Sign of the Magnetic Hyperfine Field in Dilute Iron Alloys Using the Mössbauer Effect

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(Received 6 May 1964)

The sign of the hyperfine field in several dilute iron alloys has been determined. The technique makes use of the Mössbauer effect, which is used to establish the helicity of gamma rays emitted and absorbed in a longitudinal magnetic field. Experiments were performed which show that the field induced at the iron nucleus by the application of an external magnetic field H_0 is negative (i.e., opposite to H_0) in Mössbauer sources of Co^{57} in Cu and Au and in absorbers of Mo and Rh containing about 1% Fe^{57}. The spontaneous field H_{int} observed in a ½% Fe^{57}-in-Cr alloy below 50°K is positive.

I. INTRODUCTION

THE magnetic field H_{nuc}, responsible for the measured nuclear magnetic hyperfine interaction, may be described phenomenologically by a sum of components: the external applied field H_0, the spontaneous field H_{int}, present in the absence of H_0, and a field H_i, induced by H_0. The sign of H_i or H_{int} is defined to be positive or negative according to whether it is in the same or opposite direction to the applied field H_0. The sign of H_i may be inferred from a measurement of the sign and magnitude of H_{nuc}, which in turn can be determined by a number of techniques; among these being the precession direction of a nuclear gamma-ray correlation, the angular distribution of radiations from polarized nuclei, the scattering of polarized neutrons from polarized nuclei, and the effect of an external magnetic field on the Mössbauer hyperfine pattern. Using this last technique, we have determined the sign of the hyperfine field at the Fe^{57} nucleus in several dilute iron alloys.

The magnitude of the internal field H_{int}, acting on an iron nucleus in metallic iron, was first determined using Mössbauer techniques by Hanna *et al.*[1] to be 331 kOe. In a subsequent investigation[2] they showed that the sign of H_{int} was negative by demonstrating that the magnetic hyperfine interaction was diminished on applying an external magnetic field. The importance of this result to the understanding of the mechanisms responsible for ferromagnetism led to similar experiments[3] on other ferromagnetic materials. This technique is not applicable to the measurement of H_i. For example, consider an effective field described by

$$H_{nuc}=H_0+H_i=H_0[1+C(T)].$$

If, as is often the case, C is temperature-dependent and in particular, approaches zero at high temperature, then the measurement of the hyperfine spectrum as a function of both H_0 and T will uniquely determine the sign of C. If, however, C is not temperature-dependent, or if temperature-dependent experiments are impractical, then still another technique is necessary since the sign of C is ambiguous unless the sign of H_{nuc} is measured.

The sign of H_{nuc} is reflected in the sense of the circularly polarized hyperfine transition gamma rays (i.e., the gamma-ray helicity) emitted along the field direction.[4] The helicity of a hyperfine transition reverses with field reversal. The helicity of the radiation can be analyzed by using a longitudinally polarized absorber whose internal field direction is known. Frauenfelder *et al.*[5] have suggested that this fact can be used to measure the sign of an unknown internal field by analyzing the unknown field with a polarized Mössbauer source or absorber in which the sign of the internal field is known. We have used this method to determine the sign of the H_i in several iron alloys. The technique is discussed in the next section and the results in the final section. We consider in some detail examples in which the technique corroborates the sign inferred from temperature- and field-dependent measurements (Cu and Au) as well as an example in which temperature-dependent measurements are impractical and the circular polarization technique is necessary (Cr).

II. TECHNIQUE

For completeness, the decay scheme of Co^{57} and the magnetic hyperfine structure of the 14.4-keV transition is reproduced in Fig. 1. The relative intensities of the

* This paper is based on part of a dissertation submitted to the faculty of Branders University by Norman Blum in partial fulfillment of the requirements for the degree of Ph.D. in physics.

† Supported by the U. S. Air Force Office of Scientific Research.
‡ Supported in part by U. S. Atomic Energy Commission Contract AT(30-1)-2098.

[1] S. S. Hanna, J. Heberle, C. Littlejohn, G. J. Perlow, R. S. Preston, and D. H. Vincent, Phys. Rev. Letters 4, 177 (1960).

