which is negative and increases with Fe concentration x, (iii) in the intermediate-temperature region, $|S|$ as a function of temperature goes through a broad peak; the peak value (S_{max}) increases while the peak position (T_{max}) shifts to higher temperatures as x increases, (iv) for $T > T_{max}$, S again varies linearly with temperature, but this time with a positive slope that, within the error limits, is independent of composition, and (v) for the alloys with $x = 10$ and 13, S does not show any anomaly in the critical region even though the present alloys are known[14,15] to exhibit a well-defined magnetic phase transition at the Curie point T_C.

Similar dependence of S on temperature has been previously observed in metallic glasses containing $3d$ transition metals (Fe, Co, and Ni).[9,10] By comparison, the nonmagnetic glasses exhibit a completely different TEP behavior, i.e., over a wide range of temperatures, S is positive and increases roughly linearly with temperature.[5,6] In view of the fact that the pseudo-Kondo model[3] does not distin-

guish between the magnetic and nonmagnetic glasses so far as the effect of structure on their transport properties (e.g., ρ and S) is concerned, the above comparison suggests that such a scattering process, if at all present, should give to TEP a contribution that is negligibly small compared to those arising from other scattering processes.
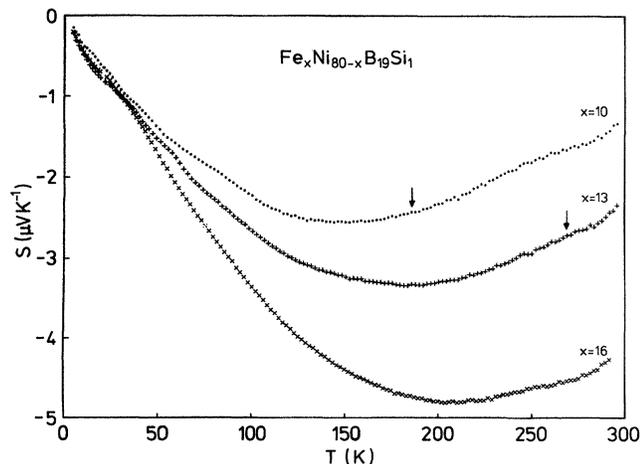
Guided by the result of the Monte Carlo calculations of the effective-field distribution $P(H)$ in glassy ferromagnets,[4] namely that even in a concentrated amorphous ferromagnet a small fraction of magnetic atoms can exist in the regions of zero effective field, Nagel and his co-workers[10] argue that the Kondo scattering of electrons from such moment-bearing atoms accounts not only for the coexistence of Kondo-type resistivity minimum and ferromagnetism in these amorphous materials, but also for their nonlinear TEP, and hence for the difference in the TEP behavior found in magnetic and nonmagnetic glasses. On the basis of this argument, for the alloys with $x = 10$—16 in the amorphous Fe$_x$Ni$_{80-x}$B$_{19}$Si$_1$ alloy system [i.e., in the concentration range where the values of both T_C and the magnetic moment fall very rapidly with decreasing x (Ref. 16)] the peak of $P(H)$ should not be far from $H = 0$, and as such $P(H)$ will vary rapidly around the origin, and the entire curve should shift to higher values of H as x increases. In such a case, one would expect a significant reduction in the Kondo effect as x increases from 10 to 16, a prediction that obviously contradicts our observations (see Fig. 1). Again, the interpretation of $S(T)$ data in terms of a pure Kondo effect is not justified on the grounds that (i) the value of the ratio $(\rho_{4.2K} - \rho_{min})/\rho_{4.2K}$ does not change significantly[14] in the

