Sign of conduction-electron polarization in iron

V. Ya. Gamlitskiĭ, O. A. Gurkovskiĭ, V. I. Nikolaev, I. N. Nikolaev,
V. M. Cherepanov, and S. S. Yakimov

I. V. Kurchatov Institute of Atomic Energy

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It is found experimentally that in two contiguous metals, one of which is ferromagnetic, the conduction electrons (CE) diffusing into the nonmagnetic metal from the ferromagnet retain their polarization. The effect was revealed by the appearance of hyperfine splitting of the Mössbauer spectrum of pure tin, thin layers of which were evaporated in vacuum between iron layers. It is shown that in this way information can be obtained on the polarization of the CE in magnetic substances and on the electron-spin mean free path in a nonmagnetic metal. The CE polarization in iron is found to be negative.

1. INTRODUCTION

Conduction electrons (CE) play an important role in the mechanism of magnetic ordering of ferro- and antiferromagnetic metals and alloys[1]. Thus, it is customarily assumed that in f metals and in dilute solutions of d and f metals in nonmagnetic matrices, the magnetic order is due to indirect exchange interaction via the conduction electrons. As to d metals, it is still unclear whether the onset of ferromagnetism in these metals can be explained by considering only the direct d-d exchange coupling. It is assumed at present that an important role in the formation of exchange interaction in d metals should be played by the CE. It is therefore important to know such characteristics of CE in magnets as the sign and magnitude of their polarization.

In spite of the very large number of papers devoted to this problem, the question of the sign of the total spin polarization of the CE is still under debate even in the case of iron, which has been most exhaustively investigated. A clear illustration of this situation is provided by the contradictory results obtained by different experimental methods. On the one hand, the conclusion that the CE in iron have negative polarization was based on experiments on the scattering of polarized neutrons[2], on the angular correlations of the γ rays in positron annihilation[3], and on the dependence of the hyperfine field and of the magnetization on the pressure[4]. On the other hand, in the analysis of the experimental data on the magnitude and sign of the hyperfine field at nuclei of a nonmagnetic impurity dissolved in iron[5], and also of the results of investigations of the Mössbauer effect (ME) on Fe^{57} nuclei in disordered alloys based on iron[6], a positive total polarization of the 4s electrons was obtained.

We do not discuss here in detail the data obtained by the aforementioned experimental methods. A critical review was presented by Duff and Das[7] and by Sedov[8]. We note only that the discrepancy between the obtained results may be connected with the fact that in most investigated methods it is necessary to carry out a very complicated reduction of the experimental data, and their interpretation is based on theoretical calculations and assumptions whose validity is not obvious in many cases.

In this paper we use a fundamentally new method, which makes it possible to obtain direct information on the polarization of the CE in magnetic metals and alloys.

2. IDEA OF THE EXPERIMENT

Consider the contact between two metals, one of which is ferromagnetic. If the CE in the ferromagnets are polarized, it can be assumed that the polarization will extend also to the nonmagnetic metal. Owing to the contact Fermi interaction, a hyperfine field H_n is produced thereby at the nuclei of the nonmagnetic-metal atoms, and the value of this field will obviously depend on the degree of polarization of the CE in the ferromagnet. The Mössbauer effect can be used to find the value of H_n, and experiments in an external magnetic field can be used to determine (as will be shown below) the sign of the polarization of the CE in the ferromagnet. The dependence of H_n on the average length over which the electron preserves the spin direction makes it possible to determine experimentally the mean free path of the electron spin in a nonmagnetic metal.

3. THE SAMPLES

The method proposed in this paper was used to study the polarization of CE in iron. The investigated samples were alternating thin layers of iron and tin enriched with Fe^{67} and Sn^{119}. The samples were prepared in the following manner: We evaporated in vacuum in succession, on a mica substrate, layers of iron (150 Å), tin (from 20 to 300 Å), and iron (150 Å). To prevent oxidation in air, the samples were coated with a layer of aluminum (~2000 Å). The number of simultaneously prepared samples was such that when they were placed on top of one another the total thickness was large enough for absorption required for the observation of the Mössbauer effect. The vacuum in the course of evaporation was not worse than 4×10^{-7} mm Hg; the deposition rate was ~1 Å/sec. During the course of evaporation, a mass-spectrometer analysis was carried out of the evaporated substances, and also of the composition of the residual gases in the chamber. The film thickness was monitored with a quartz measuring device. The total error in the determination of the thickness was 25%.