[2] S. S. Hanna, J. Heberle, G. J. Perlow, R. S. Preston, and D. H. Vincent, Phys. Rev. Letters 4, 513 (1960).

[3] See, for example, A. J. F. Boyle, D. St. P. Bunbury, and C. Edwards, Phys. Rev. Letters 5, 553 (1960); R. W. Grant, M. Kaplan, D. A. Keller, and D. A. Shirley, Phys. Rev. 133, A1062 (1964).

[4] L. Grodzins and F. Genovese, Phys. Rev. 121, 228 (1961).

[5] H. Frauenfelder, D. E. Nagle, R. D. Taylor, D. R. F. Cochran, and W. M. Visscher, Phys. Rev. 126, 1065 (1962).

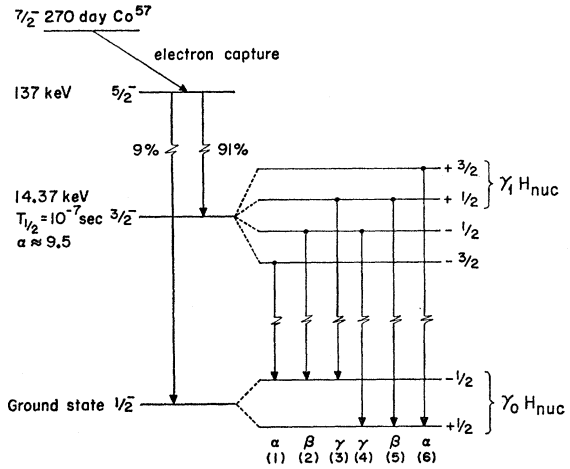

FIG. 1. The decay scheme of Co^{57} and the magnetic hyperfine structure of the 14.4-keV transition of Fe^{57}.

hyperfine lines α, β, γ are given by

$$\Delta m(\pm\tfrac{3}{2}\rightarrow\pm\tfrac{1}{2})=\tfrac{3}{2}(1+\cos^2\theta),$$
$$\Delta m(\pm\tfrac{1}{2}\rightarrow\pm\tfrac{1}{2})=2\sin^2\theta,$$
$$\Delta m(\pm\tfrac{1}{2}\rightarrow\mp\tfrac{1}{2})=\tfrac{1}{2}(1+\cos^2\theta).$$

If the source is unpolarized, averaging over θ gives the intensity ratios 3:2:1 for $\alpha:\beta:\gamma$. For a polarized source, the intensity ratios are 3:4:1 or 3:0:1 for magnetization perpendicular or parallel to the quantization axis, respectively. The Mössbauer pattern for an unpolarized absorber, in the absence of an applied field, versus a single line source is shown in Fig. 2(a). (The enhancement of the $\Delta m=0$ lines results from the predominance of domain magnetization in the plane of the foil.) Figure 2(b) shows the observed Mössbauer spectrum obtained with a longitudinally applied field of 86 kOe. The $\Delta m=0$ lines disappear; the contraction of the pattern indicating that $H_{\rm int}$ is negative is evident.[6]