FIG. 1. TEP, S, of amorphous Fe$_x$Ni$_{80-x}$B$_{19}$Si$_1$ alloys as a function of temperature: The arrows denote the Curie temperature T_C for the alloys with $x = 10$ and 13.
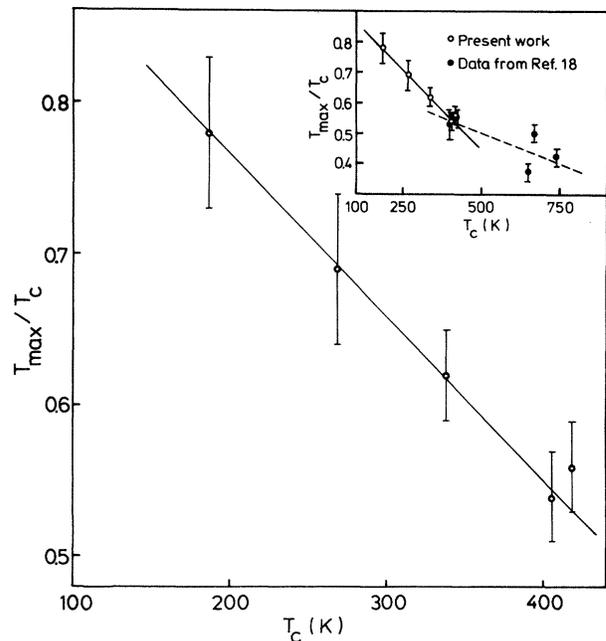
FIG. 2. T_{max}/T_C vs T_C for glassy Fe$_x$Ni$_{80-x}$B$_{19}$Si$_1$, Fe$_{20}$Ni$_{60}$B$_{19}$P$_1$, and Fe$_{20}$Ni$_{60}$B$_{20}$ alloys. Inset shows T_{max}/T_C plotted against T_C for the present alloys and for amorphous Fe$_x$Ni$_{80-x}$B$_{20}$ alloys.

concentration interval $10 \leq x \leq 16$, and (ii) the resistivity minima in these alloys occur at temperatures that are lower by an order of magnitude[14] than those at which the peaks in $S(T)$ are observed.

$T_{\max}/T_C$ plotted against T_C is shown in Fig. 2. Our preliminary results on the amorphous $Fe_{20}Ni_{60}B_{19}P_1$ and $Fe_{20}Ni_{60}B_{20}$ alloys have also been included in this figure. It is noticed that $T_{\max}/T_C$ exhibits a linear variation with T_C and increases with increasing Ni content and reaches a value ~ 0.8 at $x = 10$. The latter observation is consistent with the experimental result[17] that the peak in $S(T)$ for crystalline Ni occurs at $0.83T_C$, whereas for Fe it occurs at $0.77T_C$. These observations strongly suggest that the peak in $S(T)$ is of ferromagnetic origin. A relation of the type $T_{\max} = aT_C - bT_C^2$ between $T_{\max}$ and T_C has also been previously observed[18] for the glassy $Fe_xNi_{80-x}B_{20}$ alloy system (see the inset of Fig. 2), but with the values of the coefficients a and b that are very much different from those found in the present work. The foregoing arguments do not completely rule out the possibility of a Kondo scattering contribution to TEP, but instead serve to demonstrate that for $T < T_C$ the magnetic scattering gives a dominant contribution to S. However, a small Kondo- or pseudo-Kondo-scattering contribution to TEP, if present, will be weighted by the ratio[9,10] $\rho^K/\rho^T \sim 10^{-3} - 10^{-2}$ (Nordheim-Gorter rule[19]), where ρ^K is the Kondo or pseudo-Kondo contribution to the total resistivity ρ^T, and hence will not produce any structure in $S(T)$ within our detection limit (0.02 μV/K). It can, therefore, be safely concluded that the observed nonlinear temperature dependence of TEP arises solely from the magnetic scattering. The latter remark restricts the choice of models, in terms of which our experimental results could be discussed, to only three, e.g., the Mott s-d scattering model, Korenblit's theory, and the Kasuya localized-spin model. Before discussing the results in the light of these models, it is worth mentioning that the Ziman model, even with the Faber modification,[20] which, in addition, includes the temperature dependence of the structure factor,[8] cannot account for the pronounced curvature in the S-vs-T plots (Fig. 1).

