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ANNIHILATION OF POLARIZED POSITRONS IN FERROMAGNETIC METALS

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In the annihilation of polarized positrons in a magnetized ferromagnet, the two-quantum annihilation rate depends on the relative orientation of the direction of polarization of the positrons and the direction of magnetization of the sample. In the present work we have investigated this effect in iron and nickel. For the quantity $K = (N_+ - N_-)/(N_+ + N_-)$ the following data were obtained: $K_{Fe} = (1.5 \pm 1.5) \times 10^{-4}$, $K_{Ni} = (-0.5 \pm 1.5) \times 10^{-4}$ (N_+ and N_- are the respective numbers of two-quantum annihilations recorded in a given time with the magnetic field direction along and opposite to the direction of polarization of the positrons). On the basis of the results obtained we have drawn conclusions regarding the polarization of 4s electrons in these metals.

INTRODUCTION

WITH the aid of effects observed in the annihilation of polarized positrons in ferromagnetic metals, it is possible to draw conclusions regarding the micromagnetic structure of these metals. The possibility of studying ferromagnetic metals by this means is based theoretically on the fact that the most probable annihilation process, two-quantum annihilation, can take place only for a spin-singlet state of the annihilating electron-positron pair. Thus, the probabilities of annihilation of polarized positrons with electrons of the different groups participating in ferromagnetism depend on the relative orientation of the magnetization direction in the sample and the direction of polarization of the positrons.

It is well known that the study of the electronic structure of solids by means of positrons is based on the ability of these particles, after incidence on the target and prior to their annihilation, to be thermalized, i.e., to lose almost completely their initial kinetic energy. In the study of ferromagnets it is important that in the thermalization the initial polarization of the positrons is essentially unchanged.[1] Thus, a thermalized, polarized positron is a test particle which allows us to obtain information on the magnetic electronic structure of the ferromagnetic metals.

A number of authors[2-6] have studied the effect of the relative orientation of the direction of polarization of the positrons and the direction of magnetization of ferromagnetic samples on the angular distribution of the annihilation γ-ray pairs. The data obtained in these studies have been interpreted in terms of the simplest representations of the electronic structure of ferromagnetic metals. For this purpose a simplified model is usually used in which the spectrum of electronic states is separated into two qualitatively different bands corresponding to 4s and 3d levels of isolated atoms. Such models have not taken into account the possibility of "hybridization" of the bands, the contribution of the orbital magnetic moment, and the possibility of a nonuniform polarization and oscillation of the polarization of the 4s electrons. Also not taken into account is the splitting of 3d-like states by the intracrystalline field.

In the experimental studies mentioned it was observed that for large values of the angle θ the quantity $\Delta n(\theta) = n_+(\theta) - n_-(\theta) > 0$ ($n_+(\theta)$ and $n_-(\theta)$ are functions describing the angular correlation of the annihilation γ-ray pairs for magnetic field directions along and opposite to the direction of polarization of the positrons). This result means that the direction of the total spin magnetic moment of the strongly localized electrons (3d electrons) coincides with the direction of the magnetization vector of the sample. In clarification of the question of polarization of weakly localized electrons (4s electrons) by means of annihilation γ-ray angular correlation effects, certain difficulties arise.[4]

In principle it should be possible to obtain data for the polarization of 4s electrons, proceeding from a detailed study of the shape of $\Delta n(\theta)$ curves for single crystals. This possibility exists as the result of the difference between the Fermi surfaces for the two subbands of 4s electrons with different spin directions in the case when these electrons have a polarization different from zero. However, because of the difficulties in interpretation of the experimentally observed situation, no definite conclusions have been drawn from these studies.[6]

For the case of a polarized positron in a ferromagnetic metal, the ratio of the total probabilities of 2γ and 3γ annihilation depends to a certain degree on the relative directions of the positron polarization and the magnetization of the sample. One of the present authors[3] has suggested using this effect to determine the polarization of the 4s electrons. It was noted that, since a value $N_+ - N_- < 0$ was obtained for nickel (N_+ and N_- are the number of 2γ annihilation events recorded in a given time for magnetic field directions along and opposite to the direction of polarization of the positrons), in terms of the simplest band model it follows from this fact that the polarization of the 4s electrons in this metal is negative.

In the present work we have performed direct measurements of the quantity $K = (N_+ - N_-)/(N_+ + N_-)$ for nickel and iron. From the results of these studies it follows that the value previously obtained[3] for this quantity is erroneous and, if we take into account the accuracy achieved in the measurements, is not definitely negative. Since for this reason the conclusion drawn by Sedov[3] that the polarization of the 4s electrons is negative is in question, in the present article we have made numerical estimates of the polarization of the 4s electrons of nickel on the basis of the newly obtained result. The experimental value of K for iron is compared with the calculated value of this quantity corresponding to the case when the 4s electrons in iron are not polarized.