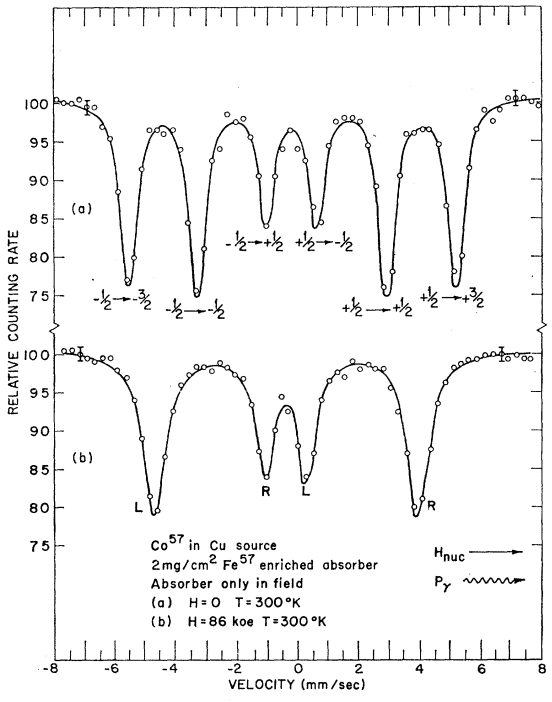

FIG. 2. Resonant absorption spectra of Fe^{57} in metallic iron, using a single-line source, showing hyperfine structure (a) in the absence of an external magnetic field and (b) in a longitudinal field of 86 kOe.

If both source and absorber exhibit hyperfine splittings due to colinear longitudinal fields, then the conservation of angular momentum requires that the hyperfine radiation emitted from the source be absorbed by the corresponding transition which conserves angular momentum; that is, the emitted gamma ray carrying ±1 unit of angular momentum can only be absorbed in a transition which can accept that ±1 unit of angular momentum.

Consider the hyperfine splittings of source and absorber in colinear internal fields, shown schematically in Figs. 3(a) and 3(b). In 3(a) the internal fields are parallel, in 3(b) they are antiparallel. As the source is swept over the absorber, an absorption spectrum of eight lines results, namely: *Ab*, *Ad*, *Ba*, *Bc*, *Cb*, *Cd*, *Da*, and *Dc* for antiparallel fields. For parallel fields the complementary pairings obtain, namely: *Aa*, *Ac*, *Bb*, *Bd*, *Ca*, *Cc*, *Db*, *Dd*. When the magnitudes of the fields are equal, degeneracy occurs. The resulting absorption spectra are sketched in Figs. 3(a) and 3(b). Figure 4 presents the experimental results for the parallel case. Both the source and absorber are iron foils in a longitudinal field of 108 kOe.

The striking differences in the spectra between the two cases are maintained in the general situation of fields of unequal magnitudes. Figure 5 is a nomograph showing the absorption positions as a function of H_1/H_2 from -1 to $+1$. The ordinate is scaled for $H_2=331$ kOe. The spectra for the parallel and antiparallel cases are distinguished both by the magnitude of the splittings and by the intensity ratios: the magnitude of the splittings is greater in the antiparallel case; the easily distinguished outer doublet has an intensity ratio of 3:1 in the antiparallel case and 1:3 or 1:1 in the parallel case. Thus, if the sign of the internal field of either the source or absorber is known, as it is for metallic iron, then the sign of the unknown field, as well as the magnitudes of both fields can be determined.

III. EXPERIMENTAL RESULTS

The results presented here on the sign of $H_{\rm nuc}$ and H_i were obtained in the course of investigations of the localized moment on the Fe^{57} atom in various metal

[6] The observed value of the contraction corresponds to 64 kOe, implying a demagnetization field of about 22 kOe. A series of careful measurements of the contraction of the hyperfine field in iron due to an external field indicates that the external field has the effect of slightly shifting the temperature axis on the Curie-Weiss magnetization curve; these results will be reported later.