To determine the influence of the evaporation conditions on the Mössbauer spectrum of the Sn^{119} nuclei, we prepared also samples containing only tin. The sources in the Mössbauer experiments were $Sn^{119}O_2$ and Co^{57} in Cr.

4. EXPERIMENTAL RESULTS AND DISCUSSION

1. The Mössbauer measurements were performed in an external field H ~ 200 Oe, sufficient (according to magnetic-measurement data) to ensure an identical direction of the magnetization of the iron films, and by the same token of the polarization of the CE penetrating into the tin from both directions.

For a correct interpretation of the results obtained by measuring the Mössbauer effect on tin, it is important to know whether the iron films are fully ferromagnetic and

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single-phase (in connection with the possible formation of alloys or Fe-Sn compounds). To this end, we investigated the Mössbauer effect on the Fe^{57} nuclei of all samples, this being one of the most sensitive phase-analysis methods. The hyperfine field obtained at room temperature was 333 ± 3 kOe (Fig. 1), in good agreement with the analogous value for pure iron. The distortions in the central part of the spectrum, as shown by control experiments, were due to the iron contained in the mica. The smaller symmetry of the individual peaks can be attributed to the influence of residual gases in the iron shown on the value of H_n. As established earlier (see, e.g., [9]), nonmagnetic impurities lead to a decrease of the hyperfine field at the Fe^{57} nuclei, which are the nearest neighbors of the impurity atom. Thus, measurements of the Mössbauer effect on Fe^{57} nuclei show that in our samples the iron is full paramagnetic and contains no noticeable amount of alloys or compounds of the Fe-Sn system.

The main feature of the Mössbauer spectra obtained with the $Sn^{119}O_2$ source for the Fe-Sn-Fe samples with tin film thickness from 20 to 300 Å is the line broadening (in comparison with the spectrum of the film containing only tin), which increases with decreasing temperature and with decreasing thickness of the tin film. Figure 2 shows spectra of an Fe-Sn-Fe sample with average tin thickness ~60 Å in comparison with the spectrum of Sn film.

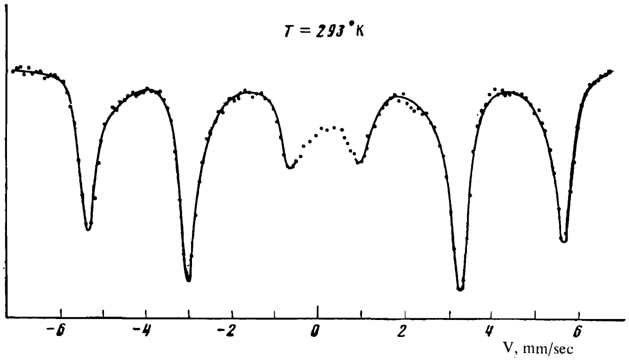

FIG. 1. Mössbauer absorption spectra of 14.4-keV γ quanta of Fe^{57} nuclei in Fe-Sn-Fe samples (iron thickness 150 Å, tin thickness 60 Å).

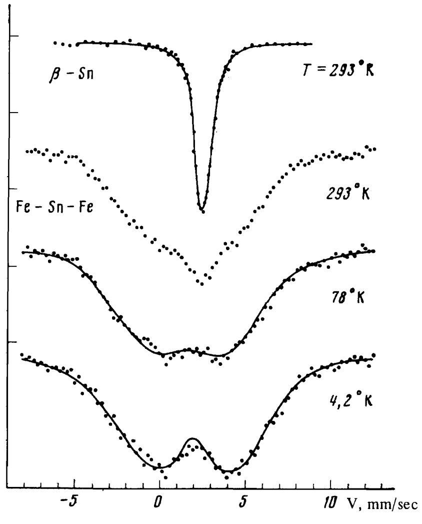

FIG. 2. Mössbauer absorption spectra of 23.8-keV γ quanta of Sn^{119} nuclei in Sn film and in Fe-Sn-Fe samples (iron thickness 150 Å, tin thickness 60 Å). The solid lines show the computer-calculated spectra.

