[6]I. Langmuir, Phys. Rev. 26, 585 (1925).

[7]A value for the mfp of the same order of magnitude as this has recently been derived from a quite different analysis based on the nonlinear treatment. See the review article by Y. Klimontvich and V. P. Silin, in Plasma Physics, edited by J. E. Drummond (McGraw-Hill Publishing Company, Inc., New York, 1961), Chap. 2.

[8]See the review article by J. E. Drummond in Plasma Physics, edited by J. E. Drummond (McGraw-Hill Publishing Company, Inc., New York, 1961), p. 2.

POLARIZED POSITRON ANNIHILATION IN FERROMAGNETS*

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It is well known that the measurement of the angular correlation of the two photons emitted during positron annihilation in solids can provide information concerning the momentum distribution of the electrons annihilating with the positron.[1] Positron annihilation with spin-aligned electrons in ferromagnets was first used by Hanna and Preston[2] for positron-spin analysis. They also indicated how such measurements can yield information regarding the momentum spectrum of the ferromagnetic electrons.[3] Their use of cylindrical geometry, particularly in its integral form, did not reveal some details of interest to the study of ferromagnetism; they also reported a null effect in nickel. Lovas[4] used parallel-slit geometry in a similar experiment, but with insufficient angular resolution and statistics. We have performed several measurements with improved statistics using a parallel-slit system.[5]

Recently Mijnarends and Hambro[6] also announced new, independent measurements in iron and interpreted their results as definite proof for the polarization of the conduction electrons. In this Letter we would like to present some of our new data in iron and nickel and to interpret the results in a rather different light. The experimental setup used was similar to one described previously,[7] with the addition of an electromagnet used to saturate the ferromagnetic samples. Positrons from an 8-mCi Na^{22} source were collimated by a circular aperture and allowed to strike the $\frac{3}{8}$-inch diameter samples. The magnetic field was set parallel ($\uparrow$) or antiparallel ($\downarrow$) to the incident positron momentum. The photons from positron-electron singlet annihilation were counted by the 7-inch long NaI crystals behind parallel lead-slit collimators. The slits subtended an angle of 0.75 mrad or 2 mrad in various runs. At each angle two counting rates, $N_\uparrow(\theta)$ and $N_\downarrow(\theta)$, were obtained with an automatic system by several cyclings through the predetermined angular range.

The model used in describing the observed

$$p(\theta)=\frac{N_\uparrow(\theta)-N_\downarrow(\theta)}{N_\uparrow(\theta)+N_\downarrow(\theta)}$$

assumes that the polarized positrons slow down to their ground state $\psi_+(\vec{r})$ in the crystal lattice but that their original spin direction is not changed appreciably.[8] Let the annihilation with the lth electron result in a momentum distribution

$$\rho_l(\vec{p})=\text{const}\left|\int\psi_l(\vec{r})\psi_+(\vec{r})e^{-i\vec{p}\cdot\vec{r}}\right|^2,$$

where $\psi_l(\vec{r})$ is the lth-electron wave function; the positron-electron interaction is neglected. The parallel-slit setup integrates over dp_x and dp_y, and one obtains

$$n_l(p_z=mc\theta)=\iint\rho_l(\vec{p})dp_x dp_y,$$

m being the mass of the electron. Using the incident positron momentum direction as the axis of quantization, let P_p and P_l be the positron and the lth-electron polarizations ($P=0$, unpolarized particle, $P=\pm1$, completely polarized particle along and opposite the quantization axis). The probability for a spin-singlet overlap with the lth electron is then proportional to $\frac{1}{4}(1\mp P_lP_p)w_l$ and that for a spin-triplet overlap is proportional to $\frac{1}{4}(3\pm P_lP_p)w_l$ where $w_l=\int n_l(p_z)dp_z$; w_l is obviously proportional to $\int|\psi_+(\vec{r})\psi_l(\vec{r})|^2d\vec{r}$. Thus we find that with the superimposed magnetic field $\uparrow$ or $\downarrow$

$$N_{\uparrow,\downarrow}(\theta)=C_{\uparrow,\downarrow}\sum_l(1\mp P_lP_p)n_l(\theta). \qquad (1)$$

It is to be noted that in ferromagnets the excess spin polarization is a negative number in the above notation. We shall return to the constants $C_\uparrow$ and $C_\downarrow$.