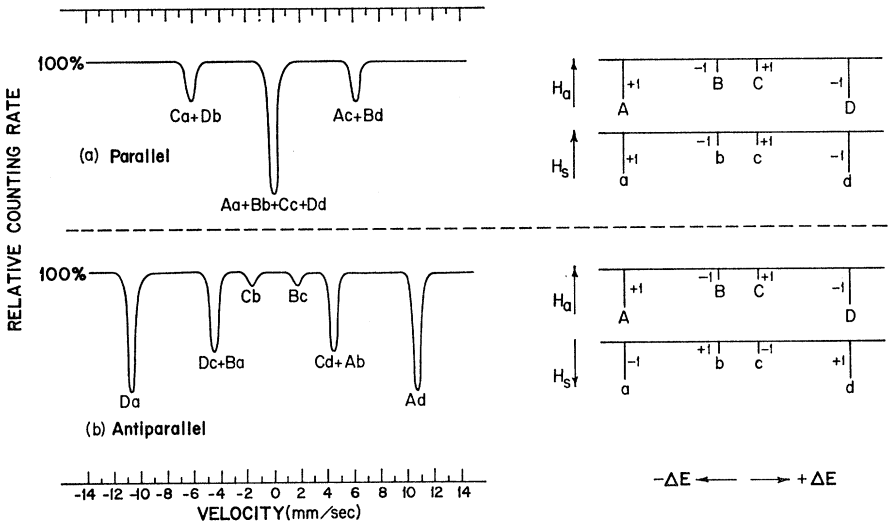

FIG. 3. Sketch of absorption patterns obtained with both source and absorber levels split by longitudinal hyperfine fields of the same magnitude which are (a) parallel and (b) antiparallel. On the right are the hyperfine lines of source and absorber with the helicity indicated by $+1$ or -1.

hosts.[7] The experiments utilized alloys containing small concentrations of Fe^{57} (Co^{57} in the case of sources). Three examples are described in detail: Co^{57} in copper, Co^{57} in gold, and $\frac{1}{2}\%$ Fe^{57} in chromium. The first of these is particularly simple, and the sign is easily inferred from the behavior of the hyperfine field with H_0 and T. The source in gold is a bit more complicated since H_{nuc} changes sign as a function of H_0/T. The direction of H_{nuc} is still determined, however, on the basis of a simple model to account for the induced field. In this case a direct independent determination of the sign simplifies and strengthens the analysis of the temperature dependence data and allows an immediate knowledge of the induced field from a measurement of the hyperfine splitting. Finally, the absorber in chromium illustrates the case where the sign of the field is not determined by measurements of the magnitude of the hyperfine field alone; here it is necessary to make a separate determination of the sense of the circular polarization of the individual hyperfine transitions.

Both source and absorber were placed inside a liquid-helium cryostat covering the temperature range from room temperature down to 1.2°K. The cryostat was suspended inside a Bitter solenoid so that the source and absorber were in the same magnetic field (to within $\sim 0.5\%$) which could be varied up to about 100 kOe. The counting equipment used a xenon-filled proportional detector because of its good resolution for 14-keV gamma rays and because of its insensitivity to the stray magnetic fields, together with a conventional nuclear pulse amplifier, a single-channel analyzer to select the 14-keV gamma ray, and a multichannel analyzer to store transmitted gamma-ray counts as a function of source velocity. The velocity drive was an electromechanical constant acceleration system similar

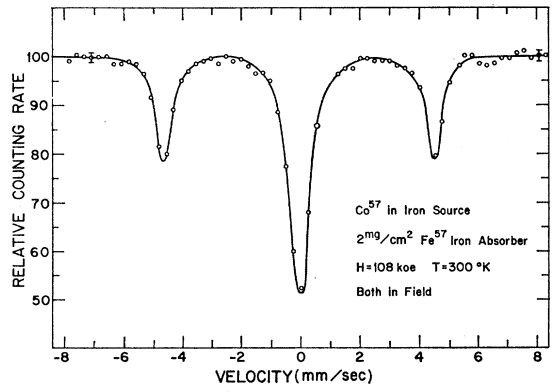

FIG. 4. Experimental absorption spectrum for a Co^{57} in iron source versus an Fe^{57} in iron absorber; both source and absorber are in a 108 kOe longitudinal field.

[7] N. Blum, A. J. Freeman, and L. Grodzins, Rev. Mod. Phys. 36, 406 (1964). Results of this work are being prepared for publication.

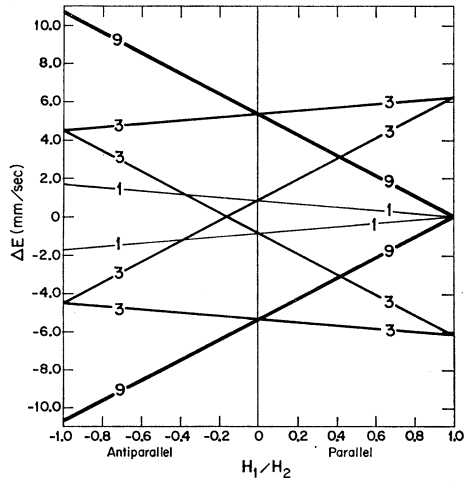

FIG. 5. Nomograph showing the positions of the lines of an absorption spectrum for colinear longitudinal fields as a function of the ratio of the hyperfine fields in source and absorber. The velocity axis is scaled for $H_2=331$ kOe.

