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Fermi surface of lithium studied by positron annihilation

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Two-dimensional angular correlation of positron-annihilation radiation in a lithium single crystal has been measured. A continuous determination of the Fermi-surface anisotropy in the $(1\bar{1}0)$ crystallographic plane is obtained for the first time in lithium. The maximum anisotropy of the Fermi surface is found to be $2.8 \pm 0.6\%$.

I. INTRODUCTION

The measurement of the electron momentum density by angular correlation of positron-annihilation radiation has been used for a long time to obtain information about the shape of the Fermi surface (FS) of lithium.[1-3] The interpretation of the experimental data in Li obtained by positron annihilation is far from well established, however, because of the existence of a significant proportion of higher-momentum components in the electron wave function which complicates the analysis. The conventional methods of FS determination are also made difficult by the occurrence of a partial martensitic phase transition at 78 K. By using samples in the form of a dispersion of randomly oriented metallic lithium spheres of micrometer size, Randless and Springford[4] have been able to suppress the martensitic transformation down to liquid-helium temperature and to measure the de Haas–van Alphen effect. These measurements gave the maximum anisotropy of the FS of lithium, but did not allow a detailed determination of the FS geometry. With the development of two-dimensional angular correlation of positron-annihilation radiation (2D-ACPAR), the results obtained by positron annihilation can be directly related to FS dimensions, and the high-momentum components (HMC) can be determined precisely for the alkali metals. A more accurate knowledge of the FS of lithium could be achieved. For the above reasons, a new positron-annihilation experiment in lithium appears to be very interesting. Preliminary results obtained at a temperature below the martensitic transition[5] were already reported.

II. TWO-PHOTON MOMENTUM DISTRIBUTION

The two-phonon momentum density $\rho^{2\gamma}(\mathbf{p})$ for positron annihilation in a solid is given, in the independent-particle model (IPM), by

$$\rho^{2\gamma}(\mathbf{p}) = \sum_{\mathbf{k},l} n_l(\mathbf{k}) \left| \int d\mathbf{r}\, e^{-i\mathbf{p}\cdot\mathbf{r}} \psi_{\mathbf{k},l}(\mathbf{r}) \psi_+(\mathbf{r}) \right|^2 , \quad (1)$$

where $\psi_{\mathbf{k},l}(\mathbf{r})$ and $\psi_+(\mathbf{r})$ are the electron and positron Bloch wave functions and $n_l(\mathbf{k})$ is the occupation number of the state $\mathbf{k}$ in band l. For a periodic potential at zero temperature Eq. (1) reduces to[6]

$$\rho^{2\gamma}(\mathbf{p}) = \sum_{\mathbf{k},l} \sum_{\mathbf{G}} n_l(\mathbf{k}) |A_{\mathbf{k},l}(\mathbf{G})|^2 \delta(\mathbf{p}-\mathbf{k}-\mathbf{G}) , \quad (2)$$

where $A_{\mathbf{k},l}(\mathbf{G})$ are the Fourier coefficients of the positron-electron wave-function product and $\mathbf{G}$ is a reciprocal-lattice vector. A Bloch electron in a state $\mathbf{k}$ can contribute not only at momentum $\mathbf{p}=\mathbf{k}$ but also at higher momentum $\mathbf{p}=\mathbf{k}+\mathbf{G}$, with weights $|A_{\mathbf{k},l}(\mathbf{G})|^2$. Because only the occupied states $\mathbf{k}$ enter in the summation of Eq. (2), the two-photon momentum distribution exhibits breaks at the Fermi momentum $\mathbf{p}=\mathbf{k}$ and also at the various other $\mathbf{p}=\mathbf{k}+\mathbf{G}$. The measured 2D-ACPAR distribution is

$$N(p_x,p_y) \propto \int \rho^{2\gamma}(\mathbf{p})\, dp_z . \quad (3)$$

It contains thus two sets of information. The sharp breaks in $N(p_x,p_y)$ reveal the topology and size of the FS while the shape of $N(p_x,p_y)$ reflects more details of the wave functions of the electron and the positron via the HMC. For lithium, results on the HMC of the electron and the positron wave functions have already been reported.[7]

III. EXPERIMENTAL PROCEDURE AND MEASUREMENT

The single crystal of lithium was grown by a modified Bridgman technique. The method used is similar to those used by Nash and Smith,[8] with some modifications, however. The sample was prepared from lithium with a purity of 99.9%. Preparation of the sample was carried out under the protection of mineral oil. The experiment was done on the bcc phase at 100 K, which is well above the martensitic transition temperature.[9] The single crystal was oriented using transmission Laue x-ray photographs. The orientation was chosen such that the z axis of Eq. (3) is parallel to the $[1\bar{1}0]$ direction.