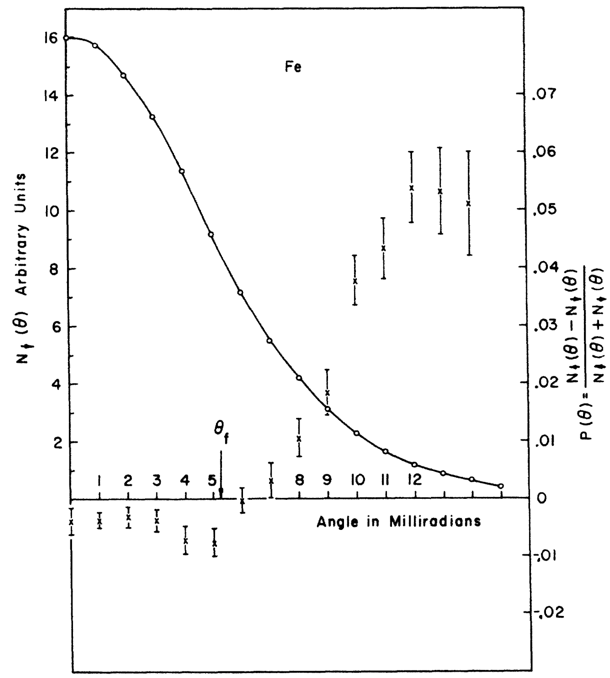

FIG. 1. $N_\downarrow(\theta)$ (left-hand scale) and $p(\theta)$ (right-hand scale) for polycrystalline iron. The statistical errors for $N_\downarrow(\theta)$ are smaller than the circles. The angular resolution is 0.75 mrad. Arrow indicates the angle θ_F corresponding to the free Fermi momentum based on one electron per atom in the conduction band.

In Fig. 1 we have plotted our result $N_\downarrow(\theta)$ for polycrystalline iron and on the same graph

$$p(\theta)=\frac{N_\uparrow(\theta)-N_\downarrow(\theta)}{N_\uparrow(\theta)+N_\downarrow(\theta)}.$$

Our data, when replotted as $N_\uparrow(\theta)-N_\downarrow(\theta)$, are in reasonable agreement with Fig. 1 of Mignarends' and Hambro's recent Letter.[6] They attribute the change of sign in the data to a negatively polarized conduction-band contribution. We shall now show that our model predicts such a change in sign even for an unpolarized conduction band. The ratio of the total 2γ- to 3γ-annihilation probability in the l-electron system is

$$\left(\frac{p_{2\gamma}}{p_{3\gamma}}\right)_{\uparrow,\downarrow}=\frac{\sigma_{2\gamma}}{\sigma_{3\gamma}}\frac{\sum_l(1\mp P_lP_p)w_l}{\sum_l(3\pm P_lP_p)w_l},$$

where $\sigma_{2\gamma}$ and $\sigma_{3\gamma}$ are the 2γ and 3γ annihilation cross sections,[9] and $\sigma_{2\gamma}/\sigma_{3\gamma}=1115$. Using $p_{2\gamma}+p_{3\gamma}=1$ we can obtain

$$\Delta p_{2\gamma}=\frac{p_{2\gamma\uparrow}-p_{2\gamma\downarrow}}{p_{2\gamma\uparrow}+p_{2\gamma\downarrow}}=f(P_lP_p,w_l).$$