In accordance with the results of the investigation of the Mössbauer effect on iron, it can be stated that the obtained samples are individual alternating layers of iron and tin, and that the line broadening is due to a hyperfine field induced at the tin nuclei by the conduction electrons polarized in the iron. A certain shift of the center of gravity of the spectrum of the Fe-Sn-Fe sample relative to the position of the Sn line may be connected with the influence of the film thickness, and also the structure of the substrate, on the lattice parameters of the films[10], which should lead to a change in the isobaric shift. We note that the line shift can also be due to a change in the isobaric shift. We note that the line shift can also be due to a change in the temperature shift, owing to the increase of the effective Debye temperature of the tin, which is particularly significant at low temperatures.

The most characteristic distinguishing feature of the spectrum is the fact that it has no resolved structure in spite of its relatively large total width. Owing to the scattering of the electron spin in the tin, the degree of polarization of the conduction electrons decreases with increasing distance from the iron boundary, and consequently, H_n decreases. Thus, the observed tin spectrum is a superposition of spectra corresponding to different values of the hyperfine fields H_n. The splitting of the spectrum obviously depends on the effective depth of penetration of the polarization δ into the tin film. The value of δ is determined by the electron mean free path λ_e and by the electron spin mean free path λ_s. Since δ should increase with decreasing temperature and accordingly with increasing λ_e, this explains the observed temperature dependence of the broadening of the Mössbauer spectrum of the tin in the Fe-Sn-Fe sample.

The computer reduction of the spectra was based on the assumption that the conduction-electron polarization decreases exponentially with increasing distance from the iron boundary. In this case the dependence of H_n at the tin nuclei on the distance l (Fig. 3) to the boundary (with allowance for the fact that the iron films are equally magnetized) should be of the form

$$H_n=2H_{n0}\exp(-l_0/\delta)\operatorname{ch}(l/\delta), \qquad (1)$$

where H_{n0} is the value of the hyperfine field at the nuclei of the Sn atoms located at the boundary with the iron.

The resultant spectrum is then described by the expression

$$F(E)=\sum_{i=1}\int_{-l_0}^{l_0}\frac{dl}{1+(4/\Gamma^2)\,[E-2E_{i0}\exp(-l_0/\delta)\operatorname{ch}(l/\delta)]^2}, \qquad (2)$$

where E_{i0} is the position of the i-th line of the spectrum for the Sn atoms at the boundary with the iron.

In the calculation, we varied the parameters H_{n0} and δ. It was found that the best agreement between the calculated curves and the experimental data is obtained at

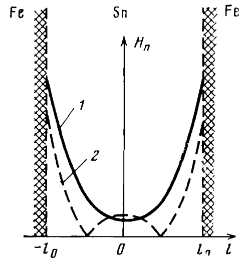

FIG. 3. Distribution function of the field H_n induced at the Sn^{119} nuclei by conduction electrons. Curve 1—external magnetic field H = 0, curve 2—H = 25 kOe.

$H_{n0} \approx 120$ kOe and $\delta \approx 30$ Å for 4.2°K. We note that the tin-film thicknesses given by us correspond to mean values determined from the mass of the evaporated material. Therefore, taking into account the possible "island-like" structure of the tin film, it must be assumed that $\delta \gtrsim 30$ Å. The fact that δ turned out to be relatively small may be due to scattering by impurities (the purity of the Sn^{119} isotope employed by us was 99.95%).

