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Study of magnetic and hyperfine interactions in the quaternary amorphous alloy $Fe_{81}B_{13.5}Si_{3.5}C_2$

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Transmission ^{57}Fe Mössbauer spectroscopy has been used to investigate magnetic and hyperfine interactions in the quaternary metallic glass $Fe_{81}B_{13.5}Si_{3.5}C_2$ (Metglas® 2605SC) over a temperature interval 77—900 K. The Curie temperature T_C and the complete crystallization temperature are found to be 698±2 K and 835±2 K, respectively. The reduced average hyperfine magnetic field $H_{\text{eff}}(T)/H_{\text{eff}}(0)$ vs T/T_C shows a flatter curve in comparison with the Brillouin curve for $S=1$. This behavior is explained on the basis of Handrich's model of amorphous ferromagnetism in which the parameter δ, which is a measure of fluctuations in the exchange interactions, is assumed to have a temperature dependence $\delta=\delta_0(1-T^2/T_C^2)$, as proposed earlier by us, with $\delta_0=0.60$. The average hyperfine field $H_{\text{eff}}(T)$ could also be fitted to the expression $H_{\text{eff}}(T)=H_{\text{eff}}(0)\ [1-B_{3/2}(T/T_C)^{3/2}-C_{5/2}(T/T_C)^{5/2}-\cdots]$, with $H_{\text{eff}}(0)=290\pm5$ kOe, $B_{3/2}=0.29\pm0.02$ and $C_{5/2}=0.29\pm0.02$ in the range $0.11<T/T_C<0.92$, which indicates the presence of spin-wave excitations over rather large temperature interval. The distribution of a hyperfine-magnetic-field $P(H)$ curve at each temperature has a prominent and almost symmetrical peak. The full width at half maximum of the peak decreases monotonically with increase in temperature. Temperature dependence of $H_{\text{eff}}(T)$ has been further analyzed to obtain the critical exponent β, and is found to be 0.31±0.02 in the temperature range $0.11\leq T/T_C\leq 0.9$.

I. INTRODUCTION

There have been extensive efforts to experiment with different alloy compositions based on the basic iron-boron metallic glass system to understand the role of transition-metal and metalloid atoms in glass formation, and to understand their influence on the thermal stability and the magnetic properties of metallic glasses. Considerable studies have been reported on the binary and ternary glassy alloys such as Fe-B,[1] Fe-B-Si,[2,3] etc. Currently, there is an interest in the study of quaternary systems such as Fe-B-Si-C alloys.[4]

This paper reports the results of a study of the metallic glass alloy $Fe_{81}B_{13.5}Si_{3.5}C_2$ (Metglas® 2605SC) by ^{57}Fe Mössbauer spectroscopy up to its crystallization temperature with an emphasis on the hyperfine magnetic interactions. A preliminary report of this work has already been published previously.[5,6] A detailed account of the crystallization studies of metallic glass $Fe_{81}B_{13.5}Si_{3.5}C_2$ by Mössbauer spectroscopy, electrical resistivity, differential thermal analysis, and x-ray diffraction has recently been reported from this laboratory.[7] While this work was in its final stages, the Mössbauer study on the same alloy by Saegusa and Morrish, published recently, was brought to our attention.[8,9] Therefore, we discuss here the features such as temperature dependence of the parameter δ, which represents the fluctuations in the exchange interaction, hyperfine-field distributions in the Mössbauer spectra of amorphous $Fe_{81}B_{13.5}Si_{3.5}C_2$, critical exponent, etc., which are not discussed by Saegusa and Morrish. The Curie and crystallization temperatures of $Fe_{81}B_{13.5}Si_{3.5}C_2$ reported here are compared with those of Saegusa and Morrish. An effort is made to critically look at the variations in the analysis of hyperfine-field distribution by varying the number of terms in the Fourier series, and also by closely comparing the results in related crystalline systems with the amorphous material under investigation.

II. EXPERIMENTAL

Metglas 2605SC (nominal composition $Fe_{81}B_{13.5}Si_{3.5}C_2$) was supplied by the Allied Chemical Corporation (U.S.A.) in a ribbon form, 2.54 cm wide and ~30 μm thick. The investigations were carried out in the temperature range 77—900 K. Experimental details and methods of analysis of data can be found elsewhere.[4]

III. RESULTS AND DISCUSSION

A. Mössbauer spectra, magnetization axis, and linewidths

Mössbauer spectra of the "as-received" glassy alloy $Fe_{81}B_{13.5}Si_{3.5}C_2$ recorded in the temperature range 77—900 K are shown in Figs. 1 and 2. The six-line pattern, which is indicative of the ferromagnetic state of the sample is observed up to 640 K. The absorption lines are, as usual, characterized by rather large widths Γ (approximately 0.6—1.6 mm/s), in comparison with the linewidth (0.28 mm/s) in the Mössbauer spectrum of a thin iron foil taken under the same experimental conditions. At 675 K, the