Within the framework of the Mott s-d scattering model, which takes into account the splitting of the d band into the spin-up $(d\uparrow)$ and spin-down $(d\downarrow)$ d subbands, the TEP is given by[17,21]

$$S(T) = -\frac{\pi^2 k_B^2}{2|e|E_F}T + \frac{\pi^2 k_B^2}{3|e|}\frac{N'_{d\uparrow}(E_F) + N'_{d\downarrow}(E_F)}{N_{d\uparrow}(E_F) + N_{d\downarrow}(E_F)}T$$

$$= S^D(T) + S^M(T), \qquad (1)$$

where $N_{d\uparrow}(E_F)$ and $N_{d\downarrow}(E_F)$ are the density of states in the $d\uparrow$ and $d\downarrow$ subbands at the Fermi level E_F, and

$$N'_{d\uparrow,\downarrow}(E_F) = \frac{dN_{d\uparrow,\downarrow}(E)}{dE}\bigg|_{E=E_F}.$$

From Eq. (1) it is evident that a quantitative interpretation of the data would require a detailed knowledge of the band structure, which is presently lacking for the alloys in question. This limits us to only a qualitative discussion of the results. Since $S^D(T)$, on its own, is too small to ac-

count for the observed values of S even at temperatures as low as 4.2 K, $S^M(T)$ gives a major contribution to S, and hence is mainly responsible for the sizeable composition dependence of TEP and also for its nonlinear variation with temperature. In view of Eq. (1), S can assume large negative values only when both $N'_{d\uparrow}(E_F)$ and $N'_{d\downarrow}(E_F)$ are negative. This implies that both the d subbands should be almost full; a deduction that is consistent with our previous magnetization[16] and magnetoresistance[22] results on the same alloy system as the present one.

While the observed variation of S with T can be qualitatively understood in terms of Eq. (1) when the arguments similar to those of Kolomoets and Vedernikov[17] are followed, the composition dependence of TEP, by comparison, is not so straightforward and as such deserves a brief discussion. As x increases, E_F progressively approaches the top edges of the d subbands since the magnetic moment (or the difference in the population of $d\uparrow$ and $d\downarrow$ subbands) and T_C (or the magnitude of the d-band splitting) both increase.[16] Consequently, with increasing x both $N'_{d\uparrow}(E_F)$ and $N'_{d\downarrow}(E_F)$ (and so also S) take larger negative values, while

$$N_d(E_F) = N_{d\uparrow}(E_F) + N_{d\downarrow}(E_F)$$

increases only slightly, as pointed out by the recent specific-heat measurements (whose results have been collected in Fig. 7 of Ref. 22) on the amorphous $Fe_xNi_{80-x}B_{20}$ alloys. To summarize, the Mott model is capable of explaining the temperature $(T \leq T_C)$ as well as the composition dependence of TEP in the present glasses, but then it predicts a sharp anomaly in $S(T)$ at T_C that we do not observe (Fig. 1).

Recently, Korenblit[11] has proposed a theory, based on the band model of ferromagnetism, which attributes the unusual behavior of TEP in amorphous ferromagnetic alloys to a simultaneous presence of two scattering processes, namely, the inelastic electron-magnon scattering at the host atoms and the elastic scattering of electrons at the solute atoms which depends on the electron-spin direction. The final expression for TEP, obtained through a solution of the Boltzmann equations for spin-up ($\uparrow$) and spin-down
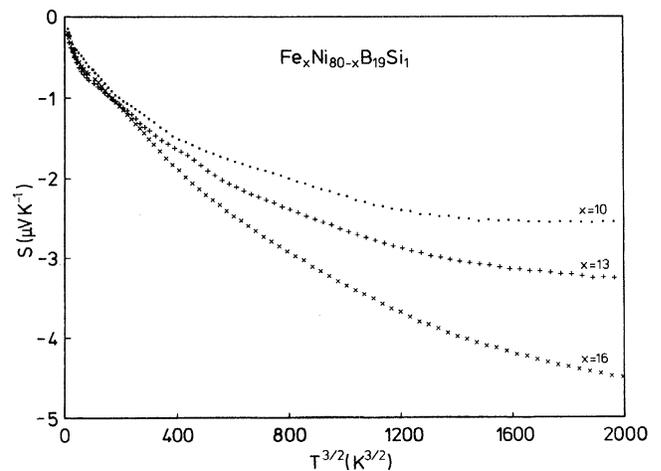
FIG. 3. S-vs-$T^{3/2}$ plots for the alloys with $x = 10$, 13, and 16.