TABLE I. Comparison of expected and observed outer lines.

Material	H_0 (kOe)	T (°K)	Positions of outer lines[a] Expected (mm/sec)			Observed (mm/sec)			
Co^{57} in Cu	66	5	−5.2 ±0.1	−4.7 ±0.1	+4.7 ±0.1	+5.2 ±0.1	−5.1 ±0.1	−4.4 ±0.2	+4.4 ±0.2	+5.1 ±0.1
Co^{57} in Au	72	80	−5.6 ±0.1	−4.5 ±0.1	+4.5 ±0.1	+5.6 ±0.1	−5.6 ±0.1	−4.6 ±0.1	+4.6 ±0.1	+5.6 ±0.1
Co^{57} in Au	67	5.5	−5.0 ±0.2	−3.6 ±0.2	+3.6 ±0.2	+5.0 ±0.2	−5.2 ±0.1	−3.6 ±0.1	+3.4 ±0.1	+6.4 ±0.1
½% Fe^{57} in Cr	65	5.0	−1.13±0.05	−0.57±0.05	+0.57±0.05	+1.13±0.05	−1.20±0.05	−0.60±0.05	+0.65±0.05	+1.10±0.05

[a] Line positions are relative to the center of the spectrum. The expected line positions use the values of H_{nuc} obtained with the experimental sample only in an external field (Ref. 7).

to those described in detail elsewhere.[8] The velocity pickup signal, suitably amplified and biased, was used to drive the address of the multichannel analyzer. Simultaneously, for normalization purposes, a second single-channel analyzer, selecting gamma rays above the 14-keV window, was used to provide a counting rate unrelated to the source velocity; these pulses were stored in another segment of the analyzer memory.

Figure 6 shows the Mössbauer hyperfine pattern for a Co^{57} in copper source versus an enriched Fe^{57} iron foil absorber; both source and absorber were at the same applied field and temperature: $H_0=66$ kOe and $T=5$°K. The data were fit with a spectrum of eight Lorentzian lines by means of a least squares analysis performed on an IBM-709 computer.[9] The positions of the outer doublets are shown in the figure. The outer lines of the doublets are the more intense; thus, the internal fields of source and absorber are antiparallel, so that the effective field at the Fe^{57} nuclei in copper is positive, in agreement with temperature-dependent data.[7] As a check of the over-all consistency of the results, the effective fields in source and absorber were measured separately at $H_0=66$ kOe and $T=5$°K using single line absorber and source, respectively. From these measurements the internal fields were determined to be: $H_{nuc}^{s}=29$ kOe and $H_{nuc}^{a}=294$ kOe; thus $H_1/H_2=0.10$. The expected pattern of 8 lines, Fig. 5, have intensity ratios of 9:3:1:3:3:1:3:9 for the antiparallel case. The velocity positions of these lines can also be read from Fig. 5 if the vertical scale is reduced by $294/331\approx0.9$. The expected and observed line positions are given in Table I. The agreement on the outer lines is good. (The inner four lines are too close and the widths are too uncertain to attach much significance to the computer fits.) The fact that H_{nuc} is positive and smaller than H_0 implies that the induced field H_i is negative, which is also in agreement with temperature-dependent measurements.

