Positron annihilation in metal films

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Structural features have been observed in the angular distributions of annihilation γ rays from submicron metal films. These features correlate with the positron work functions of these metals.

The surface layers of materials have recently been attracting progressively increasing interest in solid state physics. Positron-annihilation methods are playing an important role in this research.[1] The primary thrust in the use of positron methods in this field, however, has been either to study the microsurface by a temporal or angular technique in finely dispersed layers with a developed specific surface area or to study the macrosurface, but by the expensive method of positron beams.[2] In the positron method, one observes the energy spectrum or annihilation characteristics of the positrons which are emitted as a result of the backward diffusion from the surface under study ("backward" with respect to the direction of the initial positron beam).

In this letter we report a use of a standard angular-correlation method to study a macrosurface, specifically, to observe the annihilation characteristics of that fraction of the positrons which does not escape from the sample but instead annihilates, being captured in the surface layer of the sample.

In an experiment with a plane-slit geometry, which is used in this case, one of course studies the z component of the momentum of the electrons of the material. The z axis runs perpendicular to the surface under study, which passes through the geometric center of the apparatus and through the slit of the fixed detector.

The test samples consist of a system of a film on a substrate. A "two-layer" model for this system was analyzed in Ref. 3. According to that analysis, the angular distribution of the annihilation γ rays for a sample with a film $[f_{12}(\theta)]$ can be found from the angular distribution of the annihilation γ rays of the substrate $[f_1(\theta)]$ and that of the film material $[f_2(\theta)]$:

$$f_{12}(\theta)=(1-\beta)f_1(\theta)+\beta f_2(\theta),$$

where β is the positron annihilation probability in the film. For thin films this probability is given by

$$\beta=\mu\rho d,$$

where μ is the mass attenuation coefficient of the positron beam, ρ is the mass density of the film, and d is the film thickness.

Measurements of the angular distribution of the annihilation γ rays, which were carried out[3] for samples with copper and aluminum films 6 μm thick, revealed a good agreement with the model in the case of the copper film, but for the aluminum film the measurements were not sufficiently accurate.

In this two-layer model, under the assumptions that all the errors are statistical and that the minimum thickness ($d_{\min}$) which can be studied by the method of angular distribution of annihilation γ rays is determined from the condition

$$\beta = 2\sigma_\beta ,$$

where σ_β is the error in the determination of β, the following expression has been derived for $d_{\min}$:

$$d_{min} = \frac{2}{\mu\rho} \frac{[\sigma_1^2(\theta) + \sigma_{12}^2(\theta)]^{1/2}}{[f_2(\theta) - f_1(\theta)]} ,$$

where $\sigma_1(\theta)$ and $\sigma_{12}(\theta)$ are the errors in the measurement in the angular distributions of the annihilation γ rays of the substrate and of the sample with the film. It follows that at an experimental error $\sim 1\%$ the minimum thickness of a copper film would be 1 μm, and that of an aluminum film 20 μm.

Some methodological refinements have been made in this connection.[4] They have made it possible to reduce the measurement error to 0.1% at the maximum of the angular distribution, so that it has become possible to carry out measurements with submicron films.

In the present experiments we used copper, tantalum, and chromium films 0.3

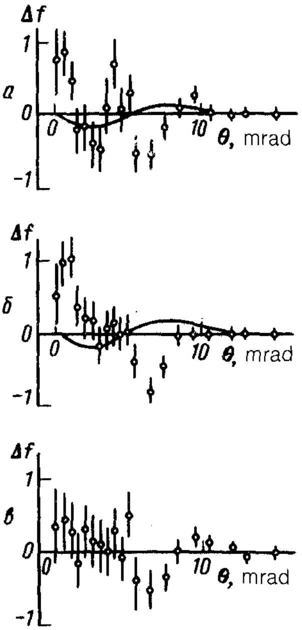

FIG. 1. Difference between the angular distributions of annihilation γ rays for samples with films of (a) copper, (b) tantalum, and (c) chromium, 0.3 μm thick, on a monosilicon substrate with a (100) surface orientation and for the substrate. The solid lines correspond to this difference for the case in which the two-layer model[3] is applicable.

μm thick deposited on a substrate with KĖF-4,5 monosilicon with a (100) surface orientation. A film of silicon oxide 0.5 μm thick was deposited on the surface beforehand to prevent damage to the surface during the deposition of the metal films. The measurements were taken at an angular step of 0.5–1.0 mrad (the angular resolution was 0.7 mrad). The positron sources were 10-mCi sources of the isotope^{22} Na. The measurement error achieved in this experiment was 0.3%.

Figure 1 shows the differences Δf between the angular distributions of the annihilation γ rays for samples with films on the substrate [$f_{12}(\theta)$; a) copper, b) tantalum, c) chromium] and for the substrate [$f_1(\theta)$]. The solid line in this figure shows this difference in the case in which the two-layer model applies. We see that the experimental difference Δf not only disagrees with the theoretical difference but also exhibits some very different types of behavior for the different films. For the copper (positron work function φ_+ of 0.8 eV; Ref. 5), for example, there are two narrow peaks at $\theta = 0$ and 4 mrad. The statistical significance of this separation is quite high (the hypothesis of a statistical scatter in the value of Δf around zero yields $\chi^2 = 37$ with 19 degrees of freedom). For tantalum ($\varphi_+ = 0.0$ eV; Ref. 5), we see only a single peak at $\theta = 0.5$ mrad; for chromium ($\varphi_+ = -2.2$ eV; Ref. 5), we find no structural features at all within the errors.

We are assuming that the appearance of peaks in Δf for copper and tantalum is a consequence of the annihilation of positrons or of positronium atoms at the surface of the metal. The negative positron work function of chromium promotes the emission of positrons into vacuum, thereby promoting their removal from the surface of the metal.

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