($\downarrow$) electrons in the strongly disordered case (i.e., when the elastic scattering dominates the inelastic one), is given by

$$S(T) = \frac{k_B}{e} \frac{\tau_\uparrow - \tau_\downarrow}{t} , \qquad (2)$$

where τ_σ is the elastic relaxation time of the electrons with spin σ and t is the mean inelastic relaxation time. Although Eq. (2) correctly predicts a nonlinear variation[11] of S with T for glassy ferromagnets, it utterly fails in many respects. (i) At low temperatures, Eq. (2) should take the form (Ref. 11) $S(T) \sim T^{3/2}$, but S-vs-$T^{3/2}$ plots for the present alloys, shown in Fig. 3, clearly demonstrate that even at the lowest temperatures such a relation between S and T does not hold. (ii) Contrary to a large value (~ -2.5 μV/K) of S at T_C, Eq. (2) gives a vanishingly small value for $S(T_C)$ because $\tau_\uparrow \simeq \tau_\downarrow$ at T_C. (iii) This

model is unable to explain the strong concentration dependence of S_{max} (Fig. 1) as evidenced from the fact that we arrive at the value $|S_{max}| \simeq 10$ μV/K for all the alloys under consideration when we set, following Korenblit,[11] $(\tau_\uparrow - \tau_\downarrow)/t \sim \rho_m/\rho_o$ (where ρ_o and ρ_m denote the residual and spin-wave resistivities, respectively) and use the experimental values of ρ_o and ρ_m from Ref. 14.

Making use of a theory, based on the localized-spin model, in which the inelastic scattering [arising from the energy absorption or emission at the ionic (magnetic) levels generated by an internal molecular field] and the elastic spin-disorder scattering are both different for electrons of opposite spin, Kasuya arrives at the following expressions[23] for the TEP for sufficiently low temperatures $(T \ll T_C)$ and for temperatures close to T_C $(T \lesssim T_C)$ using spin-wave and molecular-field approximations, respectively:

$$S(T) = -\frac{\pi^2 k_B^2}{3|e|E_F} T - \left[\frac{\pi^2 k_B^2}{6|e|E_F} \left[\frac{q_0}{k_F} \right]^2 \frac{c(g-1)J_{sd}(0)\langle j_z \rangle}{\epsilon_0} \right] \left[\left[\frac{\Phi_1 \Phi_3}{\Phi_2^2} - 1 \right]^{-1} + 3 \left[\frac{4\pi^2 \Phi_1}{\Phi_3} + 1 \right]^{-1} \right] T$$

$$= S^d(T) + S^m(T) \quad \text{for } T \ll T_C , \qquad (3)$$

with

$$\Phi_n = \int_{x_0}^{x_1} \frac{x^n dx}{(e^x - 1)(1 - e^{-x})}$$

and

$$S(T) = -\frac{\pi^2 k_B^2}{3|e|E_F} T - \frac{2k_B c(g-1)J_{sd}(0)\langle j_z \rangle}{|e|E_F} \frac{1-e^{-x}}{1+e^{-x}}$$

$$= S^d(T) + S^m(T) \quad \text{for } T \lesssim T_C , \qquad (4)$$

where $J_{sd}(0)$ is the Fourier transform of the s-d exchange integral $J_{sd}(\vec{r})$ for $q=0$, $\langle j_z \rangle$ is the mean value of the z component of the total angular momentum j located at each magnetic ion, g is the Landé splitting factor, c is the fraction of magnetic ions, the constant ϵ_0 and the Debye cutoff wave number q_0 are related through the spin-wave dispersion relation $\epsilon = \epsilon_0(q/q_0)^2 = Dq^2$, D is the spin-wave stiffness coefficient,

$$x = 3\frac{T_p}{T} \frac{\langle j_z \rangle}{s(s+1)} ,$$

T_p is the paramagnetic Curie temperature, s is the localized spin, $x_1 = \epsilon_0/k_B T$,

