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DEPENDENCE OF NUCLEAR POLARIZED Fe^{57} MÖSSBAUER SPECTRA ON THE MAGNITUDE AND SIGN OF THE HYPERFINE FIELD

P. Reivari
Department of Technical Physics, Technical University of Helsinki, Otaniemi, Finland
(Received 6 December 1968)

The Mössbauer spectra of Fe^{57}, with the Co^{57} source in iron and palladium matrices, respectively, have been measured in a He^3/He^4 dilution refrigerator. Both the magnitude and sign of the effective field at Co^{57} can be determined from nuclear polarization effects in the spectra, without recourse to an external magnetic field. Experimental data are also presented on the Kapitza thermal boundary resistance which, together with radioactive self-heating, prevents the source from cooling below 0.15°K.

The possibility of observing nuclear polarization by using the Mössbauer effect was first suggested by Dash et al.,[1] who also made the first quantitative measurements on Co^{57} nuclei in an iron matrix.[2-4] Later experiments[5,6] in our laboratory, employing a He^3/He^4 dilution refrigerator, extended this work to considerably lower temperatures. The present paper discusses similar measurements made on Co^{57} in a Pd matrix and shows that the intensity ratios of the Mössbauer lines can give not only the magnitude but also the sign of the hyperfine field on the parent Co^{57} nucleus.

When we study the nuclear polarization of Co^{57} by the Mössbauer effect we have to take into account several nuclear levels.[2] The Co^{57} ground state with spin $I_3 = \frac{7}{2}$ (the subscript refers to the nuclear level in question) decays by electron capture to the Fe^{57} 136-keV level with spin $I_2 = \frac{5}{2}$ and mean lifetime 9×10^{-9} sec. This state decays principally by the emission of a 123-keV gamma ray to the first excited state of Fe^{57}, with $I_1 = \frac{3}{2}$ and lifetime 1.1×10^{-7} sec. The well known 14.4-keV Mössbauer gamma transition occurs between this state and the Fe^{57} ground state, $I_0 = \frac{1}{2}$.

If the degeneracy of the long-lived Co^{57} parent ground state is lifted by a negative magnetic hyperfine field, as in an iron matrix,[4] then the sublevel with magnetic quantum number $m_3 = -\frac{7}{2}$ will become preferentially populated at low enough temperatures. When the Co^{57} decays, the selection rules operate in such a way that some polarization will be preserved right through the decay chain to the 14.4-keV state of iron to give for instance more $m_1 \to m_0 = -\frac{3}{2} \to -\frac{1}{2}$ than $\frac{3}{2} \to \frac{1}{2}$ gamma transitions, i.e., the line of most negative velocity in the Mössbauer spectrum will be more intense than the corresponding positive velocity line. If H_{eff} at the parent nucleus is positive the situation is reversed.

We can assume that the nuclear spin populations of the two intermediate states 1 and 2 remain constant during the decay process because their lifetimes are short compared with estimates of the nuclear relaxation time.[2] We thus need only the nuclear magnetic moment of Co^{57} in its ground state, the appropriate Clebsch-Gordan coefficients, and the temperature for calculating the hyperfine field at the Co^{57} nucleus from the intensity ratios of the Mössbauer spectrum. Whether the most enhanced line is at the positive or negative end of the velocity spectrum gives the sign of the field. This conclusion relies on our assumption that there is no significant change

in polarization during the actual decays. Depolarization effects have been observed in the K-capture process in Tutton salts,[7] but one would expect that this effect is considerably smaller in a metal than in a salt owing to the large electron mobilities. The splitting of the Mössbauer spectrum, of course, gives the effective field at the Fe^{57} nucleus.

In the present experiments Co^{57} nuclei in a Pd matrix were studied. The 3.5-mCi source was 6 mm in diameter and 0.025 mm thick. The average $3d$-transition metal concentration in the Pd was 0.1 at.%. The source was located in liquid helium inside the mixing chamber of a dilution refrigerator, along with a carbon resistor to monitor the temperature. To obtain appreciable polarization the temperature was reduced to 80 mdeg K.

The resulting spectrum is shown in Fig. 1(a) and a spectrum of Co^{57} in iron is given in Fig. 1(b) for comparison. The opposite polarization is obvious and indicates opposite signs for the hyperfine field at Co^{57} nuclei in the two different matrices. The direction of positive velocity can be checked from the isomer shift.