The results of experiments performed using a Co^{57} source in gold at two different temperatures are shown

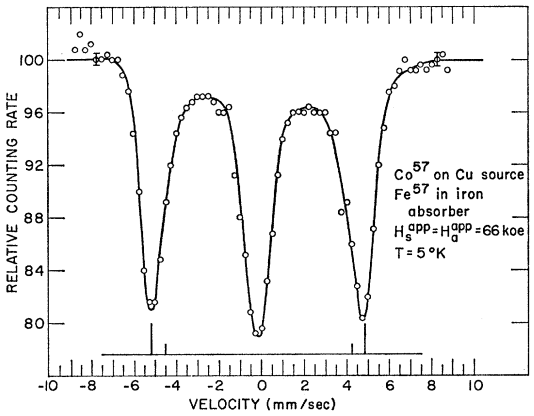

FIG. 6. Hyperfine absorption spectrum for a Co^{57} in Cu source versus an iron foil absorber enriched in Fe^{57}; both source and absorber have $H_0=66$ kOe and $T=40$°K. The index lines show the locations and relative intensities of the outer pairs of hyperfine components as calculated by the computer.

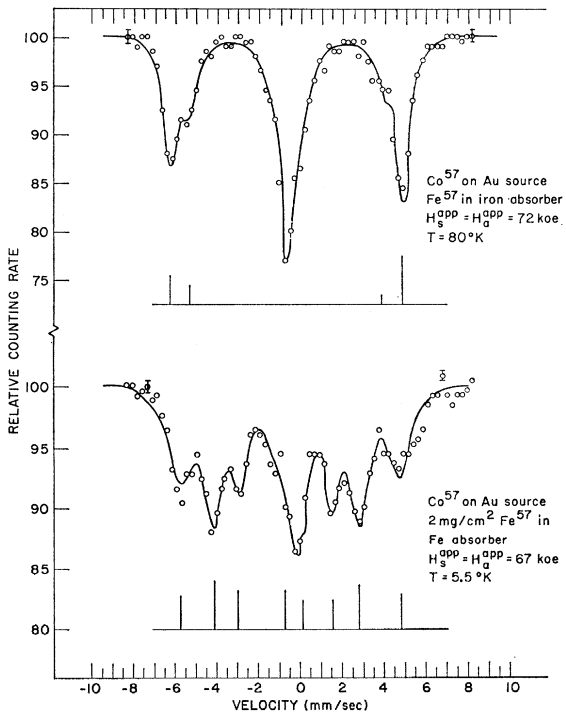

FIG. 7. Hyperfine spectra of a Co^{57} in Au source versus an enriched iron foil absorber; both source and absorber are at the same field and temperature: (a) $H_0=72$ kOe, $T=80$°K and (b) $H_0=67$ kOe, $T=5.5$°K.

[8] See, for example, D. Rubin, Rev. Sci. Instr. 33, 1358 (1962).

[9] The results for all of the data reported here were obtained by similar computer analyses. We are grateful to J. P. Schiffer of the Argonne National Laboratory for making the computer program available to us.

in Fig. 7. In both cases the absorber, iron enriched in Fe^{57}, was at the same temperature and in the same external field as the source. Consider the upper curve, Fig. 7(a), taken with $H_0=72$ kOe and $T=80°K$. The spectrum is quite similar to that in Fig. 6; the outer doublets clearly show the weaker lines inside the stronger ones. The internal fields of source and absorber are, therefore, antiparallel; H_{nuc} at Fe^{57} in a gold environment is positive for these values of H_0 and T. In Table I the expected line positions inferred from measurements using a single line absorber are compared with the observed positions. Again the agreement on the outer lines is good. As in the Cu experiment described above, H_{nuc} is positive and less than H_0, so that H_i is negative.

Figure 7(b) shows the results in the gold source for $H_0=67$ kOe and $T=5.5°K$. The spectrum has changed dramatically from the previous case. The internal fields of source and absorber are parallel as shown by the outer lines being weaker than the next inner ones. H_{nuc} is thus negative, and the induced field is, therefore, negative and large. The expected and observed line positions are given in Table I.