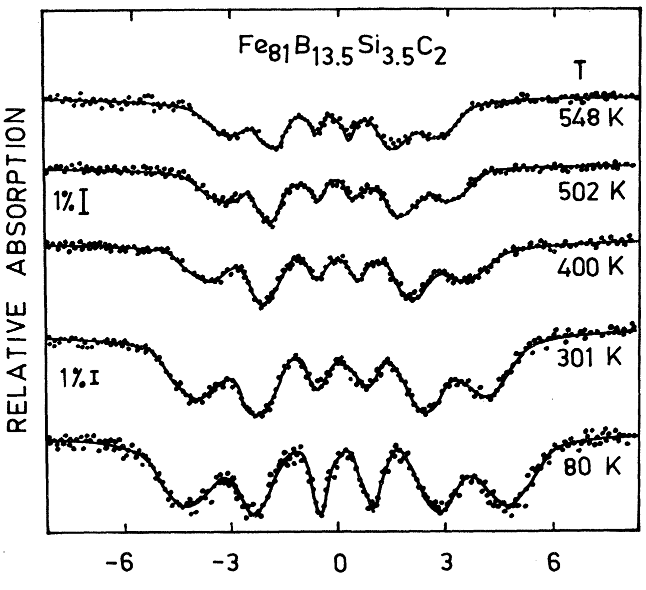

FIG. 1. Mössbauer spectra of amorphous $Fe_{81}B_{13.5}Si_{3.5}C_2$ at various temperatures below 550 K. Higher-temperature spectra are shown in Fig. 2.

six-line pattern collapses to a broad two-line pattern. This persistent line broadening indicates that the magnetic interactions are still present at 675 K. The spectrum at 700 K changes to a quadrupole-split paramagnetic doublet. The Curie temperature (T_C) of $Fe_{81}B_{13.5}Si_{3.5}C_2$ therefore lies between 675 and 700 K. At 750 K and above, the spectrum shows a complex profile due to crystallization of the glassy sample. Both Curie and crystallization temperatures have also been determined accurately by the thermal-scan method, as previously reported.[7] Analysis of Mössbauer data was performed as reported earlier.

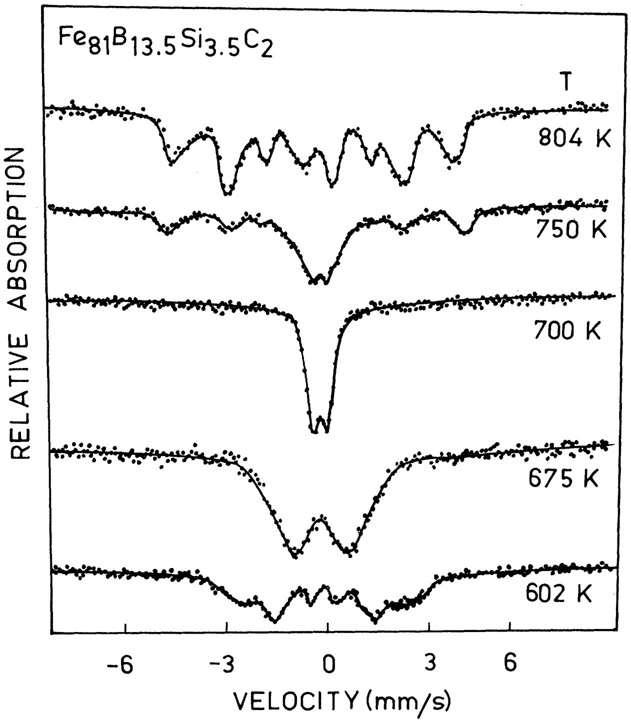

FIG. 2. Mössbauer spectra of $Fe_{81}B_{13.5}Si_{3.5}C_2$ at temperatures between 600 and 805 K. The sample is the same as in Fig. 1.

Large linewidths in six-line Mössbauer spectra of metallic glasses are usually explained by invoking the existence of a distribution of values of hyperfine magnetic fields, electric field gradients at the Mössbauer-probe atom, and isomer shifts which arise from the amorphous nature of solids. However, it is observed here that the broadening is not the same for all lines, and that the linewidth increases from the central to the outermost lines of the spectrum, i.e.,$\Gamma_{1,6} > \Gamma_{2,5} > \Gamma_{3,4}$. This shows that the major broadening is caused by the hyperfine-magnetic-field distribution, and the effect of the distribution of other hyperfine parameters is comparatively less. Furthermore, an asymmetry in linewidths as well as some asymmetry in line intensities are also observed. For example, at 301 K full widths at half maxima (FWHM) for the six-line pattern are

$$\Gamma_1:\Gamma_2:\Gamma_3:\Gamma_4:\Gamma_5:\Gamma_6::1.62:1.17:0.76:0.77:1.38:1.58 .$$

Note that $\Gamma_1 \approx \Gamma_6$, $\Gamma_2 < \Gamma_5$, and $\Gamma_3 \approx \Gamma_4$ in this case. Various explanations such as a linear relationship between the isomer shift δ_{IS} and the hyperfine magnetic field H_{eff},[10] and a linear relationship between H_{eff} and $(\delta_{IS}+\epsilon)$, where ϵ is a measure of quadrupole splitting,[11] have been proposed to explain this asymmetry. Le Caer and Dubois[12] have shown that it is possible to account for this asymmetry in the Mössbauer spectra by assuming the anisotropic hyperfine fields observed in related crystalline systems such as iron borides,[13,14] in addition to the variation of H_{eff} with δ_{IS}, the major contribution coming from the former. Information regarding the width of the hyperfine-magnetic-field distribution can be inferred from

