SPIN DISTRIBUTION STUDIES IN FERROMAGNETIC METALS BY POLARIZED POSITRON ANNIHILATION EXPERIMENTS (*)

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Résumé. — On a effectué des mesures de polarisation à trois photons $P_{3\gamma}$ à annihilation de positrons, dans Gd, Fe et Ni. On a utilisé les valeurs de $P_{3\gamma}$ pour renormaliser les expériences de corrélation angulaire 2 γ et on a obtenu ainsi la distribution des moments des électrons à spin alignés dans des monocristaux de Gd, Fe et Ni. On trouve des anisotropies importantes dans ces métaux et il semble que dans la région des moments faibles, l'annihilation des positrons se fasse avec des électrons polarisés négativement.

Abstract. — Three photon polarization measurements, $P_{3\gamma}$, were performed with positrons annihilating in Gd, Fe and Ni. The values of $P_{3\gamma}$ are used to renormalize the 2γ angular correlation experiments thus obtaining the momentum distribution of the spin aligned electrons in single crystals of Gd, Fe and Ni. The results show marked anisotropies in these metals, and indicate that in the low *momentum* region the positrons annihilate with negatively polarized electrons.

During the last decade the technique of positron annihilation has been used extensively to study the momentum distribution of electrons in solids. Due to selection rules, spin singlet positron-electron overlaps annihilate via two photons, triplets via three photons. Thus the 2 γ angular distributions obtained from polarized positron annihilation yield information about the momentum distribution of the spin aligned electrons in ferromagnetic materials [1]. In the analysis of such experiments [1, 2], it was noted that these angular distributions could not be interpreted directly in terms of electronic spin distributions in momentum space, but required a renormalization, due to the small 3 γ/2 γ ratio. In this paper we outline a theoretical analysis to obtain the renormalization in a model independent way directly in terms of the experimentally measured 3 γ/2 γ yields ; we present the results of such 3 γ measurements, and apply our proceedure to the 2 γ angular distributions obtained in single crystals of Gd, Fe and Ni.

Let us define

$$\rho^{\uparrow(\downarrow)}(\mathbf{p}) = \sum_{\uparrow(\downarrow)} \left| \int \psi_{\uparrow(\downarrow)}(\mathbf{r})\, \psi_p(\mathbf{r})\, e^{-i\mathbf{p}.\mathbf{r}}\, d^3\mathbf{r} \right|^2 \quad (1)$$

and

$$\omega_{\uparrow(\downarrow)} = \int \rho^{\uparrow(\downarrow)}(\mathbf{p})\, d^3\mathbf{p} \quad (2)$$

where $\psi_{\uparrow(\downarrow)}(\mathbf{r})$ are the electronic wave functions of a ferromagnetic metal with spin parallel (antiparallel) to an applied saturating magnetic field $\mathbf{B}$ ($\downarrow$ is the majority spin direction), and $\psi_p(\mathbf{r})$ is the ground state positron Bloch wave. We introduce a partially polarized positron with net polarization P_p. The total annihilation rates for a positron with spin parallel (antiparallel) to the applied field $\mathbf{B}$ are

$$\lambda^{\uparrow(\downarrow)} = \tfrac{1}{2}\lambda_s\, \omega^{\downarrow(\uparrow)} + \tfrac{1}{2}\lambda_t\, \omega^{\downarrow(\uparrow)} + \lambda_t\, \omega^{\uparrow(\downarrow)}$$

where λ_s and λ_t are the 2 γ and 3 γ partial annihilation rates per electron per unit volume ;

$$\beta = \lambda_t/\lambda_s = 1/1{,}114 .$$

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One obtains for the 2 γ annihilation yield with momentum $\mathbf{p}$, for $\mathbf{B}$ along the positron polarization

$$R_{2\gamma}^{\uparrow}(\mathbf{p}) = \frac{1}{4}(1 + P_p)\,\lambda_s \frac{\rho^{\downarrow}(\mathbf{p})}{\lambda^{\uparrow}} + \frac{1}{4}(1 - P_p)\,\lambda_s \frac{\rho^{\uparrow}(\mathbf{p})}{\lambda^{\downarrow}} \quad (3)$$

$R_{2\gamma}^{\downarrow}$ corresponding to the field $\mathbf{B}$ reversed is given by eq. 3 with the sign of P_p changed. Experimentally one measures an angular distribution corresponding to

$$R_{2\gamma}^{\uparrow(\downarrow)}(p_z) = K \iint R^{\uparrow(\downarrow)}(\mathbf{p})\, dp_x\, dp_y \quad (4)$$

where K is an arbitrary normalizing factor and $p_z = \theta mc$, θ being the angle between the two detectors ($\theta = 7.29$ mrad corresponds to $p_z = 1$ a. u.) (1).