The value of the function $f(P_lP_p,w_l)$ stays less than 0.001 for any reasonable value of P_l and w_l; assuming a polarization as large as $P_l=-0.3$ for all electrons and $P_p=0.7$, one obtains $f\approx 0.001$, which is an overestimate. We conclude therefore that $\int N_\uparrow(\theta)d\theta=\int N_\downarrow(\theta)d\theta$ within these limits; physically this means that one can neglect the 3γ annihilation as a competing process. Because of $\sigma_{3\gamma}\ll\sigma_{2\gamma}$ the total 2γ-annihilation probability hardly changes when the field is reversed. We thus obtain the result that $C_\uparrow\neq C_\downarrow$ in Eq. (1); Eq. (1) then becomes

$$N_{\uparrow,\downarrow}(\theta)=\frac{\sum_l(1\mp P_lP_p)n_l(\theta)}{\sum_l(1\mp P_lP_p)w_l}. \qquad (2)$$

Equation (2) will lead to a reversal in sign of $p(\theta)$ even if only d-band polarization is invoked. We find experimentally that the areas under the $N_\uparrow$ and $N_\downarrow$ curves are indeed equal to within 0.2%.

To demonstrate that reversal in sign can come from d-band polarization alone, and to test our model semiquantitatively, we can fit an inverted parabola (corrected for the estimated angular resolution), corresponding to free conduction electrons, to our experimental curve $N_\downarrow(\theta)$ and assume that the rest of the curve is due to $3d$ electrons alone. This of course neglects the small but finite contributions to the high momentum part of $N_\downarrow(\theta)$ from the $3p$ and $3s$ electrons as well as from a more realistic conduction band. An estimate of such contributions has been computed in a previous paper on annihilation in copper and aluminum.[10] Assuming seven $3d$ electrons and one conduction electron per atom ($P_d=-0.31$ at room temperature) we can predict $p(\theta)$ from the experimental $N_\downarrow(\theta)$, assuming an estimated $P_p=0.7$. The predicted $p(\theta)$ is shown in Fig. 2 together with the separation of $N_\downarrow(\theta)$ into the various contributions. It is seen that $p(\theta)$ indeed changes sign for $\theta<\theta_F$ and that one obtains the right order of magnitude for the size of $p(\theta)$ for large angles θ, but that the shape of the experimental curve is not a constant for $\theta>\theta_F$. θ_F is the angle corresponding to the free Fermi momentum based on one electron per atom in the conduction band. One way of interpreting this result is that the tight binding assumption in the d shell is not correct[10]; a broader d band would indeed correspond to what one could describe in the above picture as a θ-dependent $P_d(\theta)$.

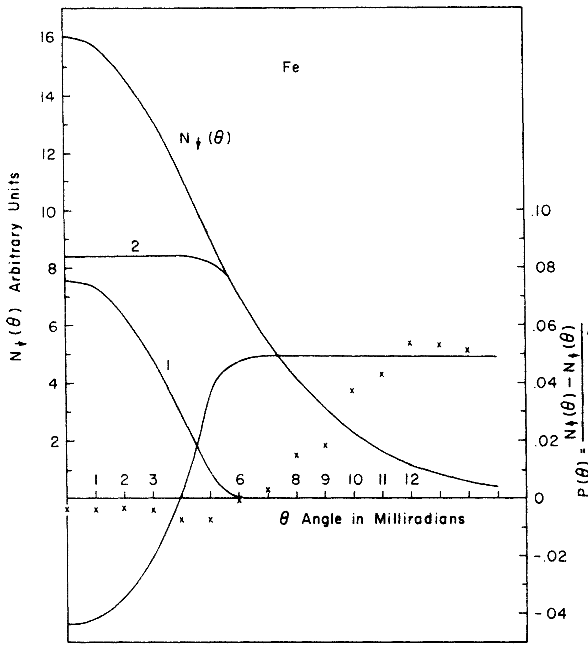

FIG. 2. Decomposition of $N_\downarrow(\theta)$ into a conduction band (1) and a 3*d* band (2) contribution. The predicted $p(\theta)$ is also plotted and the experimental points for $p(\theta)$ from Fig. 1 are reproduced without error bars.

