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# Detailed magnetization study of an amorphous ferromagnet

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Exhaustive magnetization measurements have been carried out on the amorphous ferromagnet $Fe_{40}Ni_{40}P_{14}B_6$ in the temperature range 4.2 to 700 K in fields up to 16 kOe. Magnetization at low temperatures is found to decrease in accordance with the predictions of an isotropic nearest-neighbor Heisenberg model. It is observed that in this alloy the rms range of exchange interaction equals roughly the mean nearest-neighbor distance. A theoretical interpretation has been provided for the observation that the spin-wave stiffness constant $D$ is smaller in amorphous than in crystalline ferromagnets. Magnetization data taken in the critical region, besides yielding the values for the Curie temperature and critical exponents as $T_C = 519.9 \pm 0.2$ K., $\beta = 0.38 \pm 0.01$, $\gamma = 1.31 \pm 0.02$, and $\delta = 4.46 \pm 0.04$, satisfy the magnetic equation of state characteristic of a second-order phase transition. The observed exponent values not only obey the static scaling relation but also present a reasonable agreement with the theoretical values deduced from a three-dimensional Heisenberg model. They do, however, exhibit a slight but systematic shift towards the molecular-field values. This observation has been shown to imply an increased effective range of the exchange interaction in the critical region. Long-range forces are found to have a negligible influence on the critical fluctuations of magnetization. Critical amplitude values suggest that the elementary moments involved in the magnetic order-disorder transition are those forming a part of the giant superparamagnetic moments which are found to exist for temperatures well above $T_C$. Above 575 K, the susceptibility obeys the Curie-Weiss law. A careful analysis of the susceptibility data reveals that the spin polarization of conduction electrons gives a negligible contribution to the distribution of the magnetic moment and that the moments on Ni and Fe atoms remain localized even in the paramagnetic state. No evidence has been found for the indirect exchange between spins on magnetic atoms mediated by conduction-electron spin, as conjectured by Kaneyoshi in amorphous ferromagnets, in the present glassy alloy.

## I. INTRODUCTION

Soaring interest in amorphous magnetism[1,2] during recent years has stemmed partly from the envisaged, and to some extent already realized, technical applications of amorphous ferromagnetic alloys and partly from the light these materials are expected to shed on the effects of structural disorder on the solid-state properties in general and magnetic properties, in particular. Amongst a wide variety of magnetic metallic glasses prepared by the rapid-quenching technique, perhaps the most thoroughly studied,[3-12] as far as the magnetic properties are concerned, is the glassy ferromagnet $Fe_{40}Ni_{40}P_{14}B_6$ (Metglas®2826, manufactured by the Allied Chemical Corporation, New Jersey). In spite of such intense efforts, a number of points (brought out in the following text) concerning the low-temperature spin excitations and the critical phenomena near the magnetic phase transition in this alloy still remain obscure.

Early inelastic neutron scattering and magnetization experiments reported on amorphous $Co_{80}P_{20}$,[13,14] $Fe_{75}P_{15}C_{10}$,[15,16] and $(Fe_{0.93}Mo_{0.07})_{80}P_{10}B_{10}$ (Refs. 17 and 18) alloys have demonstrated that (i) at long wavelengths, such glasses exhibit well-defined spin-wave excitations which satisfy a normal ferromagnetic dispersion relation[19]

$$\hbar\omega(q) = \Delta_g + Dq^2 + Eq^4 + \cdots \quad , \tag{1}$$

with an effective energy gap $\Delta_g << Dq^2$ arising from the dipole-dipole interactions,[18,19] (ii) at low temperatures, magnetization decreases in accordance with the Heisenberg-model prediction

$$\frac{\Delta M(T)}{M(0)} = \frac{M(0) - M(T)}{M(0)} = BT^{3/2} + CT^{5/2} + \cdots \quad , \tag{2}$$

and (iii) neutron scattering experiments yield a $D$ value which is consistently larger (by about 1.5 times) than that deduced from magnetization measurements. Several arguments[14,15,18,20,21] put forward to justify such a discrepancy between the values of $D$ as determined from magnetization and neutron-diffraction studies can be summarized as follows: (a)

The neutron-diffraction experiments give a *direct* measure of the spin-wave stiffness constant $D$ and establish the existence of the long-wavelength spin waves. But due to the lack of periodicity in noncrystalline materials, such measurements are limited to low transferred momenta (typically $q \leq 0.25$ Å$^{-1}$) and hence to small scattering angles.[14,15,20] At higher momentum transfers, the spin waves appear to be *diffused*. By contrast, magnetization measurements directly measure $B$, the coefficient of the $T^{3/2}$ term in Eq. (2), which is the "thermal average" of the total contribution arising from spin waves of *all* wavelengths. (b) Besides the conventional spin waves, there exist *additional excitations*,[18] *invisible to neutrons*, which also decrease magnetization according to the Bloch $T^{3/2}$ law and are more or less *localized* in nature. (c) Rivier and Continentino[21] contend that in amorphous magnets with negligible local anisotropy (i.e., in a ground state of noncollinear local magnetic moments) the *diffusive modes* ("diffusions") generated by longitudinal spin fluctuations contribute, besides the spin waves, to the $T^{3/2}$ decrease of the magnetization. But, unlike conventional spin waves, they show up as a broad central peak in the inelastic neutron scattering intensity.

Recent systematic spin-wave excitation, Mössbauer, and low-temperature magnetization studies on amorphous $(Fe_xNi_{1-x})_{75}P_{16}B_6Al_3$ alloys,[22-24] besides confirming the previous observations (i) and (ii), reveal that the values of $D$ obtained from magnetization and Mössbauer measurements, though in good agreement with one another for all the compositions $x$, are either very close to (for $x \geq 0.5$) or greater (by a factor $\sim 1.5$) than (for $x < 0.4$) the corresponding values measured by the neutron scattering experiments. In view of the latter observation, the above arguments (a)–(c) concerning the discrepancy between $D$ values as determined from magnetization and neutron-diffraction studies cannot be generalized. A close agreement between the results of Mössbauer and magnetization measurements has also been found in the case of amorphous $Fe_{75}P_{15}C_{10}$ and $Fe_{1-x}B_x$ alloys.[16,25-27] Contrary to the observations made on the alloys just mentioned, magnetization data[9,10] taken on Metglas® 2826 give a spin-wave stiffness constant value of $D \cong 75$ meV Å$^2$ whereas both neutron-diffraction[28] and Mössbauer[7,8,29] measurements yield identical results, viz., $D \cong 100$ meV Å$^2$. While the values of $D$ deduced from Mössbauer and magnetization studies are fairly close to one another for all the metallic glasses studied so far, there seems to be no *a priori* reason to believe that they should be widely different only in the case of Metglas® 2826.

Chien and Hasegawa[7,8] have performed Mössbauer measurements on Metglas® 2826 to study its critical magnetic behavior. They find that the temperature dependence of the hyperfine field in the vicinity of the Curie temperature can be fitted by a power law

$$H_{\text{eff}}(T)/H_{\text{eff}}(0) = D'[1-(T/T_C)]^{\beta} \ , \tag{3}$$

with the critical parameters $\beta = 0.32 \pm 0.05$ and $D' = 1.03 \pm 0.03$. In sharp contrast with the above value for the critical exponent $\beta$, bulk magnetization measurements[30-40] on a large number of amorphous magnetic alloys give a $\beta$ value which is close to 0.4 (a value well outside the error limit quoted above for $\beta$). But, because no detailed magnetic data on Metglas® 2826 in the critical region have been reported so far, a direct comparison between the $\beta$ values obtained from Mössbauer and magnetic studies on the same alloy is not possible at present.

In an attempt to tackle the above-mentioned problems, we have carried out an exhaustive magnetic measurement on amorphous $Fe_{40}Ni_{40}P_{14}B_6$ alloy in a temperature range 4.2 to 700 K in magnetic fields up to 16 kOe. The results indicate that the low-temperature magnetization decreases according to Eq. (2) up to about $0.5T_C$ with the values for the coefficients $B$ and $C$ in excellent agreement with those obtained from Mössbauer studies.[7,8,29] The observed $B$ value is found to be very close to that predicted by isotropic Heisenberg nearest-neighbor exchange-interaction model. Furthermore, the range of the exchange interaction, calculated from the experimental values of $B$ and $C$, equals just the nearest-neighbor transition-metal–transition-metal distance. A careful analysis of the experimental data in the critical region gives reliable values for the critical exponents, critical amplitudes, and the Curie temperature. In the temperature range $2.9 \times 10^{-2} \leq |\epsilon| \leq 4.2 \times 10^{-1}$ $[|\epsilon| = |(T-T_C)/T_C|]$, our measurements, like previous Mössbauer measurements,[7,8] demonstrate that Eq. (3) is satisfied with the values for the spontaneous magnetization critical exponent $\beta$ and the corresponding critical amplitude $D'$ as $0.32 \pm 0.02$ and $0.95 \pm 0.01$, respectively. However, for temperatures very close to the Curie temperature, i.e., from $|\epsilon| = 2.9 \times 10^{-4}$ to $2.2 \times 10^{-2}$ (such a temperature region being inaccessible to the Mössbauer spectroscopic method because of poor spectral resolution for $|\epsilon| < 10^{-2}$ due to broad inherent linewidths resulting from hyperfine-field distribution) $\beta$ assumes a value of $0.38 \pm 0.02$ which conforms reasonably well with the $\beta$ values reported for other amorphous alloys. As observed in other amorphous ferromagnets,[30-40] the critical exponents for the present alloy also have values close to those expected from a three-dimensional Heisenberg model, exhibit the same anomalous behavior,[36-40] and obey the static scaling relation. The experimentally observed critical exponent values suggest an increased effective range of the Heisenberg exchange interaction in the critical region. In addition, it has been inferred from the critical amplitude values that in Metglas® 2826 the elementary moments involved in the magnetic order-disorder transition are not those of individual moments as in crystalline Ni but those forming a part of the giant superparamagnetic moments which are found to exist for temperatures well above the Curie temperature. Magnetization data are observed to satisfy the magnetic equation of state characteristic of a second-order magnetic phase transition. Above 575 K, magnetic susceptibility obeys the Curie-Weiss law with the values of paramagnetic Curie temperature and Curie constant in close agreement with their values found previously by Hasegawa and O'Handley.[3] From the high-temperature behavior of the alloy, it is concluded that even in the paramagnetic state the moments on Fe and Ni atoms are localized.