It should be noted that although the measured values K are small quantities ($\sim 10^{-4}$) the study of the corresponding effect is feasible. The smallness of K is determined primarily by the fact that it is proportional to the ratio of the cross sections for 3γ and 2γ annihilation ($\sigma_{3\gamma}/\sigma_{2\gamma} = 1/372$[7]). In addition, the values of K are directly related to quantities characterizing the band structures of the ferromagnetic metals (see section 2 of the present article, equations (1)—(3)). This fact can be utilized in corresponding theoretical studies.

It should also be noted that the interpretation of the values of K does not involve or is not very sensitive to certain factors which in principle must be taken into account in studies of the band structure of ferromagnetic metals by means of annihilation γ-ray angular correlation. The following factors are in this class: a) the effect of the periodic potential of the crystal lattice on the annihilation γ-ray angular distribution pattern; b) the difference in shape of the Fermi surface for the subbands of 4s electrons with different spin directions; c) the excitation during positron annihilation of electron-hole pairs in the surrounding electron medium.[8]

1. EXPERIMENT

The basic arrangement of the apparatus is shown in Fig. 1. The measurement process consisted of recording the signals produced in a given period of time by annihilation γ-ray pairs for two mutually opposing directions of the magnetic field.

The radioactive isotope Na^{22} was used as the source of positrons. As the result of β^+ decay of the nuclei of this isotope, longitudinally polarized positrons are produced whose degree of polarization is $\langle v/c \rangle$. The activity of the source used was about 10 millicuries. The radiation was detected by scintillation counters consisting of CsI crystals 80 mm in diameter and FÉU-24 photomultipliers. In view of the smallness of the effect being sought it was necessary to shield the photomultipliers carefully from the magnetic field. For this reason the magnet was surrounded by iron plates and the detectors were placed in thick-walled iron tubes. The field strength used was 8 kG. The working surface of the samples was a flat rectangle 18 × 25 mm.

In this experiment it was necessary to keep in mind that the finite angles subtended by the detectors could be the cause of erroneous results. This is due to the fact that the finite size of the detectors leads to some selectivity in the recording of γ-ray pairs, depending on the angle between the directions of propagation of the two γ rays. For this reason, measurements were made successively at different distances of the detectors from the sample. The true effect of the change in the number of two-quantum annihilation events with change of magnetic field direction was determined by extrapolating the measured value as the distance between the detectors and the sample was reduced.

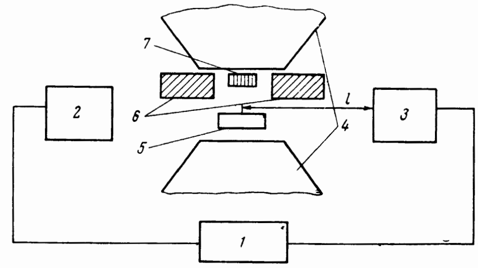


FIG. 1. Basic arrangement of the apparatus: 1 — coincidence circuit, 2, 3 — γ-ray detectors, 4 — magnet poles, 5 — sample, 6 — lead blocks, 7 — positron source (Na^{22}).

In order to avoid possible errors associated with any other instrumental effects, control measurements in a copper sample were made for each detector position. The experimental result for each detector position was obtained by recording 10^8—10^9 counts from annihilation γ-ray pairs.

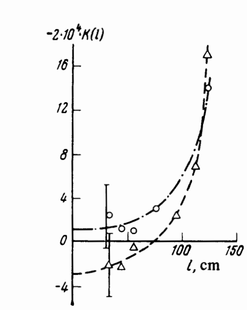
2. RESULTS

Figure 2 shows the quantity $K(l) = [N_+(l) - N_-(l)]/[N_+(l) + N_-(l)]$ obtained in the present work for iron and nickel, as a function of the distance l at which the detectors were located with respect to the sample (see Fig. 1). As we have already mentioned, we are interested in the value K(0). From the plots in Fig. 2 it can be seen that for iron and nickel the absolute value of the effect being sought does not exceed the experimental errors, which for the minimum value of l amount to $\Delta K(l_{min}) = \pm 1.5 \times 10^{-4}$. An estimate of the effect of interest made by Berko and Zuckerman[4] showed that for iron $K_{Fe} \approx 10 \times 10^{-4}$. According to the results obtained in the present work, $K_{Fe}(0) = (1.5 \pm 1.5) \times 10^{-4}$, $K_{Ni}(0) = (-0.5 \pm 1.5) \times 10^{-4}$.