As in the case of Co^{57} in iron, it was noticed that the ratios of the absorption lines approached a saturation value in spite of decreasing mixing chamber temperature. This is due to the self-heating of the source which was calculated to be 7 erg/sec and arises mainly from the absorption of conversion electrons. The intensity ratio ρ of the outermost lines is a well-known function of the source temperature T_S,[4] i.e.,

$$\rho = \rho \exp(\xi/T_S), \tag{1}$$

where $\xi = \mu H_{\text{eff}}/kI_3$ is the magnitude of the magnetic hyperfine interaction. On the other hand, according to the theory of the Kapitza thermal boundary resistance, at equilibrium

$$T_{\text{lim}}{}^4 = T_S{}^4 - T_{\text{He}}{}^4, \tag{2}$$

where T_{He} is the temperature of the helium bath around the source and T_{lim} the source temperature when $T_{\text{He}} = 0°\text{K}$. Putting the observed values of T_{He} and ρ into Eqs. (1) and (2) we get the values $T_{\text{lim}} = 0.15°\text{K}$ and $H_{\text{eff}} = +260 \pm 20$ kOe. This is in agreement with $H_{\text{eff}} = 280 \pm 20$ kOe for a 0.1-at.% solution of Co in Pd measured by conventional nuclear orientation.[8] As the field of Co in Co is negative,[9] the field in Co-Pd alloys must go through zero at some concentration. This has been found experimentally to occur at about 30-at.% Co concentration.[10]

From T_{lim} one can calculate the boundary resistance R between Pd and a dilute solution of He^3, $RT^3 = T_{\text{lim}}{}^4 A/4\dot{Q} \sim 10^{-5}$ sec °K^4 cm^2/erg, where A is the source area. The order of magnitude obtained is the same as for boundary resistances commonly measured.[11] Equations (1) and (2) give values of T_S after substitution of T_{lim} and ξ. This analysis can be only approximate as boundary resistances have been found to deviate from the T^{-3} law. Experiments show that, for example, for a boundary between copper and liq-

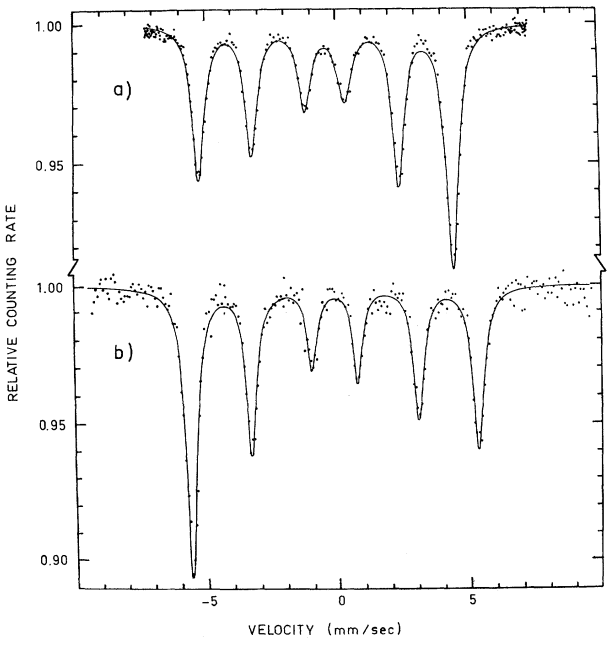

FIG. 1. (a) Polarized Mössbauer spectrum of Co^{57} in Pd at $T_S = 0.15°\text{K}$. (b) Comparison spectrum of Co^{57} in Fe at 0.13°K. The same 0.025-mm stainless-steel absorber at 300°K was used in both cases.

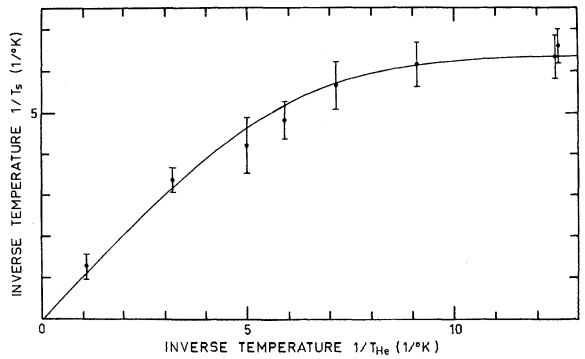

FIG. 2. Saturation of $1/T_S$ vs $1/T_{\text{He}}$. The error bars are due to the counting statistics.

uid helium this law is only valid below 0.1°K. Using the best fitting values of T_{lim} and ξ, $1/T_S$ vs $1/T_{\mathrm{He}}$ can be plotted as shown in Fig. 2.

The present experiments demonstrate that Mössbauer measurements at very low temperatures are a useful means of determining the magnitude and sign of the hyperfine field at the parent nucleus without using any external magnetic field, provided the nuclear transition parameters are known. In absorber materials the sign cannot be determined without an external field.

I would like to acknowledge useful discussions with G. J. Ehnholm, G. M. Kalvius, T. E. Katila, O. V. Lounasmaa, and G. R. Pickett.

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