It was found possible to express the physically interesting quantities $\Delta\rho(p_z) = \rho_\uparrow(p_z) - \rho_\downarrow(p_z)$ and $\sum \rho(p_z) = \rho_\uparrow(p_z) + \rho_\downarrow(p_z)$ directly in terms of the measured $\Delta R_{2\gamma}(p_z)$ and $\sum R_{2\gamma}(p_z)$ and the 3 γ effect

$$P_{3\gamma} = \frac{\Delta R_{3\gamma}}{\sum R_{3\gamma}},$$

$$\Delta\rho(p_z) = \frac{N}{P_p}\left[- \Delta R_{2\gamma}(p_z) + P_{3\gamma} \sum R_{2\gamma}(p_z)\right] \quad (5)$$

$$\sum \rho(p_z) = N \sum R_{2\gamma}(p_z) .$$

In the above we neglected terms of the order of β^2 and $\beta P_{3\gamma}$ [4]. N is an arbitrary constant.

Our 3 γ counting apparatus consisting of five NaI(Tl) detectors measuring three-fold coincidences and a sixth placed to measure the 2 γ yield (2). The experiment proved to be rather difficult because of the high singles rates ($\sim 6 \times 10^4$/s) compared to the typical

(1) $\rho^{\uparrow(\downarrow)}$ ($\mathbf{p}$) correspond to the momentum distribution of the spin $\uparrow(\downarrow)$ electrons as sampled by the positron ; they depend on the wave functions of the majority and minority spin bands and have to be distinguished from spin distributions in $\mathbf{k}$ space, since an electron with quantum number $\mathbf{k}$ can contribute at all $\mathbf{p}_l = \mathbf{k} + \mathbf{G}_l$, where the $\mathbf{G}_l$-s are the reciprocal lattice vectors [3].

(2) The counters were distributed on a 10 cm radius circle around the sample, which is saturated by an 18 kGauss field.

total three-fold coincidence rate ($\sim$ 1/s). Our final results [5] for Gd, Fe and Ni are

$$P_{3\gamma}(\mathrm{Gd}) = -0.009\,1 \pm 0.001\,8\,,$$
$$P_{3\gamma}(\mathrm{Fe}) = -0.005\,3 \pm 0.000\,9\,,$$
$$P_{3\gamma}(\mathrm{Ni}) = +0.001\,2 \pm 0.000\,9\ [6].$$

We shall now demonstrate the use of eq. 5 for these three metals.

Gadolinium. Our 2 γ polarization experiments published in an earlier note [7] are plotted in figure 1, along two crystalographic orientations. The

$$P_p(\rho_\uparrow - \rho_\downarrow)$$

curve was obtained from eq. 5 and is also plotted ; $P_p(\rho_\uparrow - \rho_\downarrow)$ exhibits an appreciable anisotropy. Also

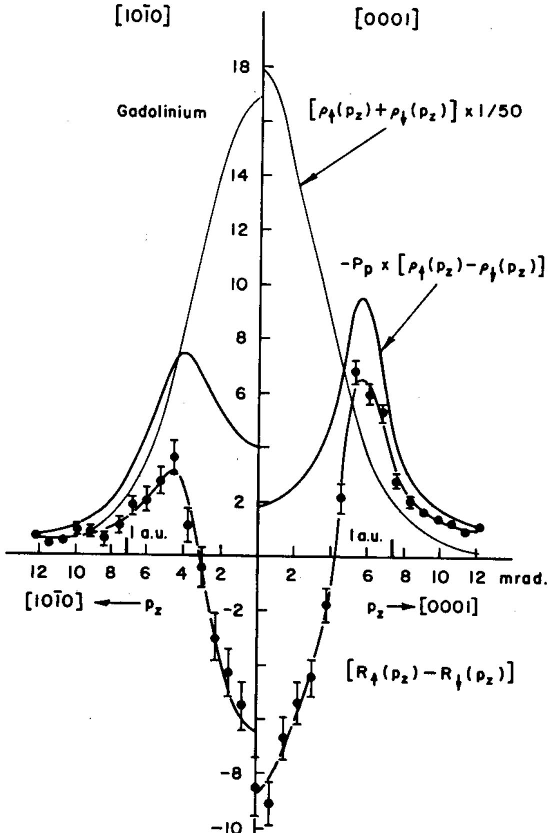

FIG. 1. — The angular distribution, $\Delta R(p_z)$, and the derived momentum distribution of the electron spin excess as seen by the positron in two orientations of Gd.

plotted are the $\rho_\uparrow + \rho_\downarrow$ curves. The anisotropy of the angular distributions in rare earths has been discussed in terms of the Fermi surface of these metals by Williams and Mackintosh [8]. Wigner-Seitz $\psi_p(\mathbf{r})$ computations [3] by S. Cushner in our laboratory indicate that the 4 f electrons contribute only 2.8 % to the total 2 γ annihilation in Gd. Thus $\Delta\rho$ is dominated by the « (5 d) – (6 s) » conduction band. Short of a ferromagnetic band computation including the positron wave function, we can only provide a qualitative discussion of our experimental results. By taking a spherical average of $\Delta\rho(p_z)$ we can convert these one dimensional distributions to a radial distribution by

$$\rho(p) = -\frac{1}{p_z}\frac{\mathrm{d}\rho(p_z)}{\mathrm{d}p_z}.$$

We find $\Delta\rho(p)$ to change sign as a function of p, corresponding to a negative spin polarization ($\Delta\rho > 0$, i. e. minority spin direction) in the low momentum regions of the conduction band, changing over to a positive (majority) spin direction at higher momenta where the 5 d parts of the conduction band predominate.