The 2D-ACPAR distribution was measured with a high-resolution spectrometer using high-density proportional chambers.[10] The angular resolution, including the thermal motion of the positron, was 0.55×0.55 mrad^2. The 2D-ACPAR distribution has been obtained with 86 000 counts at peak, after the procedure of the redundant histogram used to smooth the data.[11]

The contour map of lithium, presented in Fig. 1, shows the nonsphericity of the distribution. In the high-p regions ($p > p_F = 4.3$ mrad) it is due to the HMC while it is due to the FS anisotropy in the central part. The analysis used to measure precisely the anisotropy is described in the next paragraph.

IV. DETERMINATION OF THE FERMI-SURFACE ANISOTROPY

Positions of the breaks in the two-photon momentum density have been obtained by extracting radial distributions

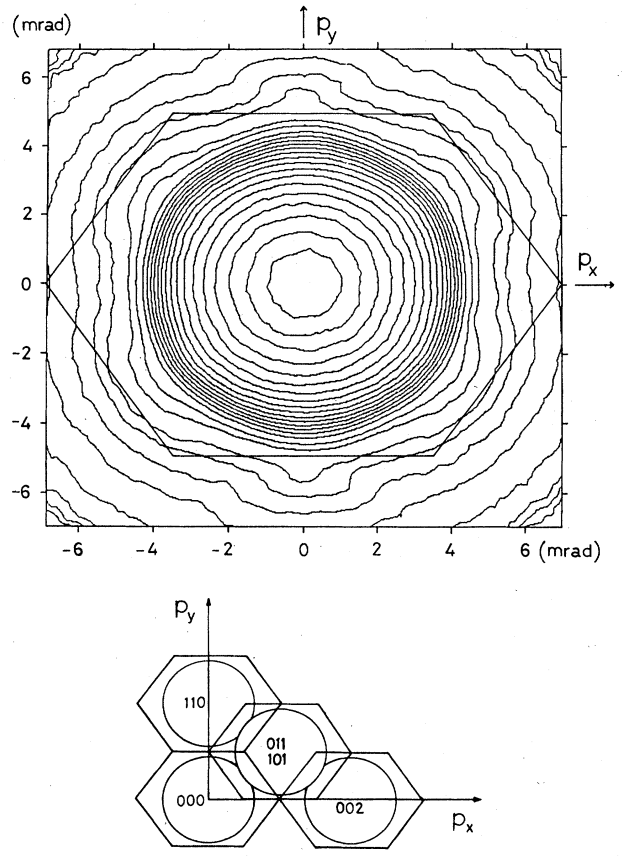

FIG. 1. Contour map of the 2D-ACPAR distribution for lithium in the $(1\bar{1}0)$ plane. p_x is along [001] and p_y along [110]. The inset indicates the projection of the HMC in this plane.

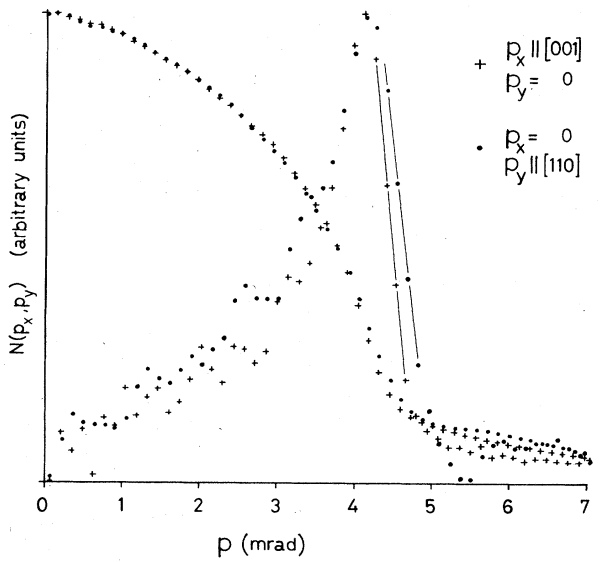

FIG. 2. Radial distributions and their derivatives along two directions in the 2D-ACPAR distribution.

from $N(p_x,p_y)$ in the $(1\bar{1}0)$ plane, and numerically differentiating them. The FS radius has been determined by analyzing the peak of the derivative in the following way. First of all, a model of the 2D-ACPAR distribution, containing several adjustable parameters, was chosen. It is the sum of three contributions.