These results can be used to obtain information on the electronic structure of these metals. On the basis of the conclusions drawn by Berko and Zuckerman,[4] it is not difficult to obtain an expression connecting K(0) with quantities characterizing the band structure of a ferromagnetic metal. If we neglect quantities of the order of $K^2(0)$ and $(\sigma_{3\gamma}/\sigma_{2\gamma})K(0)$, then the following equality occurs:

$$2K(0) = \frac{1}{f^-} - \frac{1}{f^+}, \qquad (1)$$

FIG. 2. Experimental results on the effect of relative orientation of the magnetization of ferromagnetic samples and positron polarization, on the rate of two-quantum annihilation. The quantity plotted along the vertical axis was studied as a function of the distance l at which the γ-ray detectors were located from the sample. $N_+(l)$ and $N_-(l)$ are the numbers of 2γ annihilations recorded for magnetic field directions along and opposite to the positron polarization direction. ○ — points obtained in measurements with a nickel sample, Δ — in an iron sample.



where

$$f^{\pm} = 3\frac{\sigma_{2\gamma}}{\sigma_{3\gamma}}\frac{\sum_i (1 \mp P_e{}^i P_p{}^0) W_i}{\sum_j (3 \mp P_e{}^j P_p{}^0) W_j}; \qquad (2)$$

P_e^i is the polarization of an electron in the i-th state; P_p^0 is the polarization of the thermalized positron ($-1 \le P_{e,p} \le 1$); W_j is the wave-function overlap integral of an electron in the j-th state and the thermalized positron,

$$W_j = \int |\psi_e{}^j(r)\psi_p{}^0(r)|^2 dV. \qquad (3)$$

For the simplest model of ferromagnetic metals of the iron group, which we have discussed in the Introduction, the quantities W_j are constants within one band. In this case the polarizations of 3d and 4s electrons, P_e^{3d} and P_e^{4S}, are related to the numbers characterizing the distribution of electrons in the bands by the relations $P_e^i = (n_i^+ - n_i^-)/(n_i^+ + n_i^-)$. Thus, in this case relation (1) can be considered as a certain condition connecting the four numbers which describe the electron distribution in the 3d and 4s bands. There are two other independent equations which relate these same numbers. These are the equality $n = n_{3d}^+ + n_{3d}^- + n_{4S}^+ + n_{4S}^-$, where n is the total number of valence electrons per atom, and the condition relating the numbers characterizing the electron distribution in the bands with the value of the atomic magnetic moment, $M = n_{3d}^- - n_{3d}^+ + n_{4S}^- - n_{4S}^+$ (here M is the number of Bohr magnetons per atom of the metal). If there were a fourth independent condition for the electron distribution numbers, these quantities could be determined uniquely. In the case of nickel we can use for this missing condition the suggestion which has been made by a number of authors that the 3d(−) subband in this metal is completely filled (see for example ref. 9), i.e., that $n_{3d}^- = 5$.

The positron wave functions were found by numerical solution of the Schrödinger equation for the Wigner-Seitz sphere. The potential used in these calculations consisted of the nuclear potential and a spherically symmetric potential produced by the distributed charge of the electrons inside this sphere. Here it was assumed that there are seven electrons per atom in the 3d band for iron and nine for nickel. In calculating the wave functions and the quantities W_{3d} we used the data on 3d wave functions obtained by Watson.[10] The 4s-electron states were described by plane waves.

The calculated values of overlap integrals for the electron and positron wave functions are shown in the table. The positron polarization P_p^0 with inclusion of the magnetic field effect was taken as 0.5.

We found that the value $K_{Ni}(0) = -0.5 \times 10^{-4}$ corresponds to the following electron distribution numbers: $n_{3d}^- = 5$, $n_{3d}^+ = 3.94$, $n_{4S}^- = 0.3$, $n_{4S}^+ = 0.76$. The effective polarization of 4s electrons in nickel for this configuration is + 0.43 (the plus sign indicates a negative magnetization of the 4s electrons with respect to the 3d electrons).

As noted by Sedov,[3] in the case where $K(0) < 0$ the conclusion that the magnetization of the 4s electrons is negative on the basis of the electron model used does not depend on the values of the quantities W_i and on the

	W_{3d}/W_{4s}	W_{3p}/W_{4s}	W_{3s}/W_{4s}
Fe	0.3571	0.0387	0,0524
Ni	0.3224	0.0326	0,0409

Note. W_n is the overlap integral (3) of the thermalized-positron wave function with the electron wave function. The lower index designates the electron state.

condition imposed on n_{3d}^-. The accuracy of the measurements made permits the possibility that $K_{Ni}(0) > 0$. The quantity $K_{Ni}(0) + \Delta K$, which is equal to 1×10^{-4}, corresponds to an effective 4s-electron polarization value of -0.05.