## II. EXPERIMENTAL DETAILS

Ribbon of the amorphous $Fe_{40}Ni_{40}P_{14}B_6$ alloy (prepared by the rapid-quenching technique and commercially known as Metglas® 2826), having a thickness $\sim 50$ $\mu$m and width $\sim 2$ mm, was procured from Allied Chemical Corporation, New Jersey. Using the Faraday method, magnetization versus magnetic-field isotherms for this glassy material in "as-received" condition were taken at approximately 10 K intervals from 4.2 to 300 K in fields up to 16 kOe. The details about the sample-holder design and the procedure are given elsewhere.[37,41] The sample temperature was monitored by precalibrated carbon- and platinum-resistance thermometers in the temperature ranges 4.2–50 and 50–300 K, respectively, and was held constant to within 0.1 K by controlling either the input to a resistance heater or the helium flow or both in a continuous-flow cryostat. Above 300 K, magnetization was measured in fields up to 16 kOe by a Prinston Applied Research (PAR) vibrating sample magnetometer (model 155, having a resolution of about $10^{-4}$ emu) in conjunction with a high-temperature oven assembly. The sample was heated by means of a heater (situated in the annular space, which was evacuated to a vacuum of $10^{-5}$ torr, between the inner and outer walls of the oven) while its temperature was measured by a Chromel-Alumel thermocouple placed adjacent to it in the sample chamber. A good thermal contact between the sample and thermocouple was ensured by maintaining a small positive pressure of helium-exchange gas in the sample chamber.

Having located the Curie temperature at $T_C = 520$ K from the sharp kink observed in the temperature dependence of low-field (15 Oe) magnetization, magnetization as a function of temperature was measured at constant external fields 50 Oe $\leq H_{\text{ext}} \leq$ 16 kOe in the temperature interval $(T_C - 50) \leq T \leq (T_C + 50)$ K with temperature increasing at a rate of 30 K per hour. In order to achieve sufficient accuracy in the measurement near the Curie temperature, several pieces of Metglas® 2826 ribbon (each roughly 8 mm in length) with a total weight of about 12 mg were stacked side by side in the high-temperature sample holder with the external magnetic field in the plane of the ribbon pieces so as to minimize the demagnetizing field effects. Magnetization measurements in an applied field of 10 kOe were continued to 700 K to find out the magnetic behavior of this alloy in the paramagnetic region.

## III. RESULTS AND DISCUSSION

In order to achieve a clear and coherent presentation of the present findings, the text describing the analysis and discussion of the experimental data has been divided into the following subsections.

### A. Temperature dependence of spontaneous magnetization

The assumption that spontaneous magnetization has a negligible variation with temperature between absolute zero and 4.2 K leads to a value for spontaneous magnetization at absolute zero $\sigma_s(0)$ of 100.27 emu/g which corresponds to $1.14\mu_B$ per transition-metal atom (TMA) or $M_s(0) = 754$ G [density for this alloy is taken to be 7.52 (Ref. 42)]. It is interesting to note that Hasegawa and O'Handley[3] arrived at the same $\sigma_s(0)$ value as above from their magnetic studies on Metglas® 2826. As is well known, a meaningful reduced magnetization $M_s(T)/M_s(0)$ versus reduced temperature $T/T_C$ curve can be obtained only when the Curie temperature $T_C$ is determined unambiguously. A method, whose details are given in Sec. III C has been adopted for this purpose and the value of $T_C$ was found to be $519.9 \pm 0.2$ K, which is again in excellent agreement with the $T_C$ values determined for the same alloy in Refs. 3 and 11. Having found values of $M_s(0)$ and $T_C$, the values of spontaneous magnetization at different temperatures have been obtained in the temperature range $4.2 \leq T \leq 440$ K by simply extrapolating the high-field saturated portion of the magnetization versus field isotherms to zero field (the external field being already corrected for the demagnetizing field) and from $0.85T_C$ (442 K) to $T_C$ by a *parabolic extrapolation*[11,37–39] of the Arrott-Kouvel plots (i.e., $M^2$ vs $H/M$ isotherms) to $H/M = 0$.

The temperature dependence of reduced spontaneous magnetization for amorphous $Fe_{40}Ni_{40}P_{14}B_6$ is shown in Fig. 1. Theoretical predictions based on molecular-field theory for $S = \frac{1}{2}$ and $S = 1$, which describe reasonably well[43] the temperature dependence of reduced magnetization in crystalline Ni and Fe, respectively, have been included in this figure for comparison. It becomes immediately obvious that the $M_s(T)/M_s(0)$ vs $T/T_C$ curve (henceforth referred to as universal magnetization curve) for the

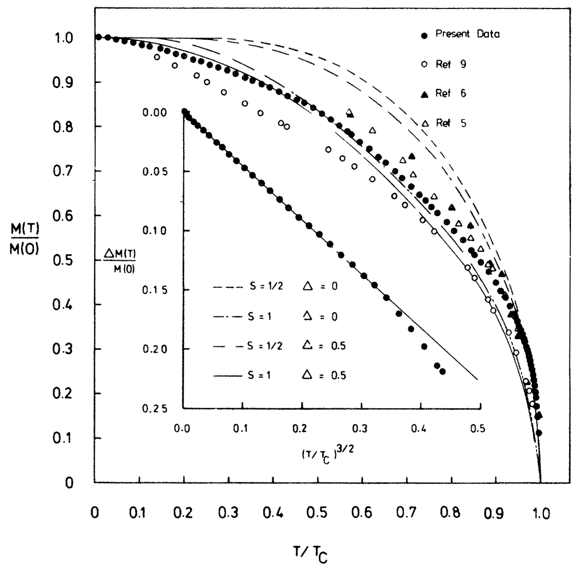

FIG. 1. Reduced spontaneous magnetization vs reduced-temperature curve for amorphous ferromagnet $Fe_{40}Ni_{40}P_{14}B_6$. Various curves through the data points are results obtained from Eq. (5). Insert shows the fractional decrease in spontaneous magnetization due to increasing temperature plotted against $(T/T_C)^{3/2}$.

present alloy falls well below that for crystalline Ni or Fe. This seems to be a feature which all the amorphous ferromagnetic alloys studies so far[13,16,17,27] have in common. Two general, but basically opposite, theoretical approaches have been proposed to justify the diminished curvature of the universal magnetization curve in amorphous ferromagnets. In the first approach, a fixed unique value is assigned to the exchange coupling constant between the spins on magnetic atoms and the amorphous nature of the alloy is taken into account by assuming a random distribution of local anisotropy field.[44,45] In the second approach, the structural fluctuations in the amorphous materials are assumed to manifest themselves in a distribution of exchange integrals.[46–53] Despite the fact that both the above-mentioned approaches demonstrate that the universal magnetization curve for an amorphous ferromagnetic material must characteristically fall below that for the crystalline counterpart, the second approach is usually preferred to the first one for comparison with the experimental results for two reasons. First, contrary to the observed Mössbauer spectra,[4,7,8,16,29] the theories under the first category predict a flat (structureless) Mössbauer spectrum[44,54] for amorphous ferromagnets below the Curie temperature. Second, the prediction based on the theory of Gubernatis *et al.*[45] that an identical value for the coefficient $B$ of the $T^{3/2}$ term in Eq. (2) should be observed for both crystalline and amorphous ferromagnets is in direct contradiction with the experimental findings (see Sec. III B). Of the various theories proposed under the second category, only the molecular-field approach of Handrich[46] provides results in a simple analytical form. Starting from the Heisenberg Hamiltonian

$$H = -\sum_{i,j} J_{ij} \vec{S}_i \cdot \vec{S}_j \tag{4}$$

and assuming $J_{ij} = \langle J \rangle + \Delta J_{ij}$ (where $\langle J \rangle$ denotes the spatially averaged value of the exchange integral between nearest-neighbor spins and $\Delta J_{ij}$ is the fluctuating part of the exchange integral arising from the structural fluctuations in amorphous ferromagnets), Handrich[46] arrives at the following expression for the reduced spontaneous magnetization in the molecular-field approximation

$$M_s(T)/M_s(0) = \tfrac{1}{2}\{B_s[(1+\Delta)x] + B_s[(1-\Delta)x]\} \; , \tag{5}$$

with

$$x = [3S/(S+1)](T_C/T)[M_s(T)/M_s(0)] \; ,$$

where $B_s$ is the Brillouin function and $0 < \Delta < 1$ is a measure of the degree of disorder and is defined as the root mean square of deviation from an average exchange integral between two nearest-neighbor spins:

$$\Delta^2 = \langle (\Delta J)^2 \rangle / \langle J \rangle^2 \; . \tag{6}$$

Equation (5) clearly shows that (i) by setting $\Delta = 0$, the well-known molecular-field expression for crystalline ferromagnets, i.e., $M_s(T)/M_s(0) = B_s(x)$ is recovered and (ii) a progressive increase in the value of $\Delta$ results in increased depression of spontaneous magnetization over the entire temperature range for $T \leq T_C$. We have attempted to fit the observed temperature dependence of spontaneous magnetization for Metglas® 2826 in terms of the relation (5) by using various values of $\Delta$ both for $S = \frac{1}{2}$ and 1 with the result that the theoretical curves for $\Delta = 0.3$, $S = 1$ and $\Delta = 0.5$, $S = \frac{1}{2}$ give a reasonable overall fit to the experimental data. Since the theoretical curves for both $\Delta = 0.3$, $S = 1$ and $\Delta = 0.5$, $S = \frac{1}{2}$ exhibit roughly the same variation, only the theoretical curve corresponding to $\Delta = 0.5$, $S = \frac{1}{2}$ is included for the sake of clarity in Fig. 1. Based on the Mössbauer studies on Metglas® 2826, Chien and Hasegawa[8] too have arrived at the same result.