2. Measurements in an external magnetic field H = 25 kOe have made it possible to determine the sign of the polarization of the conduction electrons in iron. Figure 4 compares the Mössbauer spectra of the Sn^{119} nuclei in the investigated sample, obtained at a temperature 77°K in the absence of a magnetic field and in a field H = 25 kOe. The overall narrowing of the spectrum in the external magnetic field means that the hyperfine field $\mathbf{H}_n$ at the nucleus, induced by the polarized conduction electrons, has a direction opposite to that of the external field **H**. Since the Knight shift for tin is positive[11] (the polarizations of the conduction electrons and of $\mathbf{H}_n$ are of the same sign), the polarization of the conduction electrons in tin is opposite to **H**, and consequently opposite to the magnetization of the iron. It is natural to assume that on going through the film boundary the polarization of the conduction electrons remains unchanged, so that it follows from our results that the sign of the polarization of the conduction electrons in iron is negative.

The relatively small narrowing of the spectrum in an external magnetic field H = 25 kOe is connected with the fact that $H > H_n$ in the central part of the tin film. The approximate dependence of the absolute value of the magnetic field at the nucleus on the distance, following application of an external field, is shown in Fig. 3 (curve 2). It is seen that H_n decreases by an amount H not in the entire thickness of the tin film. At the Sn-atom nuclei located in the central layers, the magnetic field at the nucleus decreases by an amount smaller than H, and even increases in absolute magnitude. Therefore the criterion for the determination of the sign of the conduction-electron polarization should be not so much the narrowing of the spectrum in an external field as the absence of large broadening. If the conduction-electron polarization were positive, application of an external field would cause the resultant field at the Sn^{119} nuclei to be not less than 25 kOe in the entire film, and this would lead to a rather large splitting of the spectrum (Fig. 4).

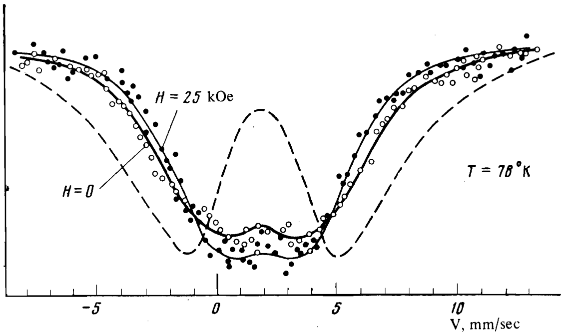

FIG. 4. Mössbauer spectra of Sn^{119} nuclei in Fe-Sn-Fe sample in the absence of a magnetic field and at H = 25 kOe. The solid lines show the computer-calculated spectra. The dashed line shows the spectrum calculated under the assumption of positive polarization of the conduction electrons in the iron.

We note in conclusion that the results of the calculations of the values of δ from the experimental data depend on the form of the function that expresses the dependence of H_n on the distance. It is also necessary to know exactly the actual thickness of the tin film. As to the sign of the conduction-electron polarization, this result does not depend on the structure of the film and on the form of the function $H_n(l)$.

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[1] S. V. Vonsovskiĭ, Magnetizm (Magnetism), Nauka (1971), p. 512.
[2] C. Shull and Y. Yamada, J. Phys. Soc. Japan **17**, Suppl., B-III, 1, 1962.
[3] V. L. Sedov, L. V. Solamatina, and L. A. Kondrasheva, Zh. Eksp. Teor. Fiz. **54**, 1626 (1968) [Sov. Phys.-JETP **27**, 870 (1968)].
[4] D. H. Anderson, Solid State Commun., **4**, 189, 1966.
[5] E. Daniel, in: Hyperfine Interactions, ed. by A. J. Freeman and R. B. Frenkel, Academic, 1967.
[6] M. B. Stearns, Phys. Rev. **B4**, 4081, 1971.
[7] K. J. Duff and T. P. Das, Phys. Rev. **B3**, 2294, 1971.
[8] V. L. Sedov, Usp. Fiz. Nauk **94**, 417 (1968) [Sov. Phys.-Uspekhi **11**, 163 (1968)].
[9] G. K. Wertheim, V. Jaccarino, J. H. Wernick, and D. N. E. Buchanan, Phys. Rev. Lett. **12**, 24, 1964.
[10] K. L. Chopra, Thin Film Phenomena, McGraw, 1969.
[11] T. J. Rowland, Nuclear Magnetic Resonance in Metals, Pergamon.

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