Certain factors must be mentioned which could, generally speaking, change the calculated values of P_e^{4S}.

1. The calculations of the numbers $n_i^{\pm}$ have not taken into account the spatial polarization of the electronic medium by the positron and the effect of annihilation with simultaneous production of additional excitations in the electronic medium. The contribution of these effects to the total probability of annihilation is appreciable. This follows from comparison of the calculated annihilation probabilities, based on the data given in the table, and the experimental values of these quantities. The calculated annihilation probability values for nickel and iron are respectively 2.8×10^9 sec^{-1} and 2.6×10^9 sec^{-1}. The experimental data, according to Bisi et al.,[11] are $(5.10 \pm 0.21) \times 10^9$ sec^{-1} for nickel and $(5.02 \pm 0.17) \times 10^9$ sec^{-1} for iron.

The systematic inclusion of these effects is a difficult problem. If we assume that inclusion of these effects decreases the quantity W_{3d}/W_{4S}, this leads to an increase in the absolute value of the P_e^{4S} value corresponding to the upper limit of the experimental value of K.

2. The density distribution of 3d electrons in the metal within the Wigner-Seitz cell differs from the distribution corresponding to the atomic wave functions used in the present calculations.[12] Near the nucleus, these electrons are less localized in the metal than in an isolated atom. Inclusion of this fact should lead to a change in P_e^{4S} of the opposite sign from that of the assumed action of the factors noted in paragraph 1 above.

3. In the calculations of P_e^{4S} for nickel, it was assumed that $n_{3d}^- = 5$. Variation of this quantity within the limits of its physically probable value produces small changes in the quantity P_e^{4S} corresponding to $K_{Ni}(0) + |\Delta K|$. For example, for $n_{3d}^- = 4.5$ the value indicated for P_e^{4S} is -0.06.

In regard to the interpretation of the result obtained for iron, it is impossible in this case to repeat the reasoning given above for nickel. For iron there is no basis to assume that one of the 3d subbands is completely filled. In order to draw conclusions from the value obtained for $K_{Fe}(0)$, we will use theoretically calculated values of the electron distribution numbers for iron and compare the experimental value of $K_{Fe}(0)$ with the value of K_{Fe} calculated by means of these numbers. According to Wakoh and Yamashita[13] $n_{3d}^+ = 2.45$, $n_{3d}^- = 4.65$, $n_{4S}^+ = n_{4S}^- = 0.45$. The value of K_{Fe} corresponding to these numbers, calculated by means of equation (1), turns out to be 3.78×10^{-4}. Comparison of this number with the experimentally obtained value $K_{Fe}(0) = (1.5 \pm 1.5) \times 10^{-4}$ indicates that in iron the 4s electrons are effectively polarized opposite to the 3d electrons. This conclusion agrees with the results of neutronographic studies of iron carried out by Shull and Yamada.[14]

[1]A. J. Dufner and A. V. Bushkovitch, Bull. Am. Phys. Soc. 9, 562 (1964).
[2]P. Mijnarends and L. Hambro, Phys. Letters **10**, 272 (1964).
[3]V. L. Sedov, Zh. Eksp. Teor. Fiz. **48**, 1200 (1965) [Sov. Phys.-JETP **21**, 800 (1965)].
[4]S. Berko and J. Zuckerman, Phys. Rev. Letters **13**, 339 (1964).
[5]T. W. Mihalisin and R. D. Parks, Phys. Letters **21**, 610 (1966).
[6]T. W. Mihalisin and R. D. Parks, Phys. Rev. Letters **18**, 210 (1967).
[7]A. Ore and J. L. Powell, Phys. Rev. **75**, 1696 (1949).
[8]V. L. Sedov, Fiz. Tverd. Tela **9**, 1957 (1967) [Sov. Phys.-Solid State **9**, 1540 (1968)].
[9]N. F. Mott, Adv. in Phys. **13**, 325 (1964).
[10]R. E. Watson, Phys. Rev. **119**, 1934 (1960).
[11]A. Bisi, G. Faini, E. Gatti, and L. Zappa, Phys. Rev. Letters **5**, 59 (1960).
[12]J. H. Wood, Phys. Rev. **117**, 714 (1960).
[13]S. Wakoh and J. Yamashita, J. Phys. Soc. Japan **21**, 1712 (1966).
[14]C. Shull and Y. Yamada, J. Phys. Soc. Japan, Suppl. **17**, 1 (1962).

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