Finally, we consider the $\frac{1}{2}\%$ Fe^{57} in Cr alloy absorber. Previous results[10] had already shown that, below about 50°K, a spontaneous magnetization of about 35 kOe is present. Moreover, in an applied field, the effective field is essentially temperature-independent (data taken above 1.4°K) and greater than H_{int}, but less than the absolute sum of H_{int} and H_0. If we describe H_{nuc} phenomenologically as $H_{nuc}=H_0+H_{int}+H_i$, then the signs of H_{nuc} and H_{int} are the same, but could be either positive or negative depending on the sign and magnitude of H_i. Without a detailed study near the Néel temperature, the signs of H_{int} and H_{nuc} must be determined by a circular-polarization measurement.

Figure 8 shows the result for a Co^{57} in Cu source versus a $\frac{1}{2}\%$ Fe^{57} in Cr absorber, both in an external field of 65 kOe at 5°K. Here we are comparing H_{nuc} in Cr with that in Cu, which we now know to be positive. The outer, unresolved, lines are clearly weaker than the next inner ones. Using a single line source, the effective field in the absorber was determined to be approximately 65 kOe, so that the ratio $H_1/H_2\approx 28/65\approx 0.43$. From Fig. 5 it is clear that the hyperfine fields are parallel and, moreover, that the second and third lines from the end are nearly degenerate. The expected and observed positions of the outer lines, given in Table I, agree reasonably well. The effective field H_{nuc} is therefore positive, so that H_{int} is also positive; in this *phenomenological* view H_i is negative.[10]

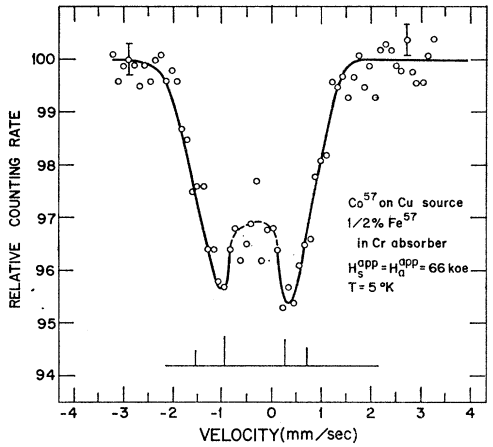

FIG. 8. Hyperfine spectrum of a Co^{57} in Cu source versus a chromium foil absorber containing $\frac{1}{2}\%$ Fe^{57}; both source and absorber have $H_0=65$ kOe and $T=5°K$.

The sign of the induced fields at the Fe^{57} nucleus in absorbers of 1% Fe^{57} in Mo and Rh have also been found to be negative.

The determination of the sign of H_{nuc}, and hence H_i, has important physical significance[11] independent of the detailed behavior of H_i as a function of H_0 and T. In the cases of Co^{57} in Cu, Au, Mo, and Rh, H_i is large and negative, implying a core polarization origin. In the Fe^{57} in Cr absorber the spontaneous field below 50°K is positive ($H_{int}\approx+35$ kOe), implying that the core polarization is not dominant.

The technique is seen to have general application for the determination of the sign of internal fields in iron alloys.[11] Moreover, in special cases the technique also has some advantages for determining the magnitude of H_{nuc}. For example, if H_{nuc} is small, than the Mössbauer pattern observed with a single line analyzer results in a broadened unresolved pattern of four lines in the favorable case of a longitudinal field. If a polarized iron analyzer is used, the observed pattern exhibits outer doublets [Figs. 6 and 7(a)]. Even though the splitting of these doublets is smaller than the total splitting, the simplicity of the pattern may make the determination of H_{nuc} quite reliable.

ACKNOWLEDGMENT

We wish to thank A. J. Freeman for his continued interest and for helpful discussions.

[10] N. Blum, A. J. Freeman, L. Grodzins, J. H. Wernick, and G. K. Wertheim, Bull. Am. Phys. Soc. 8, 43 (1963) and N. Blum, A. J. Freeman, and L. Grodzins, in *Perturbed Angular Correlations*, edited by E. Karlsson, E. Matthias, and K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1964), p. 289. Subsequent data, taken at higher external fields, may alter the interpretation given in this reference.

[11] R. E. Watson and A. J. Freeman, Phys. Rev. 123, 2027 (1961). See also the recent review by A. J. Freeman and R. E. Watson, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York, 1964), Vol. 2.