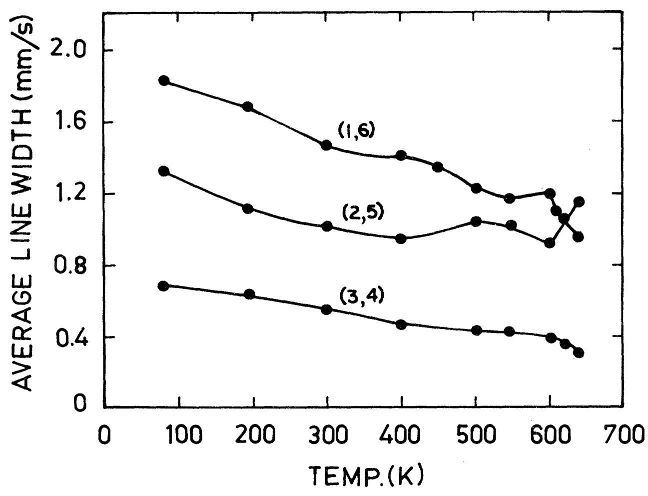

FIG. 3. Average linewidths of 1 and 6, 2 and 5, and 3 and 4 of amorphous $Fe_{81}B_{13.5}Si_{3.5}C_2$ as a function of temperature.

studying the temperature dependence of the widths of lines 1 and 6. Figure 3 shows the temperature dependence of FWHM of lines 1 and 6, 2 and 5, and 3 and 4. It is observed that $\Gamma_{1,6}$ decreases monotonically over the whole temperature range if the point $\Gamma_{1,6}$ (600 K) is ignored. This behavior indicates that the distribution in exchange interaction is narrow, as would be the case for long-range interactions, and therefore averages out the effect of the disordered structure.[15] The Curie temperature also is sharply defined for this alloy,[7,8] indicating that there are no local regions having a distribution of Curie temperatures. Therefore, the previous assumption, that the broadened Mössbauer spectra in an amorphous alloy is due to a significantly wide distribution of hyperfine magnetic fields, does not seem to be fully valid. Besides hyperfine-field distribution, other effects may also be important in the line broadening of amorphous alloys. Figure 3 also shows $\Gamma_{2,5}$ and $\Gamma_{3,4}$ as a function of temperature. Linewidth $\Gamma_{3,4}$ also shows a monotonic decrease with a rise in temperature. All these observations lead to the conclusion that the width of the distribution of hyperfine magnetic fields, whatever the width of the distribution, decreases with increasing temperature. This is further confirmed by our calculations of the probability distribution of hyperfine magnetic fields $P(H)$, discussed in a later section.

Mössbauer lines are observed to be symmetrical about the centroid, below T_C, and hence the quadrupole effect averages out to be zero in this amorphous alloy, as found in other metallic glasses. The effect of quadrupole interaction is seen only above T_C. The isomershift δ_{IS} of the Mössbauer spectrum at 300 K is found to be 0.15 mm/s relative to iron. δ_{IS} is found to be linearly dependent on temperature ($100 < T < 650$ K) with a coefficient approximately equal to -7×10^{-4} mm/s K.

B. Curie and crystallization temperatures

The Curie temperature (T_C) of $\mathrm{Fe_{81}B_{13.5}Si_{3.5}C_2}$ was deduced by two methods: (1) by plotting average hyperfine magnetic field versus absolute temperature, and (2) by the thermal-scan method. Average hyperfine magnetic fields $H_{\text{eff}}(T)$ were calculated from the least-squares-fitted line positions of the outermost lines in the six-line Mössbauer spectra. From a plot of $H_{\text{eff}}(T)$ vs T, values of $H_{\text{eff}}(0)$ and T_C are obtained by extrapolation. These values are found to be $H_{\text{eff}}(0)=290\pm5$ kOe and $T_C=690\pm10$ K. A large uncertainty in T_C in this method is due to a lower number of points near T_C. A more accurate determination was accomplished using the thermal-scan method as reported earlier (at a heating rate of 3 K/min), and was found to be 698 ± 2 K. The thermal-scan method also showed that the crystallization is complete at approximately 835 K. However, it is not easy to infer the onset of crystallization by this method, but Mössbauer spectra taken at 700 and 750 K, as shown in Fig. 2, clearly show that the onset of crystallization takes place before 750 K. Our recently reported electrical-resistivity measurements and differential thermal analysis also corroborate the onset of crystallization taking place at ~750 K.[7] Saegusa and Morrish[8] have recently reported a Curie temperature of 668 ± 2 K for this alloy, which is almost 30 K less than our value. Similarly, they have observed the onset of the crystallization of this Metglas at 655 K, which is also much less than ours, and is also less than their magnetic ordering temperature $T_C=668\pm2$ K. The temperature T_x at which fast crystallization sets in for our sample was found to be approximately 810 K (heating rate 3 K/min), and the temperature at which fast crystallization is completed is ~835 K.[7] On the other hand, Saegusa and Morrish have reported the crystallization temperature for Metglas 2605SC to be 788 ± 2 K (heating rate 5 K/min), and the crystallization is completed at approximately 800 K. Thus a difference of approximately 30 K in T_C and 20 K in T_x is observed between our measurements and those of Saegusa and Morrish,[8] their T_C and T_x having lower values.

This rather large difference in observed T_C and T_x by us and Saegusa and Morrish prompted us to repeat the thermal scan measurements and to see whether our furnace was properly calibrated. The repeated measurements on the samples taken from two different batches of the ribbon also gave us the same T_C and T_x. Our earlier measurements of T_x by the electrical resistivity and the differential thermal analysis also gave the same value of T_x within experimental accuracy as found by Mössbauer measurements, dispelling any doubt about the Mössbauer furnace calibration. In order to further investigate the calibration problem of the furnace, we ran other samples whose T_C were known, and we found that the results were reproducible within ±5 K. Thus the difference in values of T_C and T_x reported by us[7] and Saegusa and Morrish[8] must have some other origin such as a difference in the nominal composition of Metglas 2605SC. Other possibilities, such as differences in the heating rate, clamped, and unclamped sample, may be ruled out due to the large difference in the observed Curie temperature and crystallization temperature.

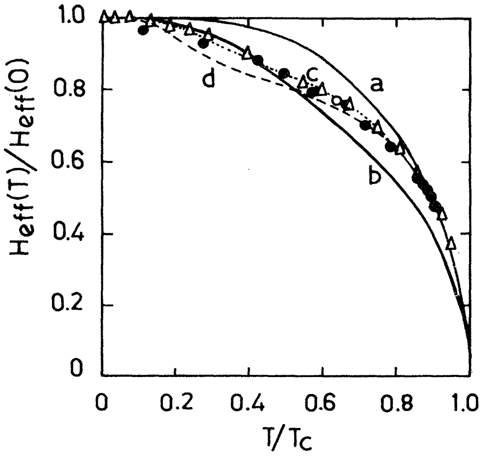

FIG. 4. RHMF vs reduced temperature of amorphous $\mathrm{Fe_{81}B_{13.5}Si_{3.5}C_2}$ for spin $S=1$. Closed circles represent our experimental points and the triangles are the experimental data of Ref. 8. The results obtained from Eq. (1) are shown by the following curves: a, for $\delta=0$; b, for $\delta=0.5$; c, for $\delta=0.6(1-t^2)$; and d, for $\delta=0.7(1-t^2)$ with $t=T/T_C$.

C. Temperature dependence of hyperfine magnetic field $H_{\rm eff}$

Reduced hyperfine magnetic field (RHMF) $H_{\rm eff}(T)/H_{\rm eff}(0)$, vs reduced temperature T/T_C is plotted in Fig. 4. Figure 4 also shows the Brillouin curve for spin $S=1$ along with the data of Saegusa and Morrish.[8] It is seen that the experimental data lie below the Brillouin curve as found for many other iron-rich metallic glasses[2,8,16–21] and other amorphous magnetic systems.[22–24] This departure of experimental data from the Brillouin curve has usually been explained, at least qualitatively, on the basis of fluctuations in the exchange interaction arising from the highly disordered atomic arrangement in glasses. Handrich[25] and Handrich and Kobe[26,27] developed a model within the molecular-field approximation which tries to explain this observation. Assuming that all atoms have the same spin S and the same Landé factor g, and taking into account of the spatial fluctuations of the exchange interactions, they were able to obtain an analytical expression for the reduced magnetization given by

$$\frac{M_s(T)}{M_s(0)}=\tfrac{1}{2}B_s((1+\delta)x)+\tfrac{1}{2}B_s((1-\delta)x)\ , \qquad (1)$$

where B_s is the Brillouin function for spin S,

$$x=\frac{3S}{S+1}\,\frac{M_s(T)T_C}{M_s(0)T}\ ,$$

and

$$\delta=\frac{\langle(\Delta J)^2\rangle^{1/2}}{\langle J\rangle}$$

is the square root of the fractional mean-square fluctuation in the exchange interaction J_{ij} between the ith and jth atoms. The value of δ lies between 0 and 1. Equation (1) does give a faster decrease of the RHMF with T/T_C if $\delta\neq 0$. However, it has been observed that even with $\delta=0.5$ or higher, there is a serious quantitative disagreement between experimental data and the Handrich-Kobe model in the low- and high-temperature regions. In order to improve upon this disagreement we have, in our earlier works, assumed δ to be a temperature-dependent parameter.[2,6,18–20] An empirical relation $\delta=\delta_0(1-t^2)$, where $t=T/T_C$, when substituted in Eq. (1) was found to give a good quantitative agreement between experimental data on iron-rich metallic glasses and Eq. (1) for values of δ_0 lying between 0.6 and 0.7.[21] A similar fitting procedure for data on $Fe_{81}B_{13.5}Si_{3.5}C_2$ also gives a good fit for $\delta_0=0.6$ and $S=1$, as shown in Fig. 4. Data points of Saegusa and Morrish which show good agreement with our data have also been shown in Fig. 4. The choice of $S=\frac{1}{2}$ or 1 does not make an appreciable difference, but we have noticed previously[19] that in some metallic glasses the agreement between experimental data and Eq. (1) can be noticeably improved by choosing either $S=\frac{1}{2}$ or 1. We believe that this may reflect the state of bonding between constituent magnetic atoms as influenced by the metalloid atoms which play an important role in the formation of glassy solids.

It is worth noting here, as pointed out by Schurer and Morrish,[15] that the comparison between amorphous magnetic alloys and crystalline elemental ferromagnets may be superfluous. This is brought out when a comparison is made between alloys having exactly the same composition in the amorphous and crystalline states, e.g., $(Fe_{50}Ni_{50})_{75}B_{25}$,[28] the disagreement between the data for the amorphous and crystalline alloys reduces considerably, although the data for the amorphous alloy still lie below that of the crystalline alloy. This observation indicates that the structural disorder does not have any appreciable effect on $M_s(T)/M_s(0)$ or $H_{\rm eff}(T)/H_{\rm eff}(0)$-vs-$T/T_C$ behavior. Therefore, one may conclude that it must be metalloids which are responsible for the observed behavior. However, similar behavior has also been reported earlier for alloys such as Fe_3Al (Ref. 29) and Fe_3Si,[30] since Al is a metal, and the suggestion of metalloids being mainly responsible for the flattening of the $H_{\rm eff}(T)/H_{\rm eff}(0)$ vs T/T_C does not hold. Therefore, it must be the presence of nonmagnetic atoms and local compositional and chemical disorder which must be responsible for large fluctuations in the exchange interactions besides some contribution from the spatial disorder in the system. Consequently, δ must represent the effect of overall disorder in the system. While some disorder may show hardly any temperature dependence, some other type of disorder may be more temperature dependent, giving rise to the temperature-dependent δ. In Sec. III A it has been said that the monotonic decrease of the linewidth $\Gamma_{1,6}$ with an increase in temperature leads to the conclusion that the distribution of hyperfine magnetic fields $P(H)$, hence that of exchange interactions, must be narrow enough to label the interactions as "long-range" type. This seems consistent with the observation that the data $H_{\rm eff}(T)/H_{\rm eff}(0)$ vs T/T_C is not much different for amorphous and crystalline $(Fe_{0.5}Ni_{0.5})_{75}Be_{25}$.[28] In our assumption, $\delta=\delta_0(1-t^2)$ reduces as $t\rightarrow 1$, and therefore should lead to a narrowing of $\Gamma_{1,6}$ and a sharpening of $P(H)$ curves as the temperature of the sample is increased. Both of these behaviors have been observed for amorphous alloy $Fe_{81}B_{13.5}Si_{3.5}C_2$. In any case, the good fit obtained between experimental data and Eq. (1), assuming $\delta=\delta_0(1-t^2)$ for a large number of iron-rich metallic glasses, cannot be ignored. Theoretically, one has to look for the origin of the suggested temperature dependence of δ.

D. Hyperfine-magnetic-field distribution

The hyperfine-magnetic-field distribution, the proposed major cause of the broadening of Mössbauer spectral line, arises from a large number of inequivalent iron sites in the glassy sample. While the temperature dependence of the linewidth $\Gamma_{1,6}$ implies the presence of long-range magnetic interactions in $Fe_{81}B_{13.5}Si_{3.5}C_2$, and therefore a narrow distribution of hyperfine magnetic fields, it is still worth looking into this distribution by independent means.

We have used the Fourier-series method developed by Window.[31] In this method it is assumed that the observed line shapes are reproduced by a continuous probability $P(H)$ for the distribution of hyperfine magnetic fields. The $P(H)$ distribution is assumed to be given by

$$P(H)=\sum_{n=1}^{M}a_n\left[\cos\left[\frac{n\pi H}{H_0}\right]-(-1)^n\right]\ , \qquad (2)$$

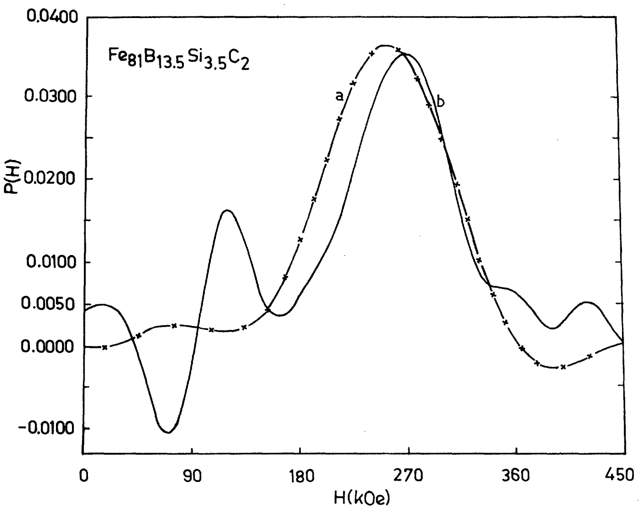

FIG. 5. Hyperfine-field distribution $P(H)$ of amorphous $Fe_{81}B_{13.5}Si_{3.5}C_2$ at 301 K; *a*, for $M=6$; *b*, for $M=15$.

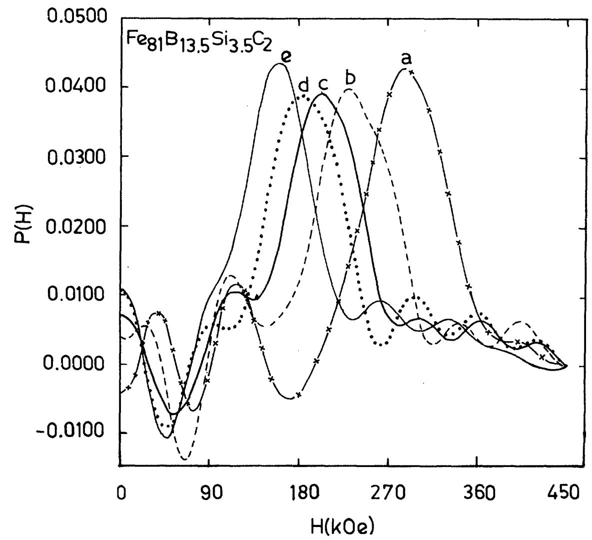

FIG. 6. Hyperfine-field distribution $P(H)$ of amorphous $Fe_{81}B_{13.5}Si_{3.5}C_2$ at *a*, 80 K; *b*, 400 K; *c*, 502 K; *d*, 548 K; and *e*, 602 K.

where H_0 is the maximum value of H when $P(H_0)=0$, and in our case it was 450 kOe, well above the H_{eff} value for $\alpha-$Fe at room temperature. Details of the procedure can be found in the paper by Narayanasamy *et al.*[32] The smoothness of the desired solution of $P(H)$ is expected to depend on "M," the number of terms used in the summation. Furthermore, a single isomer shift and the absence of quadrupole interactions have been assumed to simplify the computation. The line-intensity ratio is varied to get the minimum χ^2 value in the computer fit for a given M value. Figure 5 shows the results of $P(H)$ analysis of the Mössbauer spectrum of $Fe_{81}B_{13.5}Si_{3.5}C_2$ at 301 K when $M=6$ and 15 are used in Eq. (2). In both cases minimum χ^2 was found for the line-intensity ratio 3:2.3:1:1:2.3:3, which were 2.44 and 1.10, respectively. The $P(H)$ curve shows a nice smooth peak for $M=6$ centered around 252 kOe and FWHM equal to 121 kOe, with hardly any oscillations at either side of the peak. While in the case of $M=15$, the $P(H)$ curve shows a major peak centered around 270 kOe with FWHM of 92 kOe, with large oscillations at low values of H and a minor peak at high values of H. Oscillations in $P(H)$ around zero are the result of the truncation of the Fourier series, and therefore one need not attach much significance to it, although in this particular case these seem to be much larger than those previously observed.[33] The average hyperfine magnetic fields calculated from the $P(H)$ distributions are 235 kOe for $M=6$ and 253 kOe for $M=15$, while the measured H_{eff} is 258 kOe. For $M > 15$, the $P(H)$ curve starts showing unphysical structures. Therefore, we have analyzed the rest of the data on $Fe_{81}B_{13.5}Si_{3.5}C_2$ using $M=15$, which corresponds to the minimum χ^2. The lines drawn through the data points in Figs. 1 and 2 (up to $T=602$ K) are the theoretically simulated spectra using the $P(H)$-analysis results. Agreement seems to be reasonably good. The results of the $P(H)$ analysis carried out for the spectra between 80 and 602 K are shown in Fig. 6. All the distribution curves are found to have a pronounced maximum which shifts to lower fields with increase in temperature. Large oscillations are seen at low values of H and some minor peaks at high values of H. Looking at the low-field side of $P(H)$, a minor peak also seems to be present at ~100 kOe, which shows only a weak temperature dependence. But the shape of the major peak in $P(H)$, with increase in temperature, essentially remains unchanged.

The parameters which are of interest from this analysis are H_p, the value of H at which the maximum of the ma-

TABLE I. Most probable field, mean field, and FWHM of amorphous $Fe_{81}B_{13.5}Si_{3.5}C_2$ (Metglas 2605SC).

Temperature (K)	Mean field $\bar{H}$ (kOe)	Average magnetic hyperfine field H_{eff} (kOe)	Most probable field H_p (kOe)	FWHM of $P(H)$ distribution ΔH (kOe)
80	276.0	281.8	285.3	92.7
301	252.9	257.8	270.0	91.8
400	240.2	228.8	228.6	90.0
502	219.1	201.8	201.6	86.4
548	212.0	185.1	184.5	84.6
602	185.0	157.1	157.5	79.2

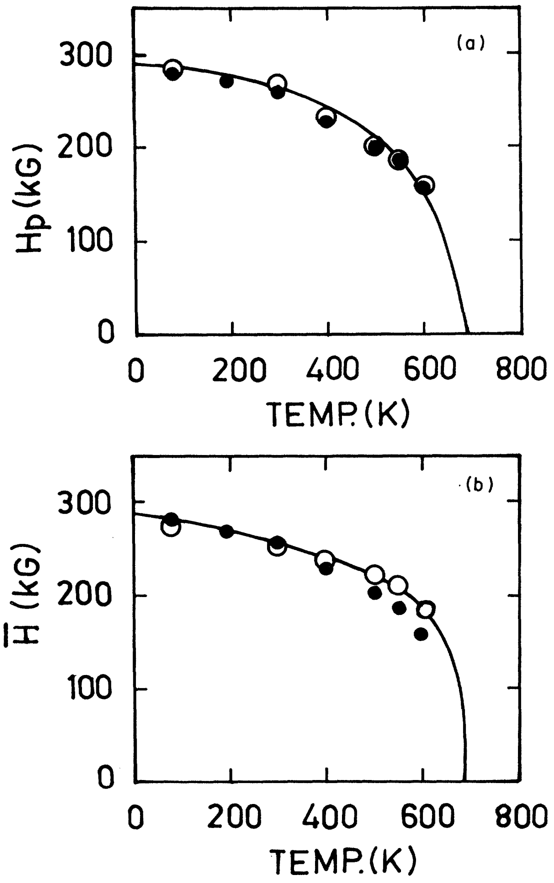

FIG. 7. (a) Temperature dependence of the most probable field of $P(H)$ for amorphous $Fe_{81}B_{13.5}Si_{3.5}C_2$, where open circles represent the most probable field and closed circles represent the average hyperfine magnetic fields taken from the least-squares fit of Mössbauer data of the sample. (b) Temperature dependence of mean field of $P(H)$ for amorphous $Fe_{81}B_{13.5}Si_{3.5}C_2$, where the circles represent the mean field and the closed circles represent the average hyperfine magnetic fields calculated from the least-squares fit of Mössbauer data of the sample.

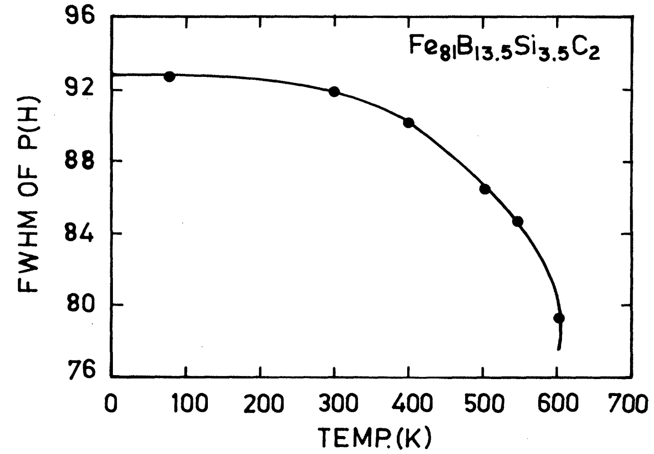

FIG. 8. Temperature dependence of the FWHM of $P(H)$ for amorphous $Fe_{81}B_{13.5}Si_{3.5}C_2$.

jor peak occurs, the FWHM of the major peak ΔH, and the mean field $\bar{H}$. Thus the mean field $\bar{H}$, the effective hyperfine magnetic field $H_{\text{eff}}(T)$, the probable field H_p, and the FWHM of $P(H)$ analysis ΔH, for different temperatures, are tabulated in Table I. The temperature dependences of H_p and $\bar{H}$ are shown in Fig. 7 along with the measured $H_{\text{eff}}(T)$. Figure 8 shows the temperature dependence of ΔH, i.e., FWHM of $P(H)$, which shows a monotonic decrease of the ΔH with temperature, implying that the distribution of hyperfine magnetic fields is becoming narrower. This is consistent with our results on the temperature dependence of $\Gamma_{1,6}$ and δ.

To get a better insight in this analysis, we also deconvoluted the thin-iron-foil Mössbauer spectrum at room temperature to obtain the corresponding $P(H)$ curve. The width of the outermost Mössbauer lines was 0.30±0.02 mm/s. The $P(H)$ curve gave a sharp peak at $H=335$ kOe with a FWHM of 34 kOe with small oscillations on either side of the main peak. Comparing this with the FWHM of $Fe_{81}B_{13.5}Si_{3.5}C_2$ at 301 K, which is approximately 92 kOe, we find that the distribution of hyperfine magnetic fields is approximately 3 times that of crystalline iron foil. It is probably narrow enough to give observed results on $\Gamma_{1,6}$, δ, and ΔH, but still large enough to broaden the Mössbauer spectral lines.

E. Spin-wave excitations

Data for $H_{\text{eff}}(T)$ vs T is fitted to the expression

$$\frac{\Delta H_{\text{eff}}(T)}{H_{\text{eff}}(0)} \equiv \frac{H_{\text{eff}}(T)-H_{\text{eff}}(0)}{H_{\text{eff}}(0)} = -BT^{3/2}-CT^{5/2}-\cdots = -B_{3/2}(T/T_C)^{3/2}-C_{5/2}(T/T_C)^{5/2}-\cdots \,, \tag{3}$$

which is Bloch's law. The proportionality $\Delta H_{\text{eff}} \propto (T/T_C)^{3/2}$ is derived from the long-wavelength spin-wave excitations and is valid for $T/T_C \ll 1$.[34,35] The plot of $\Delta H_{\text{eff}}(T)/H_{\text{eff}}(0)$ vs $(T/T_C)^{3/2}$ is shown in Fig. 9. We find that for $T/T_C \leq 0.86$, the fitted values of B ($B_{3/2}$) and C ($C_{5/2}$) are 15.9×10^{-6} $K^{-3/2}$ (0.29) and 2.3×10^{-8} $K^{-5/2}$ (0.29), respectively, with $H_{\text{eff}}(0)$ $=290\pm5$ kOe. These values favorably compare with the $B_{3/2}$ and $C_{5/2}$ values reported by Saegusa and Morrish,[8] which are 0.30±0.05 and 0.29±0.05, respectively. It is found that the values of B ($B_{3/2}$) and C ($C_{5/2}$) depend upon the temperature interval used. This is expected, since at higher temperatures, nonlinearity from $T^{3/2}$ is expected to increase, leading to the larger values of C ($C_{5/2}$). The coefficients $B_{3/2}$ and $C_{5/2}$ are consistently 2—3 times higher for amorphous ferromagnets than for the same coefficients for iron.[15,33] It has also been noted that B and C values for alloys of approximately similar compositions, one amorphous and another crystalline, are different.[32] A crystalline alloy has smaller B and C values, similar to

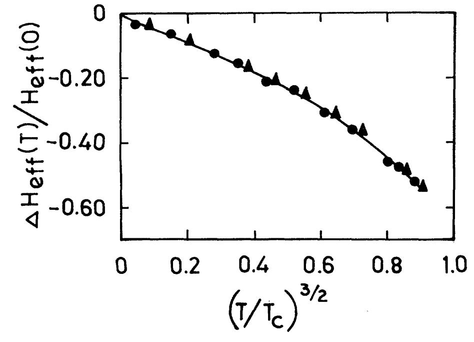

FIG. 9. Hyperfine-field change as a function of temperature in reduced units. Here closed circles represent the experimental data of amorphous $Fe_{81}B_{13.5}Si_{3.5}C_2$ and triangles represent the experimental data of Ref. 8. Also, $\Delta H_{eff}(T)=H_{eff}(T)-H_{eff}(0)$.

TABLE II. Values of the critical exponents D and β with temperature intervals $0<T/T_C\leq t$.

t	D (±0.03)	β (±0.02)
0.788	1.05	0.31
0.785	1.06	0.31
0.719	1.07	0.31
0.645	1.12	0.32
0.573	1.09	0.32
0.501	1.12	0.32
0.431	1.14	0.32
0.430	1.13	0.32
0.281	1.07	0.31
0.115	1.06	0.31

those of Fe. Thus, larger values of B ($B_{3/2}$) and C ($C_{5/2}$) seem to be the characteristic feature of the amorphous state.