$$x_0 = x_1 \frac{c(g-1)J_{sd}(0)\langle j_z \rangle k_F}{q_0 E_F} ,$$

and k_F is the Fermi wave vector. Equations (3) and (4) clearly show that, besides a normal diffusion part $S^d(T) = AT$ (common to both low- and high-temperature expressions), which is characteristic of the isotropic scattering, TEP contains a magnetic part $S^m(T)$, which characterizes the s-d interaction, and according to this

theory, is mainly responsible for the anomalous temperature dependence of TEP in magnetic systems. We have attempted a quantitative fit of the experimental data to Eqs. (3) and (4) because our earlier resistivity[14] and magnetization[15,24] data taken on a number of amorphous ferromagnetic alloys, including those presently under consideration, clearly demonstrate that the Heisenberg approximation (and hence Kasuya's theory) is better suited to amorphous ferromagnets than to the crystalline ones [for which the theory leading to Eqs. (3) and (4) was originally proposed]. Assuming the orbital momentum to be completely quenched for the $3d$ electrons, we set $g \simeq 2$ and replace $\langle j_z \rangle$ by $\bar{s}_z = [M_s(T)/M_s(0)]s$, where M_s is the spontaneous magnetization. The following fitting procedure has been adopted. At first, the experimental data taken in the temperature range $0.6T_C \leq T \leq T_C$ have been fitted to the molecular-field expression [Eq. (4)] using a least-squares computer program which treats the coefficient A and the ratio $J_{sd}(0)/E_F$ as free parameters. Setting $s=1$ and $c=0.8$, and using the T_p values estimated from the resistivity measurements[14] and our previously reported $M_s(T)/M_s(0)$ data[15,24] on glassy $Fe_xNi_{80-x}B_{19}Si_1$ alloys, we obtain the values of A and $J_{sd}(0)/E_F$ given in Table I. A simple cross check for the values of A obtained by this fitting procedure is provided by the fact that S^m in Eq. (4) vanishes at T_C so that A is nothing but $S(T_C)/T_C$. A glance at Table I reveals that (i) the corresponding values of A and $S(T_C)/T_C$ are in excellent agreement with each other, (ii) the E_F values (given in Table I within the parentheses) derived from these A values underestimate the expected magnitude of E_F by at least a factor of 3, and (iii) the ratio $J_{sd}(0)/E_F$ increases linearly with x and possesses values that compare well with those observed for crystalline Fe-Ni alloys[25] and also with the theoretical es-

TABLE I. Parameter values, obtained by fitting the TEP data to Eqs. (3)—(5) of the text, and our previously reported magnetic parameter values used for fits, for the amorphous $Fe_xNi_{80-x}B_{19}Si_1$ alloys.

x (at. %)	D (meV Å²)	T_p (K)	T_C (K)	$S(T_C)/T_C$ (10⁻² μV/K²)	A (10⁻² μV/K²)	$J_{sd}(0)/E_F$ (10⁻²)	k_F (Å⁻¹)	a (10⁻² μV/K²)	b (μV/K)	Fit range (T/T_C)
10	53±2	340±15	186.5±0.3	-1.3±0.1	-1.3±0.1 (1.9 eV)	1.2±0.02	0.15±0.02	1.0±0.2	-4.3±0.1	1.02-1.61
13	66±2	389±15	268.5±0.2	-1.0±0.1	-1.0±0.1 (2.4 eV)	2.2±0.02	0.17±0.02	1.1±0.2	-5.5±0.1	0.82-1.12
16	78±2	393±15	338.0±1.0	-1.2±0.1[a]	-1.2±0.1 (2.0 eV)	3.0±0.03	0.20±0.02	0.6±0.2	-6.2±0.1	0.65-0.89

[a]Obtained from linear extrapolation of the $S(T)$ data to T_C.

timates (10^{-2}—10^{-1}) for pure Heisenberg ferromagnets obtained within the free-electron approximation.[23] While the agreement between the A and $S(T_C)/T_C$ values, and that between the experimental and theoretical values of $J_{sd}(0)/E_F$, is in a way implicitly assumed in Kasuya's theory, observation (ii) possibly points out the well-known fact that the molecular-field approximation is not suited for temperatures in the immediate vicinity of T_C.

Now that the term $S^d(T)$ is the same for both low- and high-temperature expressions (3) and (4), $S^d(T)$ is subtracted from the observed $S(T)$ data so as to arrive at the experimentally determined variation of the magnetic contribution to TEP, $S^m(T)$, with temperature. The values of S^m at different temperatures obtained in this way are plotted against the reduced temperature T/T_C and shown in Fig. 4. One notices from this figure that it is $|S^m(T)|$ and not $|S(T)|$, as contended by Babic et al.,[26] that goes through a maximum at $T_{max} \simeq 0.5T_C$, and with increasing Fe concentration the peak value $|S^m_{max}|$ increases and the peak position T_{max} shifts slightly towards low temperatures. The next step is to see if Kasuya's model can correctly predict the temperature dependence of S^m in the spin-wave region. In order to accomplish this, the $S^m(T)$ data for $T \leq 0.4T_C$ have been fitted to Eq. (3) using the magnetic data from Ref. 24, the $J_{sd}(0)/E_F$ values[27] obtained from the high-temperature fits (Table I), and the composition-independent value[27] of ϵ_0 (equal to $Dq_0^2) \sim 170$ meV estimated from the published sound-velocity data on similar amorphous systems,[28] while treating k_F in Eq. (3) as an adjustable parameter in order to minimize the deviations of the calculated TEP values from the measured ones. The definite integrals Φ_n appearing in Eq. (3) have been computed numerically by employing a variant of the Romberg integration method. The theoretical variation of S^m with temperature that the expressions (3) and (4) predict with the choice of the parameters given in Table I is represented in Fig. 4 by the solid curves through the data points. It is noticed that the theoretical curves reproduce the experimental variation
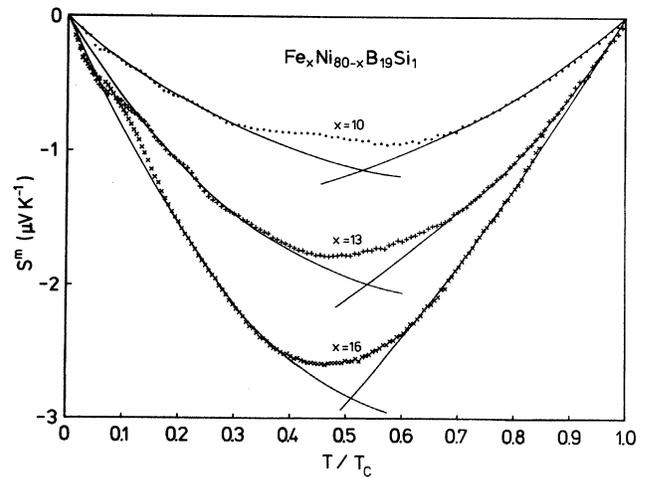
FIG. 4. Magnetic part of the TEP, S^m, as a function of the reduced temperature T/T_C. The solid curves through the data points represent the theoretical variation predicted by Eqs. (3) and (4) with the choice of the parameters given in Table I.

reasonably well. Furthermore, the magnitude of the k_F values, derived from the low-temperature fit, is comparable to that obtained by Maeda and Somura[25] for crystalline Fe-Ni alloys, but far too small to justify the use of free-electron approximation for $3d$ transition-metal alloys. To reconcile our results with the theory, we also attempted to fit the low-temperature $S(T)$ data to Eq. (3) by keeping the value of k_F fixed at 1.5Å^{-1} and allowing the ratio $J_{sd}(0)/E_F$ to vary with temperature. In direct contradiction with the expected variation of $J_{sd}(0)/E_F$ [$J_{sd}(0)/E_F$ should decrease with increasing T since $J_{sd}(0)$ decreases more rapidly than does E_F], this ratio increases with increasing temperature, such that the high-temperature value is larger, by a factor ~ 3, than the low-temperature value, which itself is roughly 1 order of magnitude higher than the values given by the high-temperature fit (Table I). The above inconsistencies beween the theory and experiment could possibly be traced to either the total neglect of electron-phonon scattering[29] or the approximations made while integrating the relaxation terms in the kinetic equation, or both, in the Kasuya model. Some of these limitations have been subsequently removed by Abel'skii and Irkhin[30] through the inclusion of electron-phonon scattering and more accurate calculations in the high-temperature limit $(T \lesssim T_C)$, again, by using a mean-field approximation. However, their final expression for TEP involves a number of unknown parameters and is so complicated that a direct comparison with the experiment is not possible without a cumbersome fitting procedure.

Finally, we return to the behavior of TEP in the critical region and for temperatures well above T_C. The important features of the results in this temperature range are (i) the approximately linear dependence of TEP,

$$S(T)=aT+b , \qquad (5)$$

on temperature for all the alloys above T_x $(T_x \simeq 190, 205, 220$ K for $x = 10$, 13, and 16, respectively), (ii) the coefficient a is positive and roughly independent of Fe concentration x, whereas b depends on x, and (iii) no anomaly in

S is observed at T_C in spite of the fact that the resistivity (ρ) shows a sharp anomaly[14] at T_C. In contrast, a sharp anomaly at T_C in both ρ and S has been observed in crystalline ferromagnets. In this sense, magnetic glasses exhibit a unique behavior of $S(T)$ in the critical region. None of the above-mentioned transport models (including the Mott and Kasuya models which correctly predict the variation of TEP with temperature for $T \lesssim T_C$) can account for features (i), (ii), and (iii). No plausible explanation can be given for these observations at present.

IV. SUMMARY AND CONCLUSIONS

The results of the TEP measurements on amorphous Fe$_x$Ni$_{80-x}$B$_{19}$Si$_1$ $(x = 10$, 13, and 16) alloys in the temperature range 4.2—300 K, when compared with the predictions of the available transport theories, reveal that none of these theories is complete in itself to account for the diversity of these results. For example, despite the fact that Mott s-d scattering and Kasuya models, of all the existing transport theories, are capable of reproducing the observed temperature and composition dependence of TEP for temperatures below T_C, both fail to correctly predict the behavior of TEP in the critical region and for $T > T_C$. Contrary to the popular belief that the nonlinear TEP in magnetic glasses arises from the Kondo effect, our results clearly demonstrate that for $T \leq T_C$ the functional dependence of TEP on temperature is mainly decided by the magnetic scattering from the moment bearing atoms. Different contributions to TEP arising from different scattering mechanisms in such glasses have been clearly identified.

ACKNOWLEDGMENTS

[1]S. R. Nagel, Phys. Rev. B 16, 1694 (1977); K. Fröböse and J. Jäckle, J. Phys. F 7, 2331 (1977); P. J. Cote and L. V. Meisel, Phys. Rev. Lett. 39, 102 (1977).

[2]N. F. Mott, Philos. Mag. 26, 1249 (1972).

[3]R. W. Cochrane, R. Harris, J. O. Strom-Olson, and M. J. Zuckermann, Phys. Rev. Lett. 35, 676 (1975); C. C. Tsuei, Solid State Commun. 27, 691 (1978).

[4]G. S. Grest and S. R. Nagel, Phys. Rev. B 19, 3571 (1979).

[5]S. R. Nagel, Phys. Rev. Lett. 41, 990 (1978).

[6]B. L. Gallagher, J. Phys. F 11, L207 (1981); B. L. Gallagher and D. Greig, ibid. 12, 1721 (1982).

[7]T. Matsuda and U. Mizutani, J. Phys. F 12, 1877 (1982); M. N. Baibich, W. B. Muir, Z. Altounian, and Tu Guo-Hua, Phys. Rev. B 26, 2963 (1982); 27, 619 (1983).

[8]R. Harris and B. G. Mulimani, Phys. Rev. B 27, 1382 (1983).

[9]M. N. Baibich, W. B. Muir, G. Belanger, J. Destry, H. S. Elzinga, and P. A. Schroeder, Phys. Lett. 73A, 328 (1979); B. Bhanu Prasad and A. K. Bhatnagar, Solid State Commun. 41, 381 (1982).

[10]S. Basak, S. R. Nagel, and B. C. Giessen, Phys. Rev. B 21,

4049 (1980); J. P. Carini, S. Basak, and S. R. Nagel, J. Phys. (Paris) Colloq. 41, C8-463 (1980).

[11]I. Y. Korenblit, J. Phys. F 12, 1259 (1982).

[12]R. B. Roberts, Philos. Mag. 36, 91 (1977).

[13]We believe that the small bump in the TEP data near 20 K for the alloys with $x = 13$ and 16 results from the phonon-drag peak of indium since the repeated measurements on several "virgin" samples taken from the same alloy batch have shown that this bump becomes more and more pronounced as the diameter of the thermocouple (hot) junction increases compared to that of the thermocouple wires. The upward trend of the data for temperatures close to 300 K is an artifact of the fitting procedure (see Sec. II).

[14]G. Böhnke and M. Rosenberg, J. Phys. (Paris) Colloq. 41, C8-481 (1980).

[15]S. N. Kaul and M. Rosenberg, Philos. Mag. 44, 357 (1981); Solid State Commun. 41, 857 (1982).

[16]S. N. Kaul, IEEE Trans. Magn. MAG-17, 1208 (1981).

[17]N. V. Kolomoets and M. V. Vedernikov, Fiz. Tverd. Tela (Leningrad) 3, 2735 (1961) [Sov. Phys.—Solid State 3, 1996

(1962)].

[18]B. Pivac and E. Babic, in *Recent Developments in Condensed Matter Physics*, edited by J. T. Devreese, L. F. Lemmens, V. E. van Doren, and J. van Royen (Plenum, New York, 1981), Vol. 2, p. 273.

[19]R. D. Barnard, *Thermoelectricity in Metals and Alloys* (Wiley, New York, 1972).

[20]T. E. Faber and J. M. Ziman, Philos. Mag. 11, 153 (1965).

[21]F. J. Blatt, P. A. Schroeder, C. L. Foiles, and D. Greig, *Thermo-electric Power of Metals* (Plenum, New York, 1976), p. 161.

[22]S. N. Kaul and M. Rosenberg, Phys. Rev. B 27, 5698 (1983).

[23]T. Kasuya, Progr. Theor. Phys. 22, 227 (1959).

[24]S. N. Kaul, Phys. Rev. B 27, 5761 (1983); 24, 6550 (1981).

[25]T. Maeda and T. Somura, J. Phys. Soc. Jpn. 44, 148 (1978).

[26]E. Babic, Z. Marohnic, M. Ocko, A. Hamzic, K. Saub, and B. Pivac, J. Magn. Magn. Mater. 15-18, 934 (1980).

[27]The choice of ϵ_0 has no significant influence on the values of the integrals Φ_n, since within the temperature range of the present interest the upper limit x_1, to a good approximation, can be set equal to infinity. In Eqs. (3) and (4), we set $J_{sd}^{Ni}(0)=J_{sd}^{Fe}(0)=J_{sd}(0)$ since these quantities are expected [see, e.g., E. P. Wohlfarth, in *Ferromagnetic Materials*, edited by E. P. Wohlfarth (North-Holland, New York, 1980), Vol. 1, p. 1] to be of similar magnitude.

[28]A. P. Malozemoff, P. H. Chang, and M. Grimsditch, J. Appl. Phys. 50, 5896 (1979).

[29]The phonon-drag contribution to TEP is expected to be negligible in our glassy systems since the scattering of phonons by electrons will be much weaker than that due to the disordered potential. Again, in magnetic glasses, the electron-phonon mass-enhancement effect will be destroyed because of the suppression of electron-phonon coupling by magnetic scattering.

[30]S. S. Abel'skii and Y. P. Irkhin, Fiz. Tverd. Tela (Leningrad) 13, 2431 (1971) [Sov. Phys.—Solid State 13, 2035 (1972)].