Although Eq. (5) is capable of yielding a lower universal magnetization curve for glassy ferromagnets, it is still far from giving a quantitative agreement with the experimental results. As is evident from Fig. 1, even for the most reasonable values of $\Delta = 0.5$ for $S = \frac{1}{2}$ and $\Delta = 0.3$ for $S = 1$ the data points lie above the theoretical curve for temperatures close to $T_C$ and below for low enough temperatures. Similar disagreement between the theoretically predicted ($\Delta = 0$) and experimentally observed variation for spontaneous magnetization with temperature has also been found in the case of crystalline ferromagnets. Such a discrepancy between the theory and experiment arises basically from the inadequacy of molecular-field approximation for temperatures either in the low-temperature region, where spin-wave description is more appropriate, or in close vicinity of the Curie temperature where, as we will see in Sec.III C, the Heisenberg model gives better quantitative agreement with the observed temperature dependence of magnetization. At this stage, it is imperative to mention that Vincze *et al.*,[55] based on their Mössbauer studies on $(Fe_{1-x}Ni_x)_{75}B_{25}$ alloys in both amorphous as well as crystalline state, find no substantial difference between the reduced hyperfine-field (hf) versus reduced-temperature curves of the corresponding amorphous and crystalline alloys and hence conclude that structural disorder has a minor effect on the shape of hyperfine-field curve whereas the high metalloid content is responsible for the rather fast decrease of hf with temperature in these materials. Contrary to the above observation, Tsuei and Lilienthal[16] find that the universal magnetization curve for amorphous $Fe_{75}P_{15}C_{10}$ alloy (this alloy crystallizes at ~400 °C into a predominant $Fe_3P$ phase with $Fe_3C$ and $\alpha$-Fe as other minor phases) lies well below that for crystalline $Fe_3P$ and the difference between them is quite significant. Therefore, the basic mechanism responsible for the faster decrease of spontaneous magnetization with temperature in amorphous ferromagnets compared with that in their crystalline counterparts is still controversial. Nevertheless, a distribution of the exchange interaction, as inferred from Mössbauer studies, remains a useful concept so far as the magnetic and electrical properties of amorphous ferromagnets are concerned (for details see Ref. 16).

### B. Low-temperature magnetization and spin-wave excitations

By now ample experimental evidence exists in the literature to demonstrate that the decrease of magnetization with increasing temperature at low temperatures not only in crystalline ferromagnetic materials[19,56] but also in amorphous ferromagnets[13,16–18,22–27,29] is adequately described by the theoretical prediction based on the Heisenberg model, Eq. (2). The results of the magnetization measurements performed on Metglas® 2826 are plotted again $(T/T_C)^{3/2}$ in the insert of Fig. 1 [note that the subscript $s$ in $M_s(T)/M_s(0)$ has been dropped both in Eq. (2) and in this figure for convenience]. It is clearly seen from this figure that Bloch's $T^{3/2}$ law holds for temperatures up to 255 K ($0.49T_C$), in agreement with the previous magnetic[9,10] and Mössbauer[4,7,8] studies on the same glassy alloy, with the coefficient $B = 38 \pm 2 \times 10^{-6}$ K$^{-3/2}$. However, a much better fit to the observed data in an extended temperature range $4.2 \leq T \leq 295$ K could be obtained by using both the terms in Eq. (2) and plotting $[\Delta M(T)/M(0)]T^{-3/2}$ against $T$. The intercept and slope of the least-squares-fit straight line through the data points gave the values $B = 38 \pm 2 \times 10^{-6}$ K$^{-3/2}$ and $C = 1.4 \pm 0.5 \times 10^{-8}$ K$^{-5/2}$, respectively. The present values of $B$ and $C$ for Metglas® 2826 together with those determined from previous magnetic[9,10] and Mössbauer[7,8,29] studies on the sample are listed in Table I, where the corresponding values for crystalline Fe and Ni and other relevant magnetic parameters have also been included for comparison. It is evident from Table I that the presently determined values for $B$ and $C$ differ from those observed in previous magnetic studies[9,10] but are in excellent agreement with the values deduced from Mössbauer measurements.[4,7,8,29] Because of the wide difference in the Curie temperatures, a meaningful comparison between the values of the coefficients $B$ and $C$ found either in different crystalline ferromagnetic materials and/or in various amorphous ferromagnets can be made only when the normalized coefficients $B_{3/2} = BT_C^{3/2}$ and $C_{5/2} = CT_C^{5/2}$, as defined by an alternative form of Eq. (2), i.e.,

$$\Delta M(T)/M(0) = B_{3/2}(T/T_C)^{3/2} + C_{5/2}(T/T_C)^{5/2} + \cdots \tag{7}$$

are used in place of $B$ and $C$. Therefore, values for the normalized coefficients $B_{3/2}$ and $C_{5/2}$ are also given within the square brackets in Table I.

Although both crystalline and noncrystalline ferromagnets follow the predictions of Heisenberg model Eq. (2), the results for the glassy alloy under consideration differ from those for crystalline ferromagnets in at least two ways: (i) the values of $B_{3/2}$ exhibiting a close agreement among themselves for crystalline ferromagnets are roughly *four times* smaller in magnitude than that observed for the present alloy, and (ii) the temperature range in which the $T^{3/2}$ term dominates is *much wider* (up to about $0.5T_C$) in the case of Metglas® 2826 than that for crystalline Fe and Ni [$0 \leq T \leq 0.15T_C$ (Ref. 56)]. In spite of the fact that the amorphous $Fe_{40}Ni_{40}P_{14}B_6$ alloy and crystalline ferromagnets differ in the above respects, they all exhibit comparable values for the coefficient of $T^{5/2}$ term, $C_{5/2}$. Similar observations have been made from studies on other amorphous ferromagnetic alloys.[22,23,25,26,29,57] Moreover, there exists sufficient evidence to prove that the higher values for $B$ are characteristic of only the glassy state.[29]

For a crystalline ferromagnet in which crystal momenta are quantized, the conventional spin-wave theory[19] shows that the $q^2$ and $q^4$ terms in the spin-

TABLE I. Magnetization and spin-wave parameters, and mean-square range of the exchange interaction for amorphous $Fe_{40}Ni_{40}P_{14}B_6$ and crystalline Fe and Ni. Numbers in the parentheses denote estimated uncertainty limits. Note that because of the large uncertainty limits of the coefficient $C$, only the highest estimate of the rms range is given for Metglas® 2826.

| Sample | $g$ | $M_s(0)$ (G) | $T_C$(K) | $B(10^{-6}\ K^{-3/2})$ $[B_{3/2}]$ Magn. | $B(10^{-6}\ K^{-3/2})$ $[B_{3/2}]$ Möss. | $C(10^{-8}\ K^{-5/2})$ $[C_{5/2}]$ Magn. | $C(10^{-8}\ K^{-5/2})$ $[C_{5/2}]$ Möss. | $D$ (meV Å$^2$) Calc. | $D$ (meV Å$^2$) Obs. | $D/T_C$ (meV Å$^2$K$^{-1}$) Calc. | $D/T_C$ (meV Å$^2$K$^{-1}$) Obs. | $\langle r^2 \rangle$ (Å$^2$) |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Metglas® | 2.05[a] | 754[b] | 519.9[b] | 38[b] | 38[c,d] | 1.4[b] | 1.2[d] | 99[b] | 100[e] | 0.19[b] | 0.19[e] | ~6.4[f] |
| 2826 | (0.02) | | (0.2) | (2) | (2) | (0.5) | (0.5) | (3) | | | | |
| | | | 520[g] | [0.45][h] | [0.47] | [0.09] | [0.08] | 78[i] | | | | |
| | | | | (0.02) | (0.03) | (0.03) | (0.03) | | | | | |
| | | | 525[a] | 56.4[j] | 40[k] | | | 75[j] | | | | |
| | | | | | (2) | | | | | | | |
| | | | 537[c] | [0.71] | [0.50] | | | | | | | |
| | | | | | (0.02) | | | | | | | |
| Fe | 2.09[l] | 1752[l] | 1043[l] | 3.4[l] | | 0.1[l] | | 286[m] | 266[n] | 0.27[l] | 0.26[o] | 16$a^2$ (Ref. f) |
| | | | | (0.2) | | (0.1) | | (11) | (20) | (0.01) | (0.01) | (2) |
| | | | | [0.115] | | [0.04] | | 278[p] | 281[q] | | 0.27[q] | |
| | | | | (0.007) | | (0.04) | | (30) | | | | |
| | | | | | | | | 285[q] | | | | |
| Ni | 2.19[l] | 508[l] | 631[l] | 7.5[l] | | 1.5[l] | | 397[m] | 433[r] | 0.64[l] | 0.69[o] | 16$a^2$ (Ref. f) |
| | | | | (0.2) | | (0.2) | | (7) | | (0.01) | | (2) |
| | | | | [0.119] | | [0.15] | | | 340[s] | | 0.54[o] | |
| | | | | (0.003) | | (0.02) | | | (45) | | (0.07) | |

[a] Reference 5.

[b] Present work.

[c] Reference 8.

[d] Reference 29.

[e] Reference 28.

[f] Values estimated from Eq. (20) when the values of $B$ and $C$ determined in the present work and Ref. 56 are used; $a$ denotes the nearest-neighbor distance.

[g] Reference 3.

[h] Numbers within the square brackets are the values of normalized coefficients $B_{3/2}$ and $C_{5/2}$.

[i] Reference 10.

[j] Reference 9.

[k] Reference 7.

[l] Reference 56.

[m] Calculated using the value of $B$ obtained in Ref. 56 in Eq. (8).

[n] G. Shirane, R. Nathans, O. Steinsvoll, H. A. Alperin, and S. J. Pickart, Phys. Rev. Lett. 15, 145 (1965).

[o] Calculated using $D$ values observed in footnotes n, r, and s.

[p] Reference 26.

[q] Reference 22.

[r] H. A. Mook, R. M. Nicklow, E. D. Thompson, and M. K. Wilkinson, J. Appl. Phys. 40, 1450 (1969).

[s] T. Riste, G. Shirane, H. A. Alperin, and S. J. Pickart, J. Appl. Phys. 36, 1076 (1965).

wave dispersion relation Eq. (1) give rise to $T^{3/2}$ and $T^{5/2}$ terms, respectively, in Eq. (2) and the coefficients of the corresponding terms in these equations are related through the expressions

$$B = \zeta(\tfrac{3}{2})[g\mu_B/M(0)](k_B/4\pi D)^{3/2} \tag{8}$$

and

$$C = \zeta(\tfrac{5}{2})[g\mu_B/M(0)](k_B/4\pi D)^{5/2}(\tfrac{3}{4}\pi)\langle r^2\rangle \ , \tag{9}$$

where $D$ is the spin-wave stiffness constant, $\zeta(\frac{3}{2}) = 2.612$ and $\zeta(\frac{5}{2}) = 1.341$ are the Rieman $\zeta$ functions and $\langle r^2 \rangle$ is the average mean-square (ms) range of the exchange interaction. Despite the fact that translational invariance is totally absent in amorphous ferromagnets and hence crystal momentum is no longer a good quantum number, neutron-diffraction measurements[14,15,18,20,22,28] clearly show that the amorphous ferromagnets do indeed exhibit well-defined spin-wave excitations at long wavelengths and the spin-wave dispersion data display the same $q$ dependence as expected for crystalline ferromagnets [i.e., Eq. (1)]. At first thought the above-mentioned observation may sound puzzling but reconciliation with such a situation is brought about by a field-theoretical calculation of Herring and Kittel[58] which reveals that for spin waves with long wavelength (wavelength exceeding many times the average atomic distances) the details of atomic arrangement, periodic or otherwise, are not important. Recently, the existence of well-defined long-wavelength spin waves in amorphous ferromagnets has been established through various theoretical treatments employing a quasicrystalline approximation.[59,60]

As already mentioned above, the term $BT^{3/2}$ in Eq. (2) arises from the fact that the quadratic dispersion relation holds at small wave vectors [or equivalently, from the finding that the spin-wave density of states $g(\omega) \sim \omega^{1/2}$ for small magnon frequency $\omega$]. The observation that $B$ or $B_{3/2}$ values for glassy ferromagnets are consistently several times larger than those for crystalline ferromagnets, therefore, implies higher low-frequency density of states for the amorphous case. In conformity with the above argument, a number of theoretical models indicate that increased structural disorder results in higher spin-wave density of states for low-frequency excitations.[51,61,62] The values of the spin-wave stiffness constant $D$ computed from Eq. (8) using the observed $B$ values are listed in Table I. The general observation[22,26,29] that $D$ has a smaller value for ferromagnets in the amorphous state than for those in the crystalline state is found to be maintained in the present system too. A theoretical justification for this finding has been provided as follows. Starting from the Hamiltonian

$$H = -\sum_{i=1}^{N}\sum_{j=1}^{N} J_{ij}\vec{S}_i \cdot \vec{S}_j \tag{10}$$

[where $J_{ij} = J(|\vec{r}_i - \vec{r}_j|) = J_{ji}$ denotes the exchange interaction between the spins $\vec{S}_i$ and $\vec{S}_j$ localized on neighboring sites $i$ and $j$ with radius vectors $\vec{r}_i$ and $\vec{r}_j$, respectively] for a system of $N$ magnetic atoms contained in a volume $V$, it can be shown that in quasicrystalline approximation the expression for the spin-wave energy of an amorphous ferromagnet is given by[59]

$$E(\vec{q}) = 2S\rho_0 \int J(r_{ij})g(r_{ij})[1 - \exp(-i\vec{q}\cdot\vec{r}_{ij})]\, d^3r_{ij} \ , \tag{11}$$

where $\rho_0 = N/V$ is the average atomic density, $r_{ij} = |r_{ij}| = |\vec{r}_i - \vec{r}_j|$, $\vec{q}$ is the wave vector and $g(r_{ij})$ is the normalized pair correlation function which denotes the probability of finding atoms at the site $j$ when some chosen atom occupies the site $i$.

For a perfectly isotropic case and in the long-wavelength (small $q$) limit, the above expression reduces to

$$Dq^2 = \mathrm{Re}[E(\vec{q})] = \left[\tfrac{1}{3}S\rho_0 \int r_{ij}^2 J(r_{ij})g(r_{ij})\, d^3r_{ij}\right]q^2 \ . \tag{12}$$

By choosing the origin at the site $i$ and denoting the distance between $i$ and $j$ sites by $r$, Eq. (12) simplifies into

$$D = \tfrac{1}{3}S \int r^2 J(r)F(r)\, dr \ , \tag{13}$$

where $F(r) = 4\pi r^2 \rho_0 g(r)$ is the radial distribution function. Next, we introduce $J(r)$ in the form

$$J(r) = J(r_0 + \overline{r - r_0}) = J(r_0 + \delta r) \ ,$$

with $r_0$ representing the mean nearest-neighbor distance and assume that the exchange interaction is confined to the nearest neighbors only. Now that $\delta r << r_0$, $J(r)$ can be expanded into a Taylor series about $r = r_0$ in powers of $\delta r$ with the result

$$J(r) = J(r_0) + (\delta r)J'(r_0) = J(r_0) + \Delta J(r) \ , \tag{14}$$

where $\Delta J$ is the contribution to the exchange integral resulting from the distribution in the nearest-neighbor distance due to the structural fluctuations in amorphous materials. In case the first peak in $F(r)$ can be approximated by a Gaussian probability density function as follows:

$$F(r) = (2/\pi)^{1/2}(Z/\Delta r)\exp[-(r - r_0)^2/2(\Delta r)^2] \tag{15}$$

[$Z$ being the nearest-neighbor (NN) coordination number, $r_0 = \int_{-\infty}^{+\infty} r F(r)\, dr$, the mean value of the NN pair distances and $(\Delta r)^2 = \int_{-\infty}^{+\infty} (r - r_0)^2 F(r)\, dr$, the mean-square deviation of the NN pair distances], a lengthy but straightforward calculation yields

$$D = D(0)\{1 - 2Z(\Delta r/r_0)[\Delta J/J(r_0)]\} \ , \tag{16a}$$

with

$$D(0) = \tfrac{1}{3}[ZSJ(r_0)r_0^2] \ . \tag{16b}$$

An independent estimate of $J(r_0)$ provided by the expression for the Curie temperature $T_C$ in the molecular-field approximation[46]

$$\begin{aligned} T_C &= [S(S+1)/3k_B]N\langle J\rangle \\ &= [S(S+1)/3k_B]\int F(r)J(r)\, dr \\ &\approx [S(S+1)/3k_B]ZJ(r_0) \end{aligned} \tag{17}$$

enables us to express $D(0)$ as

$$D(0) = [k_B T_C r_0^2/(S+1)] \ . \tag{18}$$

Taking $T_C = 519.9$ K and $r_0 = 2.54$ Å$^{-1}$,[63] Eq. (18) gives the values of $D(0)$ for the present alloy as 193 and 145 meV Å$^{-2}$ for $S = \frac{1}{2}$ and 1, respectively. These $D(0)$ values together with the values $Z = 12$ (Ref. 63) and $\Delta r/r_0 \cong 0.04$ (Refs. 64 and 65) when used in Eq. (16a) give $\Delta J/J(r_0) = 0.5$ for $S = \frac{1}{2}$ and 0.3 for $S = 1$. It is immediately noticed that the above values of $\Delta J/J$ for $S = \frac{1}{2}$ and 1 are the same as those obtained earlier by fitting the molecular-field theory to the experimental data (Fig. 1). The observation that $D$ for amorphous ferromagnets is smaller than that for the crystalline counterparts has found a similar interpretation as above in terms of the theories[66,67] that assume a random-lattice model and employ a binary collision theory. These theories, like the one proposed above, give the expression for $D$ as

$$D = D(0)(1-\Delta^2) \ , \tag{19}$$

with $\Delta$ and $D(0)$ defined by Eqs. (6) and (16b), respectively.

In addition to the above observation, Table I shows that the value of $D$ deduced from the present magnetization measurements, though in obvious disagreement with the $D$ values determined from the previous magnetization studies, is very close to that obtained from Mössbauer measurements and that actually measured by neutron-diffraction experiments. Such a close agreement between the $D$ values as determined from magnetization, Mössbauer, and neutron-diffraction studies suggests that the conventional spin-wave theory *completely accounts* for the observed low-temperature magnetic behavior and that there is *no need* to invoke the concept of *additional excitations* or *diffusive modes* at least for the glassy alloy under consideration.

The mean-square (ms) range of the exchange interaction, defined as[18]

$$\langle r^2 \rangle = \frac{\int r^2 J(r) g(r)\, dr}{\int J(r) g(r)\, dr}$$

in the case of amorphous ferromagnets, can be estimated either from Eq. (9) or from the following relation:

$$\langle r^2 \rangle = [\zeta(\tfrac{3}{2})/\zeta(\tfrac{5}{2})](16/3k_B)(C/B)D \ . \tag{20}$$

The values of ms range of the exchange interaction for Metglas® 2826 and crystalline Fe and Ni evaluated from Eq. (20) are given in Table I. From the tabulated values, it is evident that the ms range of the exchange interaction for Metglas® 2826 roughly equals the mean nearest-neighbor transition-metal—transition-metal distance whereas in crystalline Fe or Ni it is about four times the nearest-neighbor distance. This implies that at low temperatures the exchange interaction in amorphous $Fe_{40}Ni_{40}P_{14}B_6$ is essentially confined to the nearest neighbors only. The above statement is further justified by the finding that within the error limits the value of $B_{3/2}$ for Metglas® 2826 is fairly close to the value of 0.512 deduced for the case $S=1$ from a theory based on a nearest-neighbor Heisenberg model.[68]

### C. Magnetic behavior in the critical region

As already mentioned in Sec. II, magnetization as a function of temperature in an applied field of 15 Oe exhibits a sharp kink which gives the magnetic ordering temperature $T_C = 520$ K to within ±1 K. Information about the sharpness of magnetic phase transition at $T_C$ is provided by the previous (zero-field) Mössbauer measurements[8] on Metglas® 2826. From these measurements, Chien and Hasegawa[8] have found $T_C = 537$ K to be sharply defined to within 2 K. This observation implies that $T_C$ is as sharp as 0.4% and that the critical behavior can be studied for values of $|\epsilon| = |(T-T_C)|/T_C$ in excess of $1.9 \times 10^{-3}$. Moreover, the fact that the crystallization temperature for the present glassy alloy lies at least 150 K above $T_C$ makes it possible to study critical magnetic behavior without inducing crystallization in the sample.

Magnetization data taken in the temperature range $(T_C - 50)$ to $(T_C + 50)$ K when plotted in the form of $\sigma^2$ vs $H/\sigma$ (with $H = H_{\text{ext}} - 4\pi NM$, where $N$ is the demagnetizing factor) isotherms at 0.5-K intervals, give a set of smooth curves. A *parabolic extrapolation*[11,37–39] of these curves to $H/\sigma = 0$ and $\sigma^2 = 0$ yields intercepts on $\sigma^2$ and $H/\sigma$ axes equal to $\sigma_s^2$, the spontaneous magnetization squared (for $T < T_C$), and $\chi_0^{-1}$, the inverse initial susceptibility (for $T > T_C$), respectively. The temperature dependence of $\sigma_s$ and $\chi_0^{-1}$ so obtained and shown in Figs. 1 and 3 has been employed to construct the $\sigma_s(d\sigma_s/dT)^{-1}$ vs $T$ (Fig. 2) and $\chi_0^{-1}(d\chi_0^{-1}/dT)^{-1}$ vs $T$ (insert in Fig. 3)

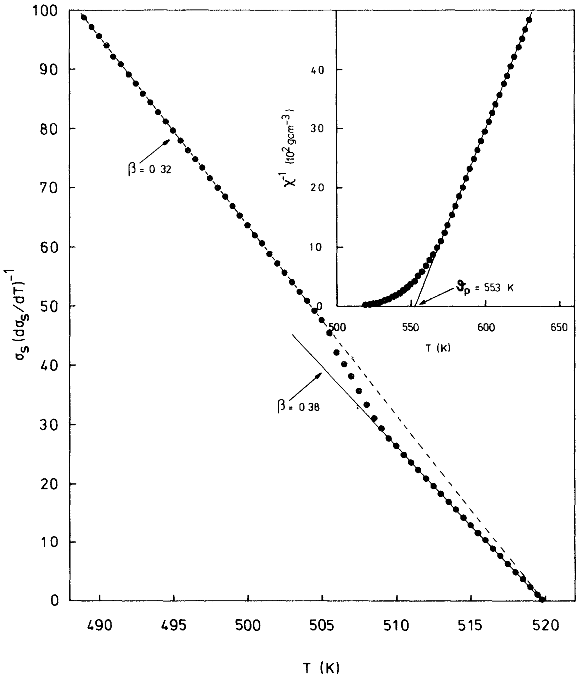

FIG. 2. $\sigma_s(d\sigma_s/dT)^{-1}$ vs $T$ plot for Metglas® 2826. This plot gives the critical exponent $\beta$ and Curie temperature (see text). Insert shows the inverse susceptibility as a function of temperature for $T > T_C$.

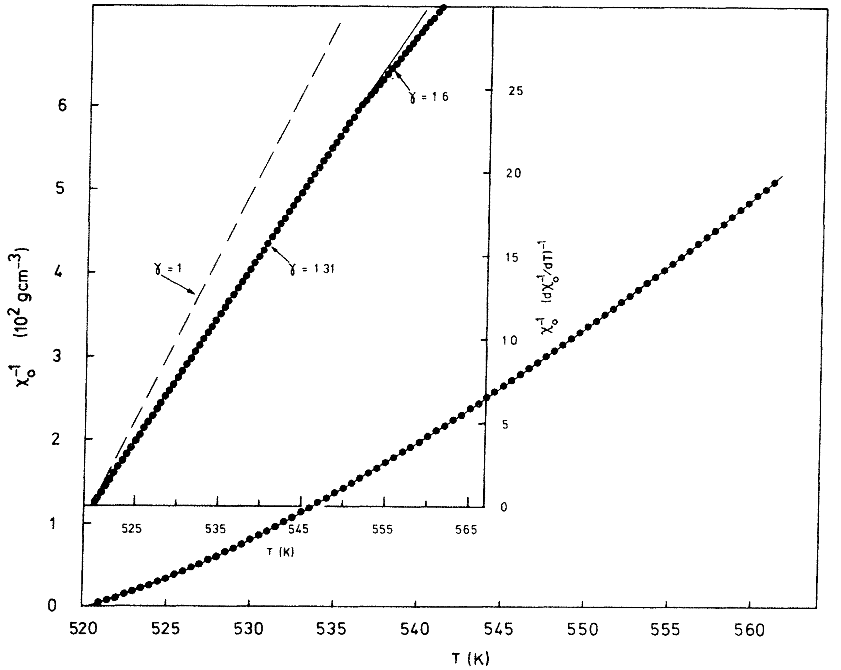

FIG. 3. Temperature dependence of the inverse initial susceptibility $\chi_0^{-1}$. The $\chi_0^{-1}(d\chi_0^{-1}/dT)^{-1}$ vs $T$ plot shown in the insert gives the critical exponent $\gamma$ and Curie temperature (see text).

plots which are known[36,37,69] to give precise values for $T_C$ and the critical exponents, $\beta$ and $\gamma$, defined by

$$\sigma_s = m_0(-\epsilon)^\beta, \quad \epsilon < 0 \tag{21}$$

and

$$\chi_0^{-1} = (h_0/m_0)\epsilon^\gamma, \quad \epsilon > 0 \tag{22}$$

[where $\epsilon = (T - T_C)/T_C$, and $m_0$ and $(h_0/m_0)$ are the critical coefficients], respectively, from the intercepts on their $T$ axes and from the inverse slopes of the straight-line portions of these plots. Furthermore, the log-log plots of $\sigma_s$ vs $|\epsilon|$ and $\chi_0^{-1}$ vs $|\epsilon|$ have been made in order to obtain values for the coefficients $m_0$ and $(h_0/m_0)$ from the intercepts on their ordinates. The values for critical parameters thereby obtained are $T_C = 519.9 \pm 0.2$ K, $\beta = 0.38 \pm 0.01$, $m_0 = 123.3 \pm 0.1$ emu/g and $\beta = 0.32 \pm 0.02$, $m_0 = 95.7 \pm 0.2$ emu/g in the temperature ranges $2.9 \times 10^{-4} \leq |\epsilon| \leq 2.2 \times 10^{-2}$ and $2.9 \times 10^{-2} \leq |\epsilon| \leq 5.9 \times 10^{-2}$ and $\gamma = 1.31 \pm 0.02$, $(h_0/m_0) = 14.05 \pm 0.01$ kOe g/emu, and $\gamma = 1.60 \pm 0.02$ in the temperature regions $6.7 \times 10^{-4} \leq |\epsilon| \leq 6.0 \times 10^{-2}$ and $6.0 \times 10^{-2} \leq |\epsilon| \leq 8.0 \times 10^{-2}$, respectively. It should be emphasized at this point that the values $\beta = 0.32 \pm 0.02$, $m_0/\sigma_s(0) = 0.95 \pm 0.01$, and $\gamma = 1.60 \pm 0.02$ match very well with their values deduced, respectively, from Mössbauer[8] and Hall-effect[70] measurements within the same or similar temperature ranges as quoted above. However, for temperatures in the immediate vicinity of the Curie point, both $\beta$ and $\gamma$ assume values which are very close to those reported[30,31,36–40] for other amorphous alloys. These values of $\beta$ and $\gamma$ together with other relevant parameters are listed in Table II. This table also includes the corresponding values found for critical-point parameters in crystalline Ni.[71] The present alloy, like other Ni-rich amorphous ferromagnetic alloys,[36–40] gives an increased value for $\gamma$ for temperatures in excess of $|\epsilon| \sim 0.06$ and thereby suggests that a short-range magnetic order persists for temperatures well above $T_C$.

In an attempt to find the value of $\delta$, the exponent for the critical isotherm, the $\sigma$-vs-$H$ data have been tested against the relation

$$\sigma = A_0 H^{1/\delta} \tag{23}$$

for the $T_C$ values in the range 519.7 to 520.1 K. These extreme values of $T_C$ are obtained from Figs. 2 and 3. The lower part of Fig. 4 shows the typical result for the value $T_C = 520$ K. It is clearly seen that Eq. (23) is obeyed in the above temperature range. This observation suggests the following relation between $T_C$ and $\delta$

$$T_C = 520.0 - 4.467(\delta - 4.44) \ . \tag{24}$$

TABLE II. Experimental and theoretical values for the critical-point parameters. Numbers in the parentheses denote estimated uncertainty in the least significant figure.

|  |  |  |  | Ising (fcc) |  | Heisenberg (fcc) |  |
|---|---|---|---|---|---|---|---|
| Parameters | $Fe_{40}Ni_{40}P_{14}B_6$ | Ni[a] | MFT[b] | $S=\frac{1}{2},\infty$[c] | $S=(\frac{1}{2})$[d] | $S=\infty$[d] | $S=\frac{1}{2},\infty$[d,e] |
| $T_C$ (K) | 519.9(2) | 627.4 |  |  |  |  |  |
| $\beta$ | 0.38(1) | 0.378(4) | 0.5 | 0.312(5) | 0.35(5) | 0.38(3) | 0.35 |
| $\gamma$ | 1.31(2) | 1.34(1) | 1.0 | 1.250(1) | 1.43(1) | 1.405(2) | 1.40 |
| $\delta$ | 4.46(4) | 4.58(5) | 3.0 | 5.00(5) |  | 5.0(2) | 5.0 |
| $\delta$[f] | 4.45(14) | 4.54(6) |  |  |  |  |  |
| $\alpha$ | −0.07(4)[g] | −0.10(2)[g] | 0.0 | 0.125 |  | −0.14(6) | −0.1 |
| $\sigma_s(0)$ (emu/g) | 100.27 | 58.57 |  |  |  |  |  |
| $\mu_0(\mu_B/\text{at.\%})$ | 0.912 | 0.616 |  |  |  |  |  |
| $m_0$ (emu/g) | 123.3(1) | 83.3 |  |  |  |  |  |
| $h_0$ (kOe) | 1732.4(2.6) | 15 720 |  |  |  |  |  |
| $m_0/\sigma_s(0)$ | 1.230(2) | 1.42 | 1.73[h] | 1.486(1)[i] | 1.69[j,k] | 1.22[j,k] |  |
| $\mu_0 h_0/k_B T_C$ | 0.204 | 1.04 | 1.73[h] | 1.52[l] |  |  |  |
| $\mu_{\text{eff}}(\mu_B)$ | 7 |  |  |  |  |  |  |
| $c$ (%) | 13 |  |  |  |  |  |  |
| $D_0(10^{-3})$ | 1.426 | 33.1 |  |  |  |  |  |
| $D_0 m_0^{\delta}/h_0$ | 1.7(4) | 1.3(3) | 1.0 | 1.81[l] | 1.54[j,k] | 1.23[j,k] |  |

[a] Reference 71.

[b] Reference 76.

[c] C. Domb, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and M. S. Green (Academic, New York, 1974), Vol. 3, p. 357.

[d] G. S. Rushbrooke, G. A. Baker, Jr., and P. J. Wood, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and M. S. Green (Academic, New York, 1974), Vol. 3, p. 245 and the references quoted therein.

[e] H. E. Stanley, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and M. S. Green (Academic, New York, 1974), Vol. 3, p. 485.

[f] Value of $\delta$ calculated using the experimental values of $\beta$ and $\gamma$ in Eq. (25) of the text.

[g] Value of $\alpha$ computed from the relation $\alpha=2(1-\beta)-\gamma$ using the observed values of $\beta$ and $\gamma$.

[h] For $S=\frac{1}{2}$.

[i] A. J. Guttmann, J. Phys. A 8, 1249 (1975).

[j] S. Milosevic and H. E. Stanley, Phys. Rev. B 6, 986, 1002 (1972); R. Krasnow and H. E. Stanley, *ibid.* 8, 332 (1973).

[k] M. Barmatz, P. C. Hohenberg, and A. Kornblit, Phys. Rev. B 12, 1947 (1975); A. Aharony and P. C. Hohenberg, *ibid.* 13, 3081 (1976).

[l] C. Domb, J. Appl. Phys. 39, 620 (1968); W. C. Muellner and J. S. Kouvel, Solid State Commun. 15, 441 (1974).

A similar relation between $T_C$ and $\delta$ has been previously reported by Arrott and Noakes[72] for crystalline Ni. A comparison of Eq. (24) with the corresponding relation obtained for crystalline Ni shows that in the latter case $\delta$ is about 25 times more sensitive to the choice of $T_C$ than that found in the present case. Several transition-metal—metalloid-glassy alloys have been found to exhibit the same behavior[36–39] so far as the relation between $T_C$ and $\delta$ is concerned. The above observation basically points to the fact that in order to arrive at a reliable value for the exponent $\delta$, $T_C$ in such amorphous systems *need not* be as *sharply defined* as in crystalline ferromagnets. The $\delta$ value that comes out when $T_C=519.9\pm0.2$ is substituted in Eq. (24) and the value for $D_0=A_0^{-\delta}$ obtained from the intercept on the ordinate of the $\ln H$ vs $\ln\sigma$ plot taken at $T_C=519.9$ K are given in Table II.

At this stage, it is worth noting that the presently determined values for the critical exponents obey the static scaling relation[73–76]

$$\delta=1+(\gamma/\beta)\ , \tag{25}$$

within the experimental error (see Table II). Moreover, compliance with the above relation implies that the data should satisfy the magnetic equation of state[73–80]

$$m=f_{\pm}(h)\ , \tag{26}$$

where plus and minus signs denote temperatures above and below $T_C$, respectively, $m\equiv\sigma/|\epsilon|^{\beta}$ is the reduced magnetization and $h\equiv H/|\epsilon|^{\beta+\gamma}$ the reduced

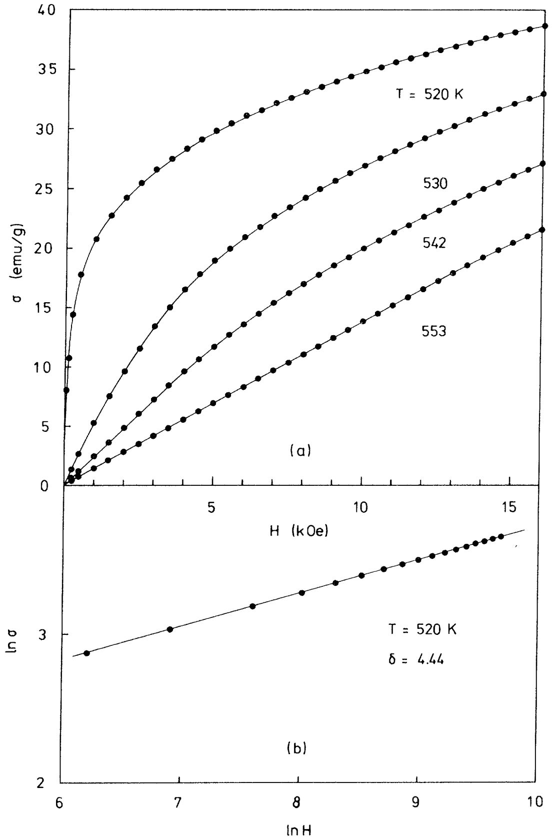

FIG. 4. Magnetization as a function of magnetic field for a few constant-temperature values $T \geqslant 520$ K (a). $\ln\sigma$ vs $\ln H$ plot for $T = 520$ K (b). The inverse slope of the least-square-fit straight line through the data points gives the value for the critical exponent $\delta$ as 4.44.

magnetic field. Equation (26) demonstrates that when $m$ is plotted against $h$, two universal curves result: $f_+(h)$ for $T > T_C$ and $f_-(h)$ for $T < T_C$. Instead of making the above-mentioned plot, it is customary[37–39,71,81] to plot $m^2$ vs $h/m$ to test the validity of Eq. (26). Such a plot shown in Fig. 5 clearly demonstrates that the present data satisfy the magnetic equation of state. In this figure, the intercepts of the universal curves with the axes specify the values of critical coefficients, $m_0^2$ ($T < T_C$) and $h_0/m_0$ ($T > T_C$), appearing in Eqs. (21) and (22). These coefficients, in turn, give the values for the normalized quantities $m_0/\sigma_s(0)$, $\mu_0 h_0/k_B T_C$ (where $\mu_0$ is the average magnetic moment per alloy atom at 0 K) and $D_0 m_0^\delta/h_0$ as shown in Table II. The theoretically predicted values for the critical exponents and critical amplitudes are also included in this table for comparison.

Table II shows that the observed critical exponent values are fairly close to the corresponding values for crystalline Ni whereas, amongst the theoretical values, they are in reasonable agreement only with those predicted by a three-dimensional Heisenberg model. Such an agreement at first sight would suggest that the short-range forces dominate in the critical region. But on closer examination, the exponent values present a slight but systematic shift (more marked in $\gamma$ and $\delta$ values) towards molecular-field values. In order to ascertain whether or not long-range forces (e.g., dipole forces) have any influence on the critical fluctuations of magnetization, we use the criterion of Kadanoff *et al.*[82] that the effect of

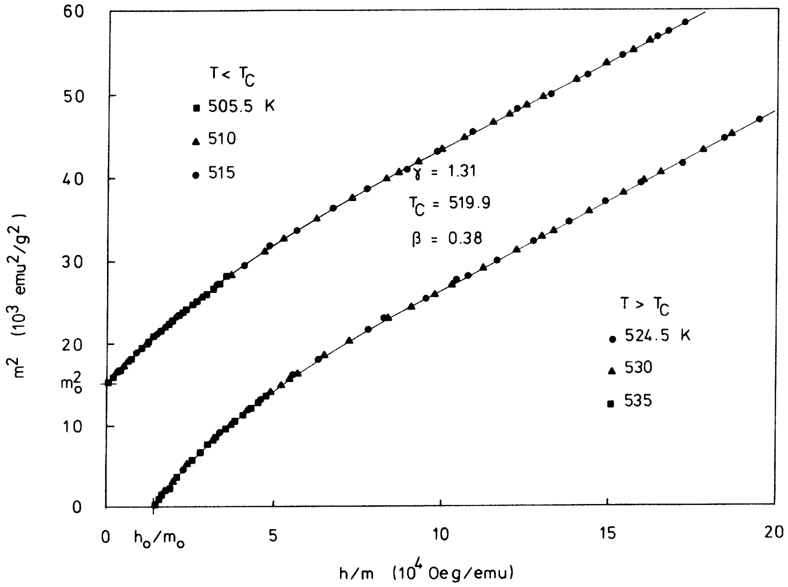

FIG. 5. Square of reduced magnetization $m^2$ vs reduced inverse susceptibility $h/m$ for temperatures below and above Curie temperature.

long-range forces can be neglected in case

$$|\epsilon| >> [\mu M_s(0)/k_B T_C]^{1/\beta(\delta-1)} \equiv t \ ,$$

where $\mu = g\mu_B S$ and $M_s(0)$ is the saturation magnetization at 0 K. Using the typical value of $t \cong 2 \times 10^{-3}$ for crystalline Fe or Ni and remembering that for our alloy the critical exponents could be determined up to $|\epsilon| = 2 \times 10^{-2}$ for $T < T_C$ and $6 \times 10^{-2}$ for $T > T_C$, we find that $|\epsilon|$ is at least $10 \times t$. It is, therefore, not surprising that the present exponent values closely follow the predictions of the Heisenberg model. Additional support to the above statement is provided by the observation that the specific-heat exponent $\alpha$, calculated using the observed values of $\beta$ and $\gamma$ in the relation $\alpha = 2(1-\beta) - \gamma$, agrees both in magnitude and sign (see Table III) with its value in terms of Heisenberg model. Furthermore, long-range forces are expected to get severely damped in amorphous materials with a very short mean-free path.[83,84] In view of the foregoing text, the finding that the critical exponent values exhibit a systematic shift towards the molecular-field values should imply that the exchange interactions in the critical region are not confined only to the nearest neighbors as normally assumed in the theories based on Heisenberg model but instead involve the next-nearest neighbors too. A firm theoretical basis for the above argument is provided as follows. Within the framework of a classical Heisenberg model, Binder and Müller-Krumbhaar employing a "self-consistent" Monte Carlo technique have shown that the values of $\beta$ and $\delta$ change from $0.33 \pm 0.02$ and $5.1 \pm 0.2$, in case only the nearest-neighbor interactions ($J_1$) (Ref. 78) are considered, to $0.39 \pm 0.02$ and $4.0 \pm 0.3$, respectively, when the next-nearest-neighbor interactions ($J_2 = 5J_1$) (Refs. 85 and 86) are also included. Using the series expansion method, Paul and Stanley[87] have demonstrated that $\gamma$ decreases slowly with an increasing $J_2/J_1$ ratio and attains a constant value of 1.2 for $J_2 \geqslant 5J_1$. One at once notices that the exponent values quoted above for the case $J_2 = 5J_1$ not only conform well with the presently determined values but also satisfy, within the error limits, the static scaling relation, Eq. (25). It is, however, not clear as to how one can reconcile to a situation wherein the exchange interactions between the next-nearest neighbors are five times stronger than those between the nearest neighbors. One possibility could be the superexchange interactions between the next-nearest-neighbor magnetic atoms mediated by a metalloid atom separating them but this does not explain why similar exponent values are also observed for crystalline Ni. At this point, it should be hastily added that the universality hypothesis, which predicts critical exponents to be independent of such *irrelevant* features of the Hamiltonian as the ratio $J_2/J_1$, has to date not found a firm support in terms of the theoretical calculations based on Heisenberg model.

Additional features presented by Table II can be summarized as: (i) While $m_0/\sigma_s(0)$ value for Ni lies between the values given by the Heisenberg model for $S = \frac{1}{2}$ and $\infty$, the corresponding value for the alloy under consideration is in striking agreement with the Heisenberg value for $S = \infty$. The above observation is consistent with the fact that the observed exponent values too are closer to the Heisenberg values for $S = \infty$ than to those for $S = \frac{1}{2}$. (ii) The experimental values of the normalized critical amplitude $D_0 m_0^\delta / h_0$ for both Metglas® 2826 and crystalline Ni compare with the theoretical values the same way as those of $m_0/\sigma_s(0)$. However, large uncertainty in the determination of $D_0 m_0^\delta / h_0$ arising basically from the observed error limits for $\delta$ makes the comparison with theory less conclusive than that for $m_0/\sigma_s(0)$ primarily because the latter normalized quantity can be estimated with much greater accuracy (see Table II) than the former one. (iii) Only in the case of Ni does the experimentally evaluated value of $\mu_0 h_0 / k_B T_C$ show any agreement with theory; the value for amorphous $Fe_{40}Ni_{40}P_{14}B_6$ alloy is at least one order of magnitude smaller. Since $h_0$ is presumably an effective exchange interaction field, the product of $h_0$ and an average effective elementary moment ($\mu_{\text{eff}}$) involved in the critical-point transition is expected to equal $k_B T_C$. Such seems to be the case for Ni when $\mu_{\text{eff}}$ is identified with $\mu_0$. However, this is clearly not the case for the alloy in question unless $\mu_{\text{eff}}$ is taken to be very much larger than $\mu_0$. Therefore, in order that $\mu_{\text{eff}} h_0$ equals the theoretical estimate (i.e., $\cong 1.6$, the mean theoretical value), we must take $\mu_{\text{eff}}$ for the present alloy to equal $7\mu_B$ (Table II). Moreover, if the concentration of such effective moments is $c$, then $c = \mu_0/\mu_{\text{eff}}$. The value of $c$ obtained in this way is given in Table II. Evidently, $c$ in our case possesses a much smaller value than the atomic concentration of either Fe or Ni. This finding implies that only a small fraction ($\sim 0.3$) of the Fe atoms (considering the fact that Ni in this alloy has a negligibly small moment[88]) plays a central role in the magnetic order-disorder transition.

### D. Magnetic behavior above the Curie temperature

Above $T_C$ Metglas® 2826 exhibits a superparamagnetic behavior as inferred from (i) the increased value of $\gamma$ above $T \sim 551$ K (insert of Fig. 3) which is a manifestation of the fact that the short-range magnetic order persists for temperatures well above $T_C$, (ii) magnetization-versus-field isotherms taken at as high temperatures as $T \sim 570$ K in fields up to 16 kOe (such isotherms are shown in the upper part of Fig. 4 for a few temperature values up to 553 K)

TABLE III. Parameter values obtained by Curie-Weiss analysis of $\chi$ vs $T$ curve for Metglas® 2826; for comparison, the corresponding values for crystalline Fe and Ni are also included. $T_d$ denotes the temperature above which the Curie-Weiss law is obeyed.

| Sample | $\mu = gS$ ($\mu_B$/TMA) | $p_{\text{eff}} = g[S(S+1)]^{1/2}$ ($\mu_B$/TMA) | $\mu_p = gS$ ($\mu_B$/TMA) | $T_C$ (K) | $\Theta_p$ (K) | $(\Theta_p - T_C)/T_C$ ($10^{-2}$) | $T_d$ (K) | $C$ ($10^{-2}$ emu K g$^{-1}$) |
|---|---|---|---|---|---|---|---|---|
| $Fe_{40}Ni_{40}P_{14}B_6$ | 1.14[a] | 3.17[a] | 2.14[a] | 519.9[a] | 553[a] | 6.35 | 575[a] | 1.58[a] |
| | 1.14[b] | 3.13[b] | 2.10[b] | 520[b] | 556[b] | 6.92 | 580[b] | 1.54[b] |
| Fe (Cryst.) | 2.216[c] | 3.20[c] | 2.32 | 1043[c] | 1093[c] | 4.79 | | 2.27[c] |
| Ni (Cryst.) | 0.616[c] | 1.61[c] | 0.85 | 631[c] | 650[c] | 3.01 | | 0.55[c] |
| | | 1.38[d] | 0.70 | | 712[d] | 12.84 | 1102[d] | 0.405[d] |

[a] Present results.

[b] Reference 3.

[c] Values taken from R. S. Tebble and D. J. Craik, *Magnetic Materials* (Wiley, New York, 1969); and A. H. Morrish, *The Physical Principles of Magnetism* (Wiley, New York, 1965).

[d] Reference 91.

show a finite curvature characteristic of a superparamagnetic behavior, and (iii) the alloy disorders magnetically only above 575 K (insert of Fig. 2). The magnetization-versus-field isotherms for $T \geqslant 520$ K have, therefore, been analyzed in terms of the relation[89,90]

$$\sigma(H,T) = \chi^{+}(T)H + \mu^{+}(T)c^{+}(T)B_s(x) \ ,$$

where $\mu^{+}$ and $c^{+}$ are the average cluster moment in $\mu_B$ and the number of clusters per gram, respectively, $B_s(x)$ is the Brillouin function with $x = \mu^{+}H/k_BT$ and $\chi^{+}(T)$, defined as $\chi^{+}(T) = \lim_{H\to\infty}(d\sigma/dH)_T$, is the high-field susceptibility which is normally temperature dependent. Using the values of $\chi^{+}$ computed at different temperatures from the slope of $\sigma$ vs $H$ curves at 15 kOe, we estimate the values for $\mu^{+}(c^{+})$ at $T = 522$ and 542 K as $2.1 \times 10^3 \mu_B \cdot (0.8 \times 10^{18}\ \text{g}^{-1})$ and $1.1 \times 10^3 \mu_B\ (1.0 \times 10^{18}\ \text{g}^{-1})$, respectively.

In the temperature region 575–645 K, the susceptibility obeys the Curie-Weiss law $\chi = C/(T - \Theta_p)$ (insert of Fig. 2) with the values for $C$, the Curie constant, and $\Theta_p$, the paramagnetic Curie temperature, given in Table III. For comparison, the corresponding values for crystalline Fe and Ni are also included in this table. The values for $C$, $\Theta_p$, and $T_C$ deduced from the present magnetization measurements are in excellent agreement with their values found in Ref. 3. Above 645 K the structural relaxation effects become apparent when the data points start deviating from the Curie-Weiss law behavior and at $T = 651$ K the susceptibility exhibits a sharp jump indicating a transition to the crystalline state. This value of the crystallization temperature $T_{\text{cr}} = 651$ K also agrees fairly well with its value determined in Refs. 3 and 11. From the Curie constant $C$, we obtain $p_{\text{eff}} = g[S(S+1)]^{1/2}$ which, in turn, gives the value for the average paramagnetic moment $\mu_p = gS$ as $2.14\mu_B$ per transition-metal atom. Expressing the sample composition as $(Fe_xNi_{1-x})_{80}P_{14}B_6$ with $x = 0.5$ and taking the moment per Ni atom ($\mu_{\text{Ni}}$) as $0.1\mu_B$ for this particular alloy composition,[88] it follows from the relation $\bar{\mu} = (1-x)\mu_{\text{Ni}} + x\mu_{\text{Fe}}$ (where $\bar{\mu}$ is the average moment per transition-metal atom and $\mu_{\text{Fe}}$ is the moment per Fe atom), using $\bar{\mu} = 1.14\mu_B$, that $\mu_{\text{Fe}} = 2.18\mu_B$. This value is very close to that determined for $\mu_p$.

In crystalline Fe and Ni, the moment in the paramagnetic state exceeds that in the ferromagnetic state (see Table III). This enhancement has been understood[91] in terms of a *nonlocalized negative* spin polarization of conduction electrons amounting to $0.21\mu_B$ per Fe atom[92] and $0.105\mu_B$ per Ni atom,[93] as determined from polarized neutron-diffraction measurements. However, in amorphous $Fe_{40}Ni_{40}P_{14}B_6$ alloy, such a moment enhancement is not obvious. This permits us to conclude that spin polarization of conduction electrons, if any, gives a negligible contribution to the distribution of the magnetic moment in the present alloy and that moments on Ni and Fe atoms are localized[94] even in the paramagnetic state.

### E. Exchange interactions

Two approaches, both based on the Heisenberg model, are generally followed to estimate the value of effective exchange interaction in ferromagnetic materials. In the first approach, the value of $J$ is determined by applying spin-wave theory to the measured low-temperature saturation magnetization [i.e., by first evaluating the value of $D$ from Eq. (8) and then

TABLE IV. Values of $J$ deduced from spin-wave theory and the Rushbrooke-Wood formula. [The values of the stiffness constant $D$ directly measured by neutron-diffraction experiments have been used in Eq. (16b) to calculate the values for $J$. Such a choice of $D$ values does not alter the results in any significant way since the $D$ values deduced from Eq. (8) using the $B$ values obtained from the magnetization measurements are quite close to those observed in neutron-diffraction studies.] SW—spin wave; HT—high-temperature series expansion.

| Material | $S$ | $Z$ | $D$ (meV Å$^2$) | $T_C$ (K) | $r_0$ (Å) | $J$ (K) [SW, Eq. (16b)] | $J$ (K) [HT, Eq. (27)] |
|---|---|---|---|---|---|---|---|
| Metglas® 2826 | 1.00 | 12[a] | 100[b] | 520 | 2.54[a] | 45 | 43 |
|  | 0.50 |  |  |  |  | 90 | 125 |
| Fe | 1.11 | 8 | 281[c] | 1043 | 2.48 | 179 | 116 |
| Ni | 0.30 | 12 | 387[d] | 631 | 2.49 | 603 | 335 |

[a] Reference 63.
[b] Reference 28.
[c] Reference 22.
[d] Because of the significant difference in the values of $D$ observed from neutron-diffraction studies on Ni, the mean value is used for computing the value of $J$.

substituting it in Eq. (16b)] whereas in the second one $J$ is calculated from the expression

$$k_B T_C / J = \frac{5}{96}(Z-1)[11S(S+1)-1] \ , \qquad (27)$$

which is obtained, as shown by Rushbrooke and Wood,[95] by expanding the susceptibility above the Curie temperature in powers of $J/k_B T$. Various symbols appearing in Eq. (27) have their usual meaning.

The values of $J$ obtained from the above-mentioned methods for Metglas® 2826 and for crystalline Fe and Ni are listed in Table IV. The main features of this table can be summarized as follows: (i) for amorphous $Fe_{40}Ni_{40}P_{14}B_6$ alloy, the deduced $J$ values are in excellent agreement with each other in the case $S=1$ whereas they are in obvious disagreement (high-temperature value being ~1.4 times greater than the low-temperature value) for $S=\frac{1}{2}$ and (ii) contrary to the above observation, the low-temperature $J$ values greatly exceed those found at high temperatures for crystalline Fe and Ni. The discrepancy between the $J$ values observed for Fe and Ni basically points to the fact that some of the $3d$ electrons in them are itinerant and hence the Heisenberg interaction remains but a poor approximation for iron-group metals. A situation similar to that found above for the case $S=\frac{1}{2}$ in Metglas® 2826 is found to occur in crystalline Gd too.[33,43] The discrepancy between the high- and low-temperature $J$ values in that case has been attributed[96] to the interactions beyond nearest neighbors which are thought to be of the oscillatory Rudderman-Kittel-Kasuya-Yosida (RKKY) type.[83] Recently, Kaneyoshi and co-workers[84,97] have conjectured that in amorphous magnetic alloys there exists an indirect exchange interaction which for crystalline ferromagnets takes the usual RKKY form. In view of the fact that the observed $g$ (Table I) and $p_{\mathrm{eff}}$ (Table III) values indicate for Metglas® 2826 a spin value $S=1.11$ (a value very close to that observed for Fe) for which both spin-wave theory and high-temperature series method yield values for $J$ in agreement with each other, the above remarks lead us to the following conclusions: (a) the present glassy alloy is a better Heisenberg ferromagnet than crystalline Fe or Ni and (b) no evidence for the type of indirect exchange interaction as conjectured by Kaneyoshi is found in amorphous $Fe_{40}Ni_{40}P_{14}B_6$ alloy. Both these conclusions are consistent with our earlier deductions.

## IV. CONCLUSIONS

Exhaustive magnetization measurements performed on the amorphous ferromagnet $Fe_{40}Ni_{40}P_{14}B_6$, besides removing the doubts raised by the previous investigations concerning the magnetic behavior of this glassy alloy, permit us to draw the following conclusions: (i) conventional spin-wave theory completely accounts for the observed low-temperature magnetization behavior, (ii) the spin polarization of conduction electrons (in the ferromagnetic state) makes a negligible contribution to the distribution of magnetic moment, (iii) moments on Fe and Ni atoms remain localized in the entire temperature range $T \leq 645$ K, (iv) the exchange interaction, though confined to the nearest neighbors only at low temperatures, involves the next-nearest neighbors also in the critical region, and (v) the elementary moments involved in the magnetic order-disorder transition are those forming a part of the giant superparamagnetic moments which are found to exist for temperatures well above $T_C$.

To summarize, the Heisenberg interaction represents an approximation better suited to amorphous ferromagnets like Metglas® 2826 than to crystalline ferromagnetic materials like Fe or Ni. A distribution of the exchange integrals in the Heisenberg model provides an adequate description for the influence of amorphousness on the magnetic properties of $Fe_{40}Ni_{40}P_{14}B_6$ alloy. Structural disorder results in a reduction of the exchange coupling constant, $J$, and the spin-wave stiffness constant, $D$.

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