(i) The central part due to annihilation with conduction electrons is represented by a free-electron sphere of radius k_{F0}.

(ii) The part due to annihilation with core electrons is well represented by a Gaussian fitted along the [100] direction where HMC have no appreciable contribution.

(iii) The contribution of the HMC is based on an empirical model which consists of decreasing exponential curves fitted, for each radial distribution, to the experimental HMC.[7]

The sum of the three contributions has been convoluted with a Gaussian angular resolution function. A nonlinear least-squares fit with the measured 2D-ACPAR is performed to determine the parameters used in the model. A radial distribution has been then calculated from the model and numerically differentiated in order to determine where the Fermi radius lies in the slope of the derivative peak. In the region of interest around the Fermi radius, the absolute value of the derivative curve is a straight line. Its very steep slope depends on the angular resolution. We have found that we can obtain a reliable Fermi radius at 84% of the derivative peak. To test this criterion with a nonspherical FS, we have chosen a phenomenological model of the FS consisting of a sphere with superimposed bumps along the [110] directions, as did Donaghy and Stewart.[1] The adjustable parameters were chosen so that the Fermi radius in the directions [110], [001], and [111] corresponds to the calculation of MacDonald.[12] By replacing the contribution (i) by this quasisphere, we have found that the criterion at 84% is reliable.

We have applied this criterion to the measured 2D-ACPAR distribution in order to determine the radius of the FS of lithium. The two radial distributions shown in Fig. 2 are extracted from $N(p_x,p_y)$ along lines parallel to [110] and [001]. At 84% of the absolute derivative of the curves, we

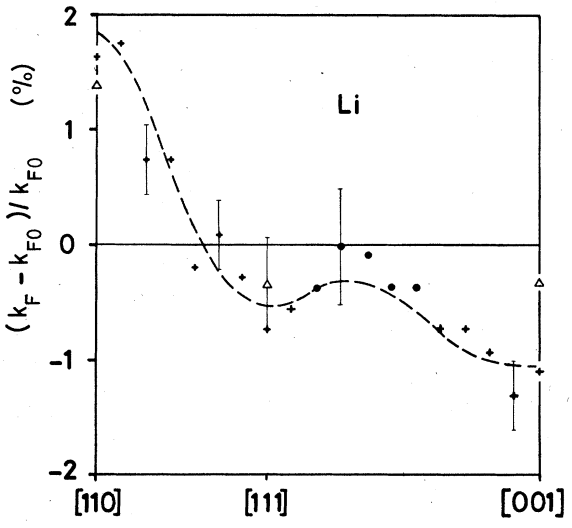

FIG. 3. Fermi-surface distortions in lithium in the $(1\bar{1}0)$ plane. The crosses represent the 2D-ACPAR results. The triangles were obtained by Basinski, Douglas, and Stewart (Ref. 3). The dashed lines correspond to the calculation of MacDonald (Ref. 12).

TABLE I. Experimental FS anisotropies for bcc lithium.

References	$(k_F - k_{FO})/k_{FO}$
1	5%
14	$< 3\%$
2	2.9%
4	$2.6 \pm 0.9\%$
3	$1.75 \pm 0.8\%$
13	$1.7 \pm 0.3\%$
This work	$2.8 \pm 0.6\%$

can directly measure the maximum anisotropy of the FS of lithium. Such radial distributions allow us to determine the Fermi radius for many other directions in the $(1\bar{1}0)$ plane. The results are reported in Fig. 3.

The FS of lithium is distorted from the free-electron sphere by an extension along the [110] directions towards the Brillouin zone boundaries with a compensating depression along the [001] directions. The FS distortions of lithium $(k_F - k_{F0})/k_{F0}$ have been estimated to be $2.8 \pm 0.6\%$. The estimation of 0.6% for the error takes into account the statistical error and the error produced by the data interpolation. As it may be seen in Table I, