

Comments

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Comment on the evaluation of hyperfine-field distributions in amorphous ferromagnets

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The iron hyperfine-field distribution in amorphous $\text{Fe}_{32}\text{Ni}_{36}\text{Cr}_{14}\text{P}_{12}\text{B}_6$ (METGLAS[®] 2826-A) at $T = 5$ K has been determined by Mössbauer spectroscopy. The obtained $P(H)$ is a very broad *single-maximum* function contrary to the recently reported *two maxima* distribution. The possible reason for this disagreement is discussed.

In a recent Mössbauer study of amorphous $\text{Fe}_{32}\text{Ni}_{36}\text{Cr}_{14}\text{P}_{12}\text{B}_6$ (METGLAS[®] 2826-A) Chien¹ has determined the iron hyperfine-field distribution $P(H)$ using the Fourier-series method developed by Window.² At 4.2 K a broad $P(H)$ was found which could be approximately described by a double-maxima function of H with a high-field component and a low-field component. These components were attributed¹ to Fe with mainly Fe and Ni and possibly some Cr atoms as neighbors and to Fe with a substantial number of Cr neighbors, respectively. This surprising result, which suggests the presence of microscopic *chemical inhomogeneities* in the amorphous system, motivated us to reinvestigate the $P(H)$ distribution. In the evaluation of $P(H)$ a different method recently proposed by Vincze³ was used. This method allows a more direct determination of the $P(H)$ because two independent Mössbauer spectra are used while usually the $P(H)$ is derived from a single spectrum.

It is well known that the relative intensities I of the individual six lines in the Mössbauer spectrum of ⁵⁷Fe are given by⁴

$$I_{1,6}:I_{2,5}:I_{3,4} = 3:b:1, \quad \text{with } b = \frac{4 \sin^2 \theta}{1 + \cos^2 \theta},$$

where θ is the angle between the γ -ray direction and the magnetic moment. b is determined by the distribution of magnetic-moment directions in the sample which in general is not known *a priori*. It is assumed in all methods used for the evaluation of $P(H)$ that b is independent of the hyperfine field, or more precisely that the average value of b is the same for each H . To our knowledge the validity of this assumption

was not investigated earlier. The *a priori* unknown b value may cause ambiguity in the determination of the $P(H)$ when the distribution is relatively broad and especially when it extends to very low fields. This ambiguity is not present in the method proposed by Vincze,³ which also provides an internal check for the earlier-mentioned assumption that b does not depend on H . This method requires that the magnetization directions in the sample and thus the value of b can be easily influenced by a small external field (~ 200 Oe). This is valid for most metallic glasses because of their soft magnetic properties.⁵

A linear combination of two spectra with different relative intensities of the lines 2 and 5 is taken in such a way that the 1,6 and 3,4 lines are removed. This can be done easily by adjusting the outer—free of overlap—part of the intensity of lines 1 and 6, which gives the coefficient for the linear combination. The two spectra of METGLAS[®] 2826-A at $T = 5$ K and the linear combination are shown in Figs. 1(a), 1(b), and 1(c). The spectra *a* and *b* were recorded in the same geometry (with the γ -ray direction perpendicular to the plane of the amorphous ribbons), in zero external field and in a small external field (~ 200 Oe) parallel to the ribbon plane, respectively. The low relative intensities of the second and fifth lines in zero external field [Fig. 1(a)] is caused by the stress⁶ between the amorphous ribbons and the adhesive tape. The small external field apparently turns the magnetic moments largely into the ribbon plane. The spectrum [Fig. 1(b)] can be described by an effective $\theta = 79^\circ$ (or $b = 3.7$) as the following analysis will show. It is worthwhile to emphasize that the only important assumption is that the average of the b values is the same at each

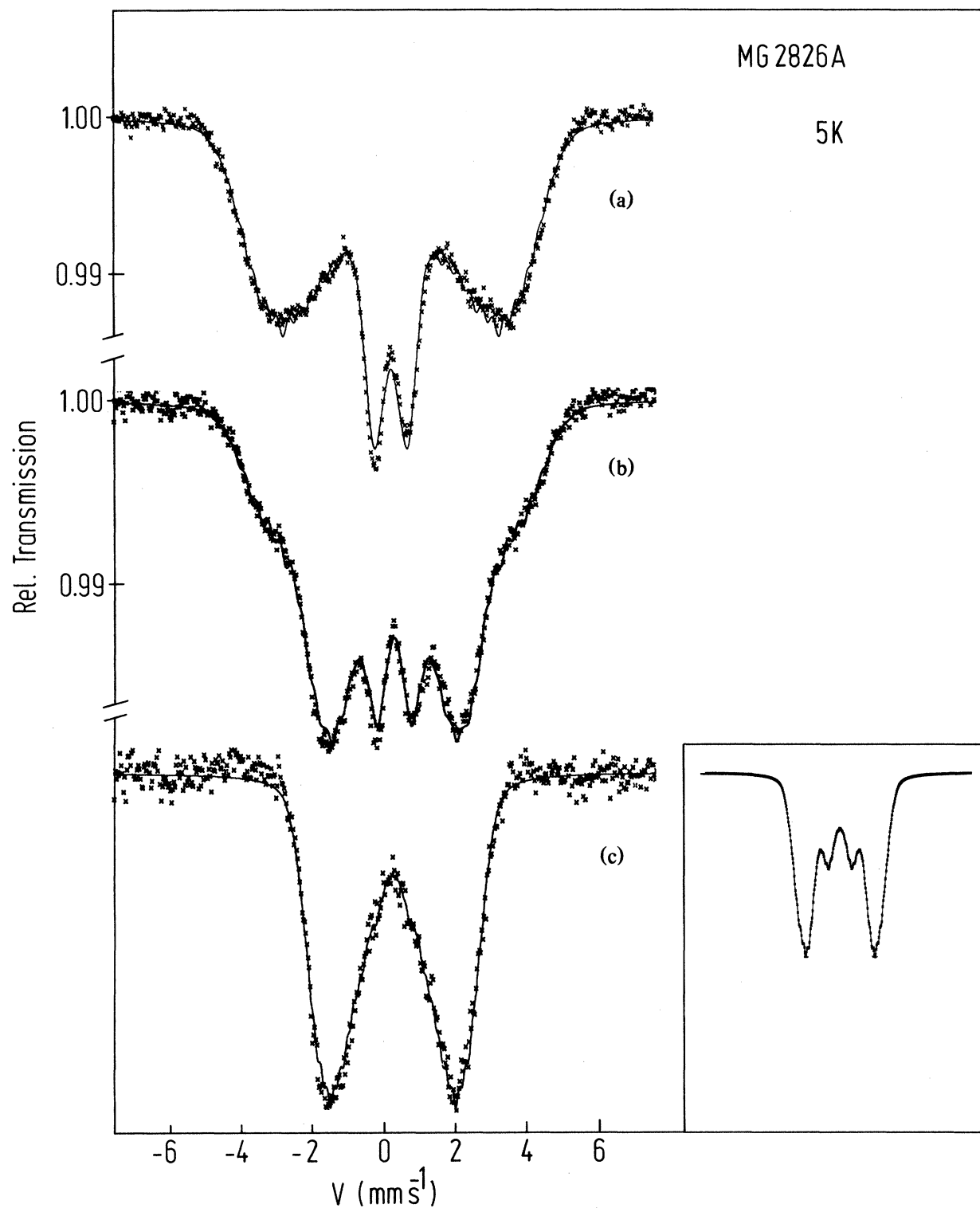


FIG. 1. Mössbauer spectra of amorphous $\text{Fe}_{32}\text{Ni}_{36}\text{Cr}_{14}\text{P}_{12}\text{B}_6$ at 5 K (a) in zero external field, (b) in a small external field (~ 200 Oe) parallel to the plane of the ribbons. (c) The lines 2 and 5 obtained from a linear combination of the spectra *a* and *b*. The continuous lines are the calculated curves based on the discrete $P(H)$ of Fig. 2. The insert shows the shape of the lines 2 and 5 calculated from the two-maxima $P(H)$ given in Fig. 7 of Ref. 1, where the oscillations at the high-field side due to the truncated Fourier series have been removed.

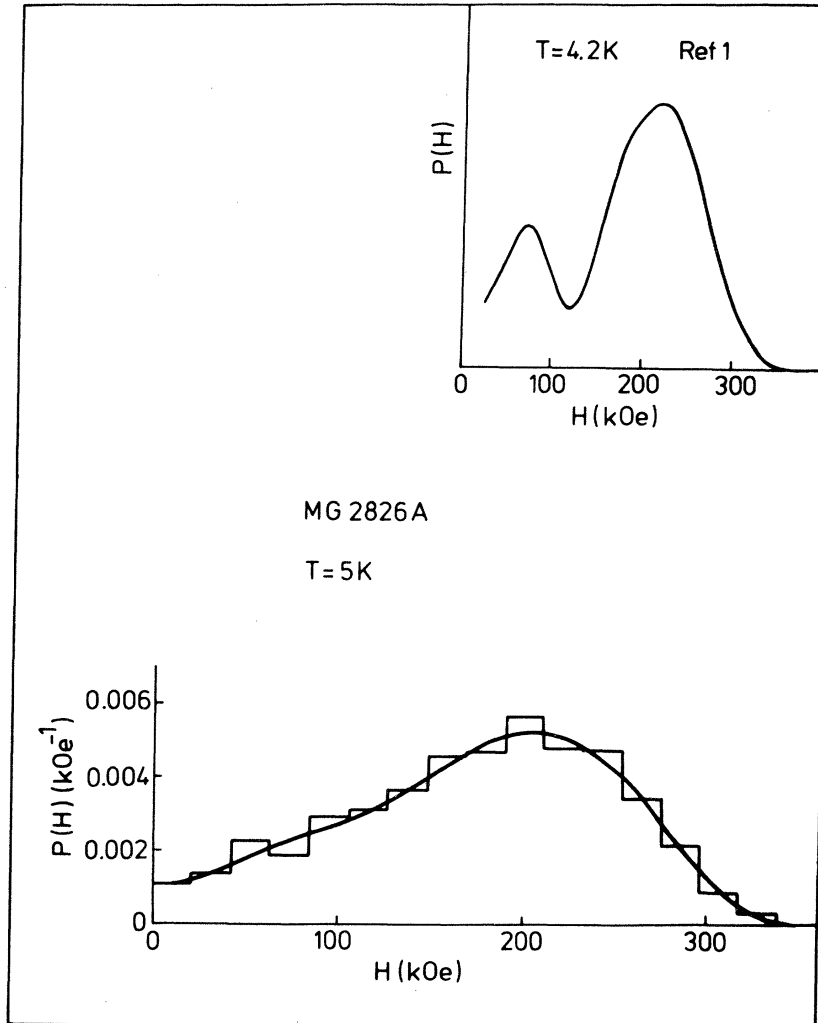


FIG. 2. Hyperfine-field distribution $P(H)$ of amorphous $\text{Fe}_{32}\text{Ni}_{36}\text{Cr}_{14}\text{P}_{12}\text{B}_6$ obtained by Vincze's method (Ref. 3) as explained in the text. The continuous line shows the $P(H)$ obtained by Window's method (Ref. 2) from lines 2 and 5 [Fig. 1(c)]. The insert shows the $P(H)$ obtained by Chien (Fig. 7 of Ref. 1).

hyperfine-field value and that complete alignment of the magnetic moments is not required. Moreover, this assumption can be checked in the evaluation process since the intensities of possible paramagnetic lines or lines due to nonaligned magnetic moments will be different in the original spectra and in the linear combination resulting in the separated 2 and 5 lines. From the lines 2 and 5 [Fig. 1(c)] we can conclude directly that the $P(H)$ is a smooth function of H extending to very low fields, and not the two-maxima function obtained by Chien¹ which would give the line shape shown in the insert of Fig. 1. We calculated a discrete $P(H)$ (Fig. 2) from the lines 2 and 5 [Fig. 1(c)] by fitting doublets with fixed H values at distances $\Delta H \approx 20$ kOe, and their relative intensity was determined (the linewidth of the Lorentzians was

0.29 mm s^{-1}). Furthermore, a single isomer-shift value was assumed. Using the $P(H)$ obtained, the original spectra were fitted with one free parameter: the relative intensity b of lines 2 and 5. Again a single isomer-shift value and zero quadrupole splitting were assumed. It has been shown by Vincze³ that the $P(H)$ distribution is not influenced by taking into account the isomer shift and quadrupole splitting distribution. The fits shown in Figs. 1(a) and 1(b) are good and the values of b are 0.1 ± 0.1 (spectrum *a*), 3.7 ± 0.1 (spectrum *b*). These values correspond to average magnetization directions described by the θ values of 13° and 79° , respectively.

The result that the original *two* spectra could be quite well described with the $P(H)$ obtained from the separated 2 and 5 lines shows that within the experi-

mental error the b values are independent of the hyperfine field which is the crucial assumption of the evaluation methods. This conclusion is further supported by the evaluation of $P(H)$ from the original two spectra by the Window method, which gives the same result for $P(H)$ in both cases, and identical to the one obtained from the 2,5 lines, when the above determined b values are used (continuous line in Fig. 2).

The resulting $P(H)$ (Fig. 2) is a rather smooth *single-maximum* function and extends to very low fields. For comparison the $P(H)$ obtained by Chien is shown in the insert of Fig. 2. In contrast with the latter one, our $P(H)$ agrees with a *random* distribution of Cr atoms, as we would expect for a *chemically homogeneous* amorphous alloy. The two maxima, or more precisely, the relative minimum between the peaks in the $P(H)$ derived by Chien¹ can be explained as the result of a too high value chosen for the parameter b . It is easy to verify that the minimum in this $P(H)$ (insert Fig. 2) corresponds to the separation of those 2 and 5 lines which belong to the 6 line patterns with the maximum probability at the high-field side. This illustrates clearly that the very basic problem of the $P(H)$ evaluation is that the value of b is generally unknown. As a consequence of the erroneous determination of $P(H)$ all conclusions of Ref. 1 concerning the interpretation of $P(H)$ are questionable. Similarly, the $P(H)$ distributions

of amorphous $(\text{Fe,Mo})_{75}\text{P}_{16}\text{B}_6\text{Al}_3$,⁷ and of amorphous Fe-Si alloys⁸ are very broad, extend to low field, and they do show the two-maxima feature discussed here and this again may be a result of the evaluation method.

For amorphous Fe-Ge alloys, which also show a broad $P(H)$ with long tail, it was shown⁹ that the two-maxima $P(H)$ obtained in a first analysis could be transformed into a smooth single-maximum function by choosing a lower value for the intensity ratio b .

The evaluation of broad $P(H)$ extending to low fields is very problematic. It has been shown that the method of Vincze³ is suitable for the analysis of such broad, overlapping spectra because of the use of overlap-free subspectra and its internal-control possibilities.

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