

Magnetic properties of amorphous $\text{Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ alloys ($x=1,2,3$): A further key to understand the magnetism of amorphous $\text{Ni}_{81.5}\text{B}_{18.5}$

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The observed apparent ferromagneticlike behavior of melt-quenched amorphous (*a*) $\text{Ni}_{81.5}\text{B}_{18.5}$ alloys has usually been interpreted as resulting either from very weak itinerant ferromagnetism of the amorphous matrix with a Curie point well above room temperature or from the contribution of Ni-rich segregations which are embedded in a Pauli paramagnetic amorphous matrix. At the same time, structural studies on melt-quenched Ni-B alloys around the eutectics at B=17 at. % revealed in several cases the presence of crystalline (Ni and Ni_3B) precipitates, indicating poor glass formation for these particular alloy compositions. On the other hand, it has been a general experience that introducing a third element promotes the formation of a more homogeneous amorphous state. By choosing Fe as the third component, we have found a significant suppression of ferromagnetism in the ternary alloys and obtained a Curie point as low as about 50 K even for 3 at. % Fe i.e., paramagnetism was observed at room temperature where the binary alloy exhibited a ferromagneticlike behavior. Measurements of the low-field magnetization and ac susceptibility and the magnetization isotherms, the latter analyzed with the help of Arrott plots, were performed for melt-quenched *a*- $\text{Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ ($x=1,2,3$) alloys, and the magnetic transformation temperatures were determined. These results gave further support for the paramagnetic behavior of the matrix of the amorphous binary alloy and for ascribing the observed ferromagneticlike magnetization to Ni-rich segregations. From the results of the present study, the magnetic phase diagram of amorphous $\text{Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ alloys could be constructed that indicated the presence of a spin-glass state as well, similarly to other Ni-Fe metalloid alloys.

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I. INTRODUCTION

Several reports¹⁻⁸ have been published on the magnetic properties of melt-quenched amorphous Ni-B alloys with the composition of about B=18.5 at. %. A common feature of the observations^{1,3,4,6} was that at and below room temperature the magnetization of these amorphous alloys was dominated by a small, ferromagneticlike component with saturating magnetization values of about 1 emu/g. The saturation field varied in a wide range (2–20 kOe), whereas the magnetization at a sufficiently high field (≥ 10 kOe) decreased only very slowly with increasing temperature, apart from the liquid helium range. The high-field susceptibility χ_{HF} was of the order of 10^{-6} – 10^{-5} emu/g and varied only slightly with temperature.

There has been a controversy in interpreting the observed magnetic behavior of melt-quenched $\text{Ni}_{81.5}\text{B}_{18.5}$ amorphous alloys. Based on an Arrott-plot evaluation of the magnetization isotherms at 4.2 and 297 K, Kaul and Rosenberg³ deduced very weak itinerant ferromagnetism (VWIF) with a Curie temperature of 450 K. Subsequently, they have reported ferromagnetic resonance experiments⁷ as well in support of the above conclusion. On the other hand, Takahashi and co-workers^{2,5} and Bakonyi and co-workers^{4,6,8} ascribed the observed ferromagnetism to strongly magnetic (Ni-rich)

segregations and concluded that the amorphous matrix of these alloys exhibits Pauli paramagnetism. Studies of the magnetic susceptibility in the liquid state of Ni-B alloys with the same composition^{2,5,6} as well as ¹¹B NMR line shift measurements⁴ yielded further evidence in favor of this interpretation.

In order to rationalize these two strikingly different interpretations, it might be worth making some considerations concerning the poor glass-forming ability (GFA) of this particular alloy composition $\text{Ni}_{81.5}\text{B}_{18.5}$ which is close to the most Ni-rich eutectics at 17 at. % B.⁹ The problem of the GFA has been widely addressed in general¹⁰⁻¹² and, specifically, also for the Ni-B alloy system.¹³ It has been established that alloy compositions around deep eutectics are especially prone to glass formation by melt quenching due to the strongly suppressed nucleation rate at these compositions.¹⁰ It turned out,^{11,12} however, that whereas glass formation is not restricted to eutectic compositions only, at certain eutectics the GFA has remained poor. This seems to be the case also for the $\text{Ni}_{83}\text{B}_{17}$ eutectic point since, although several attempts have been made to vitrify Ni-B alloys around this eutectics, the results reported remained rather controversial. Based on an x-ray diffraction (XRD) test, glass formation has been found for $\text{Ni}_{100-x}\text{B}_x$ alloys with $x=17$ – 18.5 (Ref. 14), $x=18$ (Ref. 15), $x=18.5$ (Refs.

1, 3, 16, and 17), $x=19$ (Ref. 16), and $x=20$ (Ref. 18). Neutron scattering experiments have also indicated glass formation for $x=18$ and 20 (Ref. 18). The formation of a completely crystalline state was detected by transmission electron microscopy (TEM) for $x=18$ (Ref. 19), and partially crystalline ribbons were quenched, according to the XRD analysis, from the melt for $x=17$ and 20 (Ref. 16) and for $x=15$ (Refs. 15 and 16). However, even “XRD amorphous” alloys can contain a small amount (about 2–3%: see Ref. 14) of crystallites due to the resolution limit of this detection technique. Indeed, Takahashi and co-workers^{1,16} have observed by metallurgical microscopy and TEM the presence of fine Ni and Ni₃B crystallites in an as-quenched amorphous Ni_{81.5}B_{18.5} alloy exhibiting a halo-type XRD pattern. The above results can be summarized by establishing that whereas an amorphous matrix can be obtained in melt-quenched Ni-B alloys around the eutectics at B=17 at. %, the presence of crystalline precipitates can still be expected in an amount depending on the particular quenching conditions and, therefore, Ni-rich segregations may well have occurred also in the alloys used for the above-described magnetization studies.

It is well known, however, that the introduction of a third component into a binary system can significantly improve the GFA. For the Ni_{81.5}B_{18.5} alloy, a partial replacement of B by Si (Ref. 14) or P (Ref. 20) atoms resulted in melt-quenched ternary amorphous alloys without substantial Ni segregations or precipitates. Along this line, we have now attempted to achieve an improvement of the GFA for Ni_{81.5}B_{18.5} by partially replacing Ni atoms by Fe atoms. The idea behind studying this alloying effect was that if the observed ferromagnetism in melt-quenched Ni_{81.5}B_{18.5} amorphous alloys was caused by Ni-rich segregations, the improved GFA will reduce the size and/or concentration of these segregations and, therefore, the magnetization behavior of the ternary alloy should exhibit drastic changes with respect to the binary alloy. Since Fe atoms possess larger magnetic moments than Ni atoms, a proper analysis of the variation of the magnetic properties upon introducing Fe into the Ni_{81.5}B_{18.5} alloy can give further hints at a better understanding of the origin of the observed ferromagnetism in the binary alloy.

The purpose of the present paper is to describe the results of magnetic measurements on melt-quenched a -Ni_{81.5-x}Fe_xB_{18.5} ($x=1,2,3$) alloys. Their behavior will be compared to previous observations^{1,3,4,6} on melt-quenched a -Ni_{81.5}B_{18.5} alloys. The present work was carried out to shed some more light on the controversies concerning the origin of the observed magnetic behavior of this binary alloy and, in general, concerning the appearance of ferromagnetism in the Ni-Fe-B system. The introduction of Fe, indeed, improved the GFA to the extent that the magnetic measurements indicated a magnetically (and, therefore, chemically) much more homogeneous state in the ternary alloys than was in the binary alloy. As typical for other Ni-Fe-metalloid alloys, the formation of a spin-glass state was also observed. The present results enabled us to construct the magnetic phase diagram of the a -Ni_{81.5-x}Fe_xB_{18.5} alloys.

The paper is organized as follows. Section II briefly de-

scribes the details of the sample preparation and measurements. The experimental results will be presented and analyzed in Sec. III. Then, in Sec. IV, the results obtained on the Ni-Fe-B ternary alloys will be used to discuss the magnetic properties of the binary Ni_{81.5}B_{18.5} alloy and to construct the magnetic phase diagram of the ternary Ni-Fe-B alloys. Finally, the conclusions of the present study will be summarized in Sec. V.

II. EXPERIMENT

The amorphous Ni_{81.5-x}Fe_xB_{18.5} ($x=1,2,3$) alloys were produced by a melt-spinning technique in the form of typically 1-mm-wide and 10- μ m-thick ribbons. Their amorphous state was checked by x-ray diffraction. The magnetization (σ) isotherms were measured in magnetic fields up to 16 kOe below room temperature in a vibrating sample magnetometer. The same equipment was also used to measure the magnetization in a magnetic field of $H=5$ or 10 Oe as a function of temperature either after zero-field cooling (ZFC) or after field cooling (FC) where the cooling was performed in a magnetic field of 5 or 10 Oe. Around the temperatures where a magnetic transition was indicated by the low-field magnetization measurements, the ac susceptibility was also studied for $x=1$ and 2 at 160 Hz frequency and with $H_{\max}=0.3$ Oe.

III. RESULTS AND DATA ANALYSIS

A. Magnetization isotherms

The low-temperature magnetization isotherms for the three amorphous alloys are presented in Fig. 1. For a given temperature and magnetic field, the value of the magnetization increases with increasing Fe content. However, the qualitative features of the magnetic behavior are very similar for each alloy: saturating, ferromagnetic (FM) magnetization isotherms at low temperatures and a linear increase of σ with increasing magnetic field for sufficiently high temperatures. The latter feature indicates a paramagnetic (PM) state and, thus, a FM-PM transition at intermediate temperatures.

B. Low-field magnetization and ac-susceptibility data

In order to characterize the magnetic transition temperatures of a -Ni_{81.5-x}Fe_xB_{18.5} alloys, the temperature dependence of the low-field magnetization and ac susceptibility was measured and the results are summarized in Fig. 2.

For $x=1$ [Fig. 2(a)], the FC and ZFC low-field magnetization curves are different at the lowest temperatures, indicating a spin-glass-like behavior. The same is shown by the cusp of the ac susceptibility as well. The spin-freezing temperature T_g identified by the peak position of the low-field magnetization versus temperature curve is 3.2 K. For $x=2$ [Fig. 2(b)] a reentrant spin-glass (RSG) behavior can be identified, indicated by a plateau of the low-field magnetization between 6 and 8 K. This behavior is characterized by two transition temperatures: (i) a spin-freezing temperature $T_f \approx 3.5$ K defined as the temperature of the steepest decay of the ZFC magnetization with decreasing temperature and (ii)

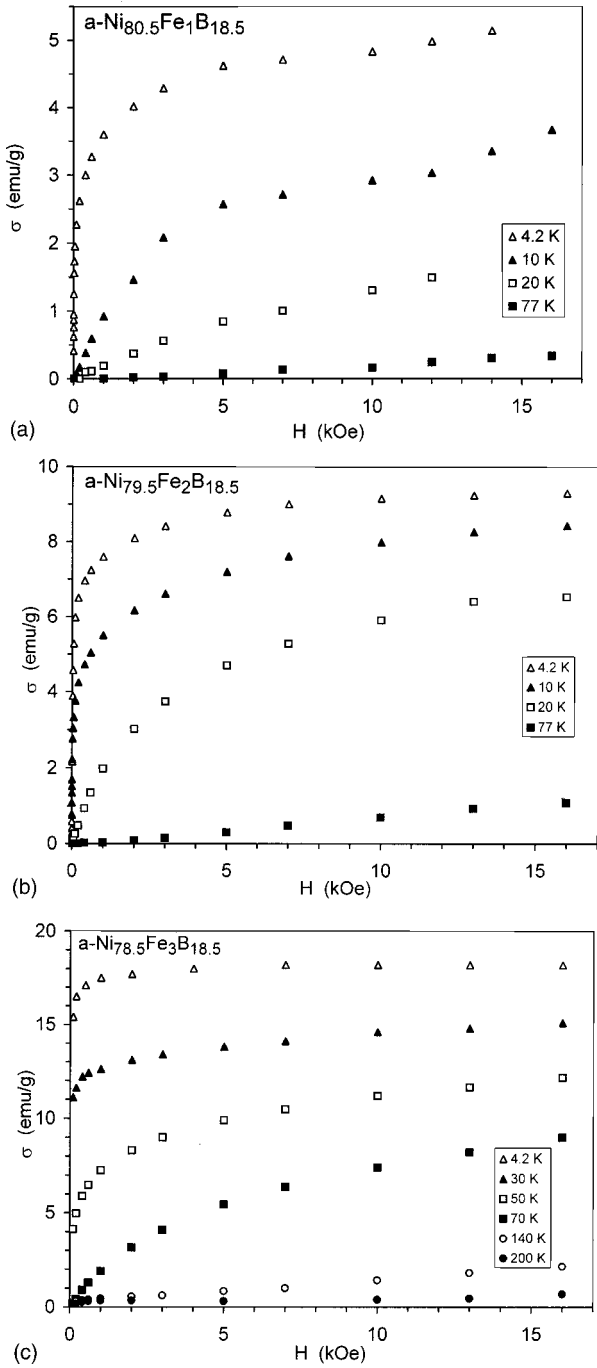


FIG. 1. Low-temperature magnetization isotherms of the amorphous $\text{Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ alloys: (a) $x=1$, (b) $x=2$, and (c) $x=3$.

a Curie temperature $T_c \approx 14.5$ K determined by extrapolating the steepest part of the high-temperature flank of the low-field magnetization curve to the abscissa. The larger T_f and T_g values from the ac-susceptibility data, identified as the ac-susceptibility peak temperatures [Figs. 2(a) and 2(b)], are due to the difference in the time scale between the two kinds of measurement technique.

For $x=3$ [Fig. 2(c)], the FC and ZFC curves completely coincide, indicating that this alloy already behaves as an ordinary ferromagnet without a spin-glass state at the lowest

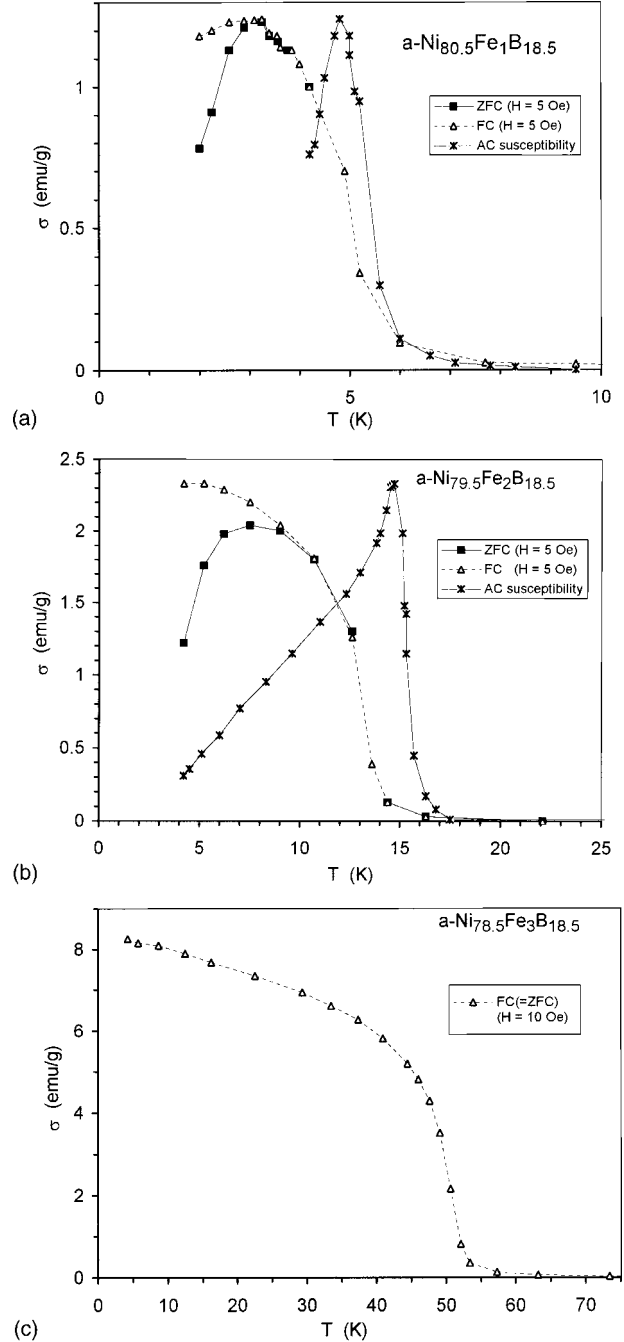


FIG. 2. Low-field magnetization σ (FC and ZFC) and ac susceptibility of the amorphous $\text{Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ alloys at low temperatures: (a) $x=1$, (b) $x=2$, and (c) $x=3$. The solid and dashed lines connecting the data points serve as a guide for the eye only. The ac susceptibility is displayed in arbitrary units.

temperatures. A Curie point of $T_c=52$ K can be determined for this alloy by the procedure used for $x=2$.

C. Arrott-plot analysis of magnetization isotherms

The magnetic behavior of ferromagnetics with relatively low Curie temperatures such as the $a\text{-Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ alloys has often been analyzed with the help of the so-called Arrott plots (σ^2 vs H/σ curves) as has been recently performed for

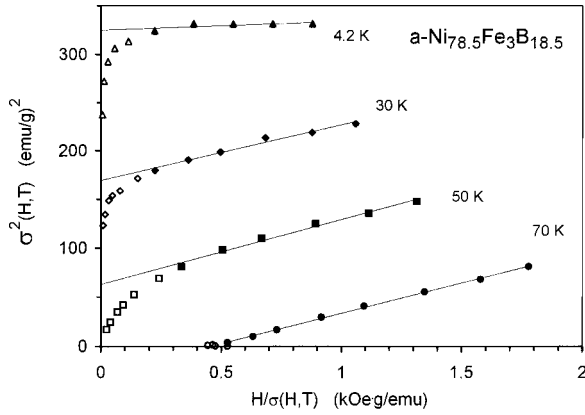


FIG. 3. Arrott plot $\sigma^2(H,T)$ vs $H/\sigma(H,T)$ of the amorphous $\text{Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ alloy with $x=3$ around the magnetic transition temperature [$T_c=52$ K from the low-field magnetization data in Fig. 2(c)]. The straight lines were fitted to the high-field data points only which are displayed by solid symbols.

amorphous Ni-P alloys²¹ around the critical concentration for the onset of spontaneous magnetic order. As Fig. 3 shows for $x=3$, for temperatures in the vicinity of the Curie point ($T_c=52$ K as deduced above from the low-field magnetization data), these Arrott plots consist of nearly parallel straight lines, apart from very low fields. This deviation from linearity can be ascribed to spatial heterogeneities of the magnetization (for more details, see Ref. 21, in which the original references on this problem can also be found). The extrapolation of the straight line portion of the isotherms at high magnetic fields to $H=0$ defines the spontaneous magnetization $\sigma(0,T)$ for a given temperature. The temperature dependence of the spontaneous magnetization for each alloy displayed as a $\sigma^2(0,T)$ vs T^2 plot in Fig. 4 can be used to determine the Curie temperatures, and the results are $T_c(\text{Fe}_1)=9.5$ K, $T_c(\text{Fe}_2)=19$ K, and $T_c(\text{Fe}_3)=64$ K. These values are upper limits only since, due to the limited number of data points for each sample, we have not considered the curvature of the $\sigma^2(0,T)$ vs T^2 plots in Fig. 4, which was the

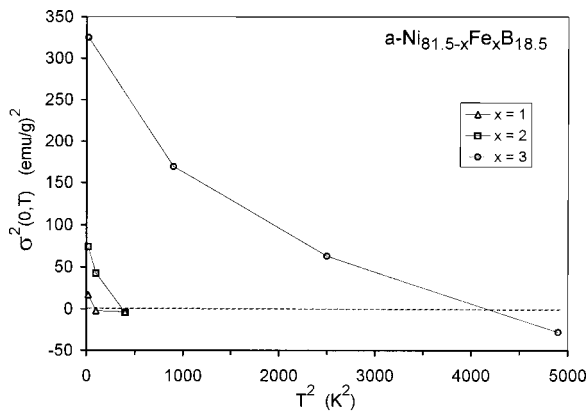


FIG. 4. Temperature dependence of the spontaneous magnetization $\sigma(0,T)$ displayed in the form of a $\sigma^2(0,T)$ vs T^2 plot for the amorphous $\text{Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ alloys with $x=1, 2$, and 3 . The $\sigma^2(0,T)$ values were obtained from the Arrott plots as shown for $x=3$ in Fig. 3. The solid lines serve as a guide for the eye only.

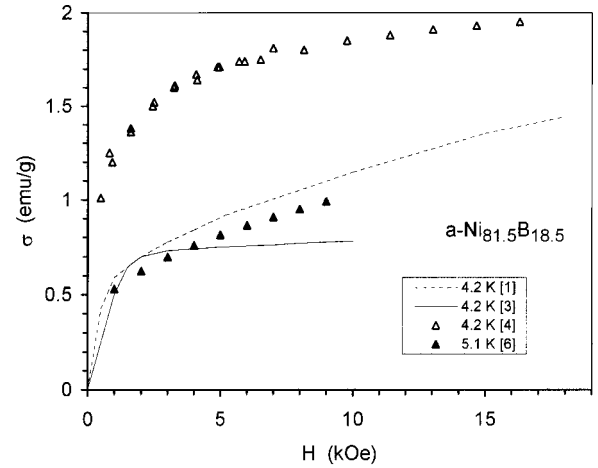


FIG. 5. Magnetization isotherms for amorphous $\text{Ni}_{81.5}\text{B}_{18.5}$ alloys in the liquid helium temperature range. The data are from the references as indicated in the legend.

typical case also for the amorphous Ni-P alloys.²¹ The higher T_c value in comparison with the low-field data (for $x=3$) or the existence of a nonzero T_c (for $x=1$ and 2) from the Arrott-plot analysis is partly due to the overestimated T_c values from Fig. 4 and partly due to the fact that the Arrott-plot analysis usually overestimates the magnetic transition temperature of an inhomogeneous ferromagnet as was discussed in detail for the case of amorphous Ni-P alloys.²¹

IV. DISCUSSION

Before discussing the spin-glass and FM states of amorphous $\text{Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ alloys with $x=1,2,3$ and the magnetic phase diagram of the Ni-Fe-B system, we should first clarify the magnetic state of the binary $a\text{-Ni}_{81.5}\text{B}_{18.5}$ alloy. For this purpose, we summarize the available magnetic data for the $a\text{-Ni}_{81.5}\text{B}_{18.5}$ alloy and compare them to the present results on the ternary system.

A. Magnetic properties of amorphous $\text{Ni}_{81.5}\text{B}_{18.5}$

The magnetization isotherms of $a\text{-Ni}_{81.5}\text{B}_{18.5}$ alloys^{1,3,4,6} measured in the liquid helium range are presented in Fig. 5, indicating that the observed magnetic behavior is very similar for all investigated samples and the magnetization is typically 1 emu/g in a magnetic field of about 10 kOe in each case. A rapid increase of the magnetization was observed at low fields ($H \leq 5$ kOe) and, at higher fields, the magnetization variation became much smaller, although saturation could not be achieved up to 50 kOe (Ref. 4). The observed magnetization isotherms indicate a ferromagneticlike behavior with a saturation magnetization of about 2–3 % of that of pure fcc Ni. The high-field susceptibility was reported to be 11×10^{-6} emu/g for the range 2–10 kOe (Ref. 3) and 3×10^{-6} emu/g for the range 20–50 kOe (Ref. 4).

In order to have a picture about the possible magnetic transformation temperatures, the results of high-field magnetization measurements are summarized in Fig. 6 for $a\text{-Ni}_{81.5}\text{B}_{18.5}$. Apart from the lowest temperatures, the magnetization decreases only slightly with increasing tempera-

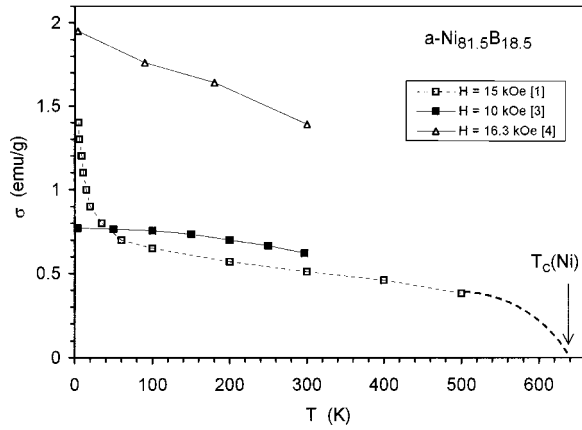


FIG. 6. Temperature dependence of the high-field magnetization for amorphous $\text{Ni}_{81.5}\text{B}_{18.5}$ alloys. The solid and dashed lines connecting the data points serve as a guide for the eye only. The dashed line extending the data of Ref. 1 to higher temperatures indicates that the Curie point of the FM-like magnetization contribution of $a\text{-Ni}_{81.5}\text{B}_{18.5}$ can be as high as even the Curie temperature of pure fcc Ni marked by the arrow.

ture in each case. The results in Fig. 6 support that a FM-PM transition occurs in these alloys well above room temperature only, probably not very far from the Curie point (631 K) of Ni indicated by the arrow. Other low-field⁴ (80 Oe) and high-field⁶ (17 kOe) magnetization studies also indicated that the magnetization decreases monotonously and smoothly from room temperature at least up to 500 K, similarly to the results of Takahashi *et al.*¹ shown in Fig. 6 (due to the onset of crystallization around 500 K, the decrease of σ cannot be measured for higher temperatures in the as-quenched amorphous state).

As mentioned in Sec. I, the observed magnetic behavior of $a\text{-Ni}_{81.5}\text{B}_{18.5}$ alloys can be explained by assuming^{2,4-6,8} that about 2–3% of the sample volume consists of Ni-rich segregations which have a transition temperature very close to that of fcc Ni (see Fig. 6). An Arrott-plot evaluation of the magnetization isotherms reported for the $a\text{-Ni}_{81.6}\text{B}_{18.4}$ alloy³ yielded, indeed, a Curie point of 450 K. We have now performed a similar Arrott-plot analysis for the magnetization data reported in Ref. 4 for an $a\text{-Ni}_{81.5}\text{B}_{18.5}$ alloy and also a high Curie point ($T_c = 510$ K) was obtained. The high T_c value for $a\text{-Ni}_{81.6}\text{B}_{18.4}$ alloy was attributed to the amorphous matrix with a VWIF character by Kaul and Rosenberg.³ However, when alloying Ni with *sp* elements in general²² and, specifically, with phosphorus in the form of an amorphous Ni-P matrix,²¹ the reduction of the saturation magnetization and Curie temperature has been found to occur at an approximately common rate with increasing metalloid concentration. This is itself a strong argument against the interpretation that an amorphous matrix of $a\text{-Ni}_{81.5}\text{B}_{18.5}$ alloys exhibits ferromagnetism with $\sigma_S \approx 1$ emu/g and $T_c \approx 500$ K. This would namely mean a ferromagnetic Ni-rich alloy for which the values of σ_S and T_c are 2% and 70%, respectively, of the corresponding values of fcc Ni, which is highly improbable. In addition to the evidences given previously,^{2,4,6} this further supports the argument that the presence of Ni-rich segregations in the Pauli paramagnetic matrix of the

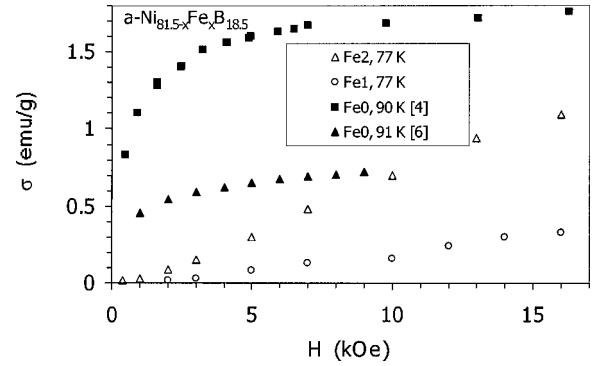


FIG. 7. Magnetization isotherms around liquid nitrogen temperature for the amorphous $\text{Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ alloys with $x = 1$ and 2 and for two amorphous $\text{Ni}_{81.5}\text{B}_{18.5}$ alloys (the data for the binary alloys were taken from Refs. 4 and 6).

amorphous $\text{Ni}_{81.5}\text{B}_{18.5}$ alloy (having probably a slight, about 0.5 at. % enrichment in B) is a more proper interpretation of the observed magnetic behavior depicted in Figs. 5 and 6 for these melt-quenched amorphous ribbons.

The rapid increase of σ when $T \rightarrow 0$ K for some alloys (Fig. 6) may indicate that the amorphous matrix contains also giant-moment paramagnetic clusters with a Curie-Weiss-type behavior,^{23,24} the contribution of which begins to dominate the observed magnetization in this temperature range, similarly to the case of amorphous Ni-P alloys just above the critical concentration of the FM-PM transition.²¹

A strong support for the presence of Ni-rich segregations in the binary alloy comes from a comparison of its magnetic behavior with the ternary alloys as well. Figure 7 shows the magnetization isotherms around liquid nitrogen temperature for two $a\text{-Ni}_{81.5}\text{B}_{18.5}$ alloys and for the $a\text{-Ni}_{80.5}\text{Fe}_{1.5}\text{B}_{18.5}$ and $a\text{-Ni}_{79.5}\text{Fe}_{2.5}\text{B}_{18.5}$ alloys. At these particular temperatures, the Fe-containing alloys show generally a smaller magnetization and a linear magnetization isotherm in contrast to the binary Ni-B alloys exhibiting here a FM-like behavior and a higher magnetization value. These results can be explained by the fact that the low GFA of the binary alloy resulted in the appearance of chemical inhomogeneities (Ni-rich segregations) during melt quenching, whereas for $x = 1$ and 2, the improved GFA led to a more homogeneous distribution of Ni and Fe in the alloy and, thus, to a decrease of the observed magnetization in spite of the presence of Fe atoms carrying a larger magnetic moment than Ni.

We can summarize the magnetic behavior of the $a\text{-Ni}_{81.5}\text{B}_{18.5}$ alloy as follows. The observed ferromagnetic behavior can be ascribed to the presence of about 2–3% Ni-rich segregations which are embedded in the amorphous matrix. The actual amount of these strongly magnetic segregations depends on the cooling rate during melt quenching. The amorphous matrix is Pauli paramagnetic with a high-field susceptibility of nearly 200×10^{-6} emu/mol (Ref. 4), which is close to that of pure Pd metal. This indicates that the amorphous matrix is strongly Stoner enhanced, and this $\text{Ni}_{81.5}\text{B}_{18.5}$ alloy composition is already close to the PM-FM transition.

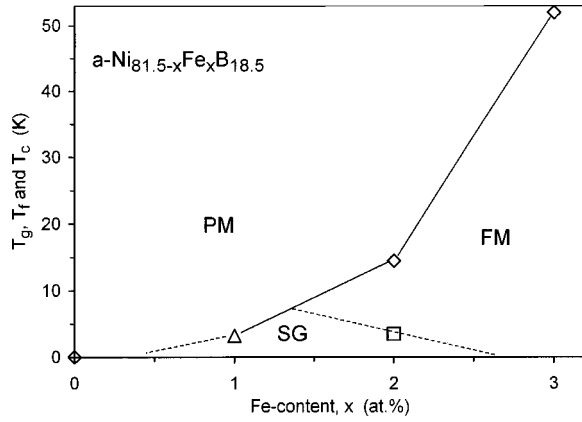


FIG. 8. Magnetic phase diagram of amorphous $\text{Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ alloys up to $x=3$ [T_g (Δ), T_f (\square), T_c (\diamond)]. Due to the few data points, it can only be estimated that the onset of the SG and FM regimes occurs around $x=0.5$ and 2.5 , respectively, as indicated by the dashed lines.

In a forthcoming paper,²⁵ it will be shown that the magnetization isotherms of $\text{Ni}_{81.5}\text{B}_{18.5}$ alloys can be ascribed to superparamagnetic particles, similarly to amorphous Ni-P alloys just around the critical concentration of the PM-FM transition.²¹

B. Magnetic phase diagram of the amorphous $\text{Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ system

In Fig. 8, the magnetic phase diagram for the amorphous $\text{Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ alloys is shown as constructed from the measured low-field magnetization data. No spin-glass state is observed for $x=0$ and 3 , whereas this state exists for the compositions in between ($x=1$ and 2). The binary amorphous alloy $\text{Ni}_{81.5}\text{B}_{18.5}$ and the ternary one diluted with 3 at. % Fe are paramagnetic and ferromagnetic down to the lowest temperatures measured, respectively. The compositions in between show a complex magnetic behavior with decreasing temperature: (i) the alloy with $x=1$ at. % Fe is a pure spin glass (SG) characterized by a magnetic transition from PM to SG at T_g and (ii) the alloy with $x=2$ at. % Fe is RSG characterized by two magnetic transitions: from PM to FM at T_c and from FM to SG at T_f . It is evident that the magnetic behavior of the amorphous $\text{Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ series ($0 \leq x \leq 3$) changes very rapidly with the Fe concentration, and this resembles the magnetic phase diagram of the amorphous $\text{Fe}_{100-y}\text{Zr}_y$ system for $7 \leq y \leq 12$ (Ref. 26). This resemblance is somewhat surprising since amorphous Fe-Zr is a Fe-rich alloy system where the rapid change of the magnetic properties takes place in a small Zr concentration range. On the other hand, comparing the magnetic phase diagram of

$a\text{-Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ alloys to that of $a\text{-Ni}_{80-x}\text{Fe}_x\text{P}_{20}$ alloys,^{27,28} it is evident that the spin-glass phase extends up to much higher Fe concentrations ($x_{\text{max}} \approx 10$ at. %) for the Ni-P-based ternary amorphous alloys than for the Ni-B-based ones studied in this paper ($x_{\text{max}} < 3$ at. %). This shows the crucial role of the type of metalloid in determining the low-temperature magnetic behavior in Ni-rich ternary Ni-Fe-metalloid amorphous alloys, and this will be the subject of a subsequent paper.

V. CONCLUSIONS

Detailed low-temperature magnetic measurements (magnetization isotherms, temperature dependence of the low-field magnetization, and ac susceptibility) were performed on melt-quenched amorphous $\text{Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ alloys with $x=1, 2$, and 3 . A pure spin-glass state was found for $x=1$ ($T_g=3.2$ K). For $x=2$, the spin-glass state ($T_f \approx 3.5$ K) transforms into a ferromagnetic phase which has a Curie temperature of $T_c=14.5$ K. The alloy with $x=3$ exhibits ferromagnetism with a Curie point of $T_c=52$ K. Based on these data, the magnetic phase diagram of amorphous $\text{Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ alloys could be constructed.

A comparison of the field and temperature dependence of the magnetic behavior of the amorphous $\text{Ni}_{81.5-x}\text{Fe}_x\text{B}_{18.5}$ ternary alloys with previous data on the amorphous $\text{Ni}_{81.5}\text{B}_{18.5}$ binary alloy supplied further evidence that the amorphous matrix of the binary alloy is a Pauli paramagnet and the observed FM-like behavior of the amorphous $\text{Ni}_{81.5}\text{B}_{18.5}$ alloy can be ascribed to Ni-rich segregations formed during the melt-quenching process owing to the low GFA of this particular alloy composition. Further detailed magnetic measurements, to be presented in a forthcoming paper,²⁵ on the amorphous $\text{Ni}_{81.5}\text{B}_{18.5}$ alloy demonstrate a superparamagnetic behavior of the Ni-rich segregations. This interpretation has been recently accepted also by Rojo *et al.*²⁹ based on the results obtained for a melt-quenched $a\text{-Ni}_{80}\text{B}_{20}$ alloy.

Finally, it should be pointed out that for inhomogeneous ferromagnets, the low-field FC and ZFC magnetization and the ac-susceptibility measurements clearly reveal that the Arrott-plot analysis overestimates the magnetic transition temperatures. In the present study, e.g., the Arrott-plot analysis yielded $T_c=9.5$ K for $x=1$, which alloy actually has a PM-SG transition only at $T=3.2$ K. This tendency is especially pronounced for weakly magnetic systems as was found to be the case also for amorphous Ni-P alloys around the critical concentration of the PM-FM transition.²¹

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