Iron and nickel. — We have applied eq. 5 to new polarization data from oriented crystals of Fe [9] and Ni, obtained in our laboratory by J. Weingart. In both metals we find appreciable anisotropies in the polarization curves, even though the $\rho_\uparrow(p_z) + \rho_\downarrow(p_z)$ curves show rather small anisotropies. The Ni results were compared to the experiments of Parks and Mihalisin [10]. Although we agree with the general trend, we have not found the fine structure discussed by them. Using the $\Delta R_{2\gamma}(p_z)$ curves and the $P_{3\gamma}$ measurements we compute $\Delta\rho(p_z)$ as for Gd. The results for Fe and Ni are shown in figure 2, in the two direc-

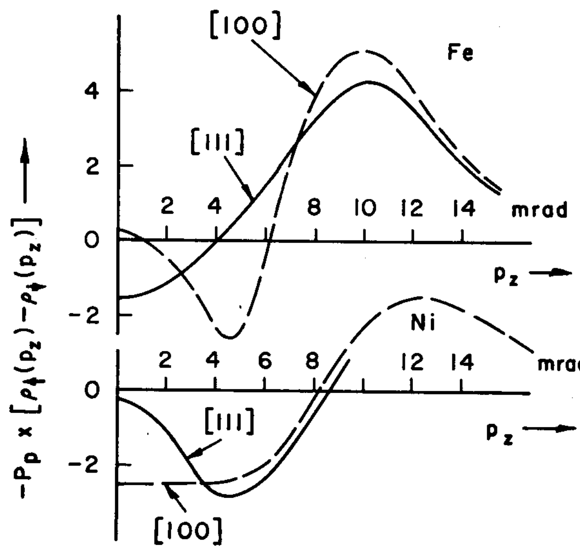

FIG. 2. — The derived momentum distribution of the electron spin excess as seen by the positron in the [111] and [100] directions for Fe and Ni.

tions [100] and [111]. We notice that the anisotropies are opposite in the two metals, i. e. the [111] curve in Ni and the [100] curve in Fe have a clear minimum. Notice also that both metals exhibit net negative regions at low momenta, attributable to negative spin polarization, at least in the region seen by the positron, i. e. in the outer regions of the cell, in agreement with the observations of Shull from polarized neutron data (³). Assuming a rather isotropic « s » distribution (the Ni necks along the [111] direction are too narrow to be observable by our 2 mrad resolution), we attri-

(³) The simple negatively polarized spherical « s » band model used for the early polycristalline data [11] has to be modified since the negative spin regions extend further in momentum space than expected from pure « s » band contributions. As noted previously such negative regions of spin density in p space do not have to be reflected in negative spin distributions in k space, but can arise from different $\psi_\uparrow(\mathbf{r})$ and $\psi_\downarrow(\mathbf{r})$.

bute the anisotropies in the region of low p_z to the « 3 d » band. Indeed, taking the momentum transformed atomic 3 d t_{2g} and e_g type wave functions in the ratios obtained by Shull, we predict anisotropies at $p_z = 0$ in the direction obtained in our experiment. The difference curves $\Delta\rho_{[111]}(p_z) - \Delta\rho_{[100]}(p_z)$ follow, in fact, rather closely the shape of the differences of the theoretical 3 d distributions at low p_z. We thus conclude that our experiments indicate predominantly t_{2g} behaviour for Ni and e_g behaviour for Fe.

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[4] In eq. 5 $P_{3\gamma}$ corresponds to the 3 γ measurements being obtained with the counters in a plane perpendicular to the field B; the formula thus includes the anisotropic 3 γ emission with respect to B.

[5] When these values were used in eq. 5, they were reduced by the measured ratio of 1.5 of P_p in the 3 γ vs. 2 γ experiments.

[6] The value obtained for the 2 γ effect by Sedov (V. L.), *Zh. Eksp. Teor. Fiz.*, 1965, **48**, 1200, is too large on physical grounds, and the more recent values (Sedov (V. L.), Solomatina (L. V.) and Kondrashova (L. A.), *Zh. Eksp. Teor. Fiz.*, 1968, **54**, 1626) should be thought of as upper limits, because of their large quoted errors.

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