In a preceding section it was mentioned that the flatter curve of $H_{eff}(T)/H_{eff}(0)$ vs (T/T_c) for amorphous ferromagnets does not seem to be due to the structural disorder, as a crystalline alloy of similar composition shows approximately the same behavior. If one looks at the data of Vincze *et al.*[28] on $(Fe_{50}Ni_{50})_{75}B_{25}$, one finds that there is a strong overlap of data $H_{eff}(T)/H_{eff}(0)$ vs T/T_C for amorphous and crystalline alloys, and therefore one would expect that B ($B_{3/2}$) and C ($C_{5/2}$) values for this alloy, in crystalline as well as amorphous form, should come out to be nearly equal for $T/T_C\leq0.4$. This seems to be in contradiction with the observation on the Fe-B system.[36] It is also hard to reconcile the fact that the disorder structure of amorphous ferromagnets influences one magnetic property (spin-wave-stiffness coefficients) quite significantly, while the other magnetic property (average hyperfine field) remains essentially unaffected.

F. Critical exponent β.

It is possible to obtain the value of the critical exponent β from the temperature dependence of H_{eff}. Since $M_s(T)\propto H_{eff}(T)$, near T_C the following relation will hold:

$$\frac{H_{eff}(T)}{H_{eff}(0)}=D\left[1-\frac{T}{T_C}\right]^{\beta}, \tag{4}$$

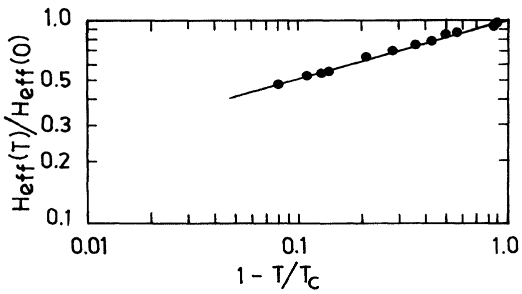

FIG. 10. $[H_{eff}(T)/H_{eff}(0)]$ vs $(1-T/T_C)$ for amorphous $Fe_{81}B_{13.5}Si_{3.5}C_2$ in log-log plot.

where β is one of the critical exponents and D is a constant close to 1.0. The variation of reduced hyperfine fields of $Fe_{81}B_{13.5}Si_{3.5}C_2$ with $1-T/T_C$ is shown in Fig. 10 on a log-log plot, which shows that this power law is followed by the $Fe_{81}B_{13.5}Si_{3.5}C_2$ amorphous alloy not only close to T_C, but over an extended temperature interval. A least-squares fit of Eq. (4) to experimental points for $1.0>T/T_C>0.70$ gives $\beta=0.31\pm0.02$ and $D=1.07\pm0.03$. These values are in good agreement with the β values of other metallic glasses.[19,35] The values of β and D calculated from Eq. (4) for different temperature regions are shown in Table II. It is interesting to note that even for $1.0>T/T_C>0.2$, β and D values are 0.31 and 1.07, which are not very different from the values for $1.0>T/T_C>0.70$. By comparison with the theory, a three-dimensional Heisenberg ferromagnet is expected to give $\beta=0.33$, which is in quite good agreement with the β value obtained here for amorphous $Fe_{81}B_{13.5}Si_{3.5}C_2$. It seems surprising that this power law should be obeyed over such an extended temperature interval, which may possibly arise due to the glassy nature of samples.

IV. CONCLUSIONS

The Mössbauer spectra measured between 77 and 900 K show that the Curie temperature T_C and the complete crystallization temperature are found to be 698±2 and 835±2 K, respectively. These values are higher than those reported by Saegusa and Morrish. $H_{eff}(T)/H_{eff}(0)$ is observed to decrease much faster with an increase in T/T_C when compared with the Brillouin curve for $S=1$. Agreement between $H_{eff}(T)/H_{eff}(0)$-vs-T/T_C data and Handrich's model of an amorphous ferromagnet, based on the mean-field—theory calculations, can be vastly improved by assuming the parameters $\delta=\langle(\Delta J)^2\rangle^{1/2}/\langle J\rangle$ in the theory to be temperature dependent, i.e., $\delta=\delta_0(1-T^2/T_C^2)$ with $\delta_0=0.6$. The variation of $H_{eff}(T)$ with T can also be described by

$$H_{eff}(T)=H_{eff}(0)[1-B_{3/2}(T/T_C)^{3/2} -C_{5/2}(T/T_C)^{5/2}-\cdots]$$

over the full temperature interval studied with coefficients $H_{\rm eff}(0)=290\pm5$ kOe, $B_{3/2}=0.29\pm0.02$, and $C_{5/2}=0.29\pm0.02$, while at low temperatures, i.e., $77<T<400$ K, $[H_{\rm eff}(T)-H_{\rm eff}(0)]/H_{\rm eff}(0)$ is proportional to $(T/T_C)^{3/2}$, which indicates the presence of long-wavelength spin-wave excitations in this amorphous alloy. At temperatures below T_C, $H_{\rm eff}(T)/H_{\rm eff}(0)$ can be expressed as $D(1-T/T_C)^\beta$ with $D=1.05\pm0.03$ and $\beta=0.31\pm0.02$. The observation of a well-defined $P(H)$ distribution monotonically narrowing with increasing temperature, the $T^{3/2}$ dependence of $H_{\rm eff}(T)$ at low temperatures, and the temperature dependence of the fluctuation parameter δ, in the Handrich model, suggest that the magnetic interactions in the amorphous $Fe_{81}B_{13.5}Si_{3.5}C_2$ are predominantly of long-range type.

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