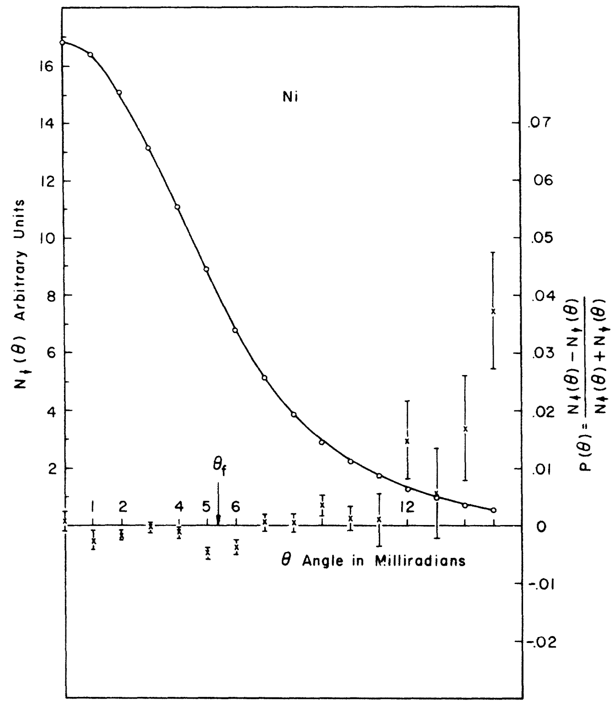

FIG. 3. $N_\downarrow(\theta)$ (left-hand scale) and $p(\theta)$ (right-hand scale) for polycrystalline nickel. The statistical errors for $N_\downarrow(\theta)$ are smaller than the circles. The angular resolution is 2.0 mrad. Arrow indicates the angle θ_F corresponding to the free Fermi momentum based on one electron per atom in the conduction band.

The peaking of the experimental $p(\theta)$ curve just in the Fermi radius region reflected even more in the $n(\theta)$ curve of Mignarends and Hambro for single Fe crystals[6] seems to indicate, however, that one indeed has conduction-band polarization with the excess antiparallel spin of the conduction band being located in that region. A simple model of two momentum spheres for the two spin directions with different radii does indeed predict via Eq. (2) a peaking of $p(\theta)$.

Figure 3 shows our results for polycrystalline nickel. Unlike Hanna and Preston,[3] we find a small polarization effect for nickel which appears, however, only at quite large angles. Again we obtain the reversal in sign and an indication of a peaking in the Fermi radius region. More experiments on single crystals of Ni are presently in progress. Similar measurements in copper gave a null effect for $p(\theta)$ as expected.

We conclude, therefore, that the interpretation of the polarization experiments is somewhat model dependent, and that the reversal in sign of $p(\theta)$ for $\theta < \theta_F$ in itself does not constitute a direct proof of the conduction-band polarization, but that the shape of $p(\theta)$ for $\theta < \theta_F$ is indeed indicative of such an antiparallel conduction-band polarization.

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[1]See, for example, the review article of P. R. Wallace, Solid State Physics (Academic Press, Inc., New York, 1960), Vol. 10, p. 1.

[2]S. S. Hanna and R. S. Preston, Phys. Rev. 106, 1363 (1957).

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[6]P. E. Mijnarends and L. Hambro, Phys. Letters 10, 272 (1964).

[7]S. Berko, Phys. Rev. 128, 2166 (1962).

[8]See, for example, A. J. Dufner and A. V. Bushkovitch, Bull. Am. Phys. Soc. 9, 562 (1964).

[9]A. Ore and J. Powell, Phys. Rev. 75, 1696, 1963 (1948).

[10]S. Berko and J. S. Plaskett, Phys. Rev. 112, 1877 (1958); see also a similar computation used to explain Hanna and Preston's results in iron: W. D. McGlinn, Nuovo Cimento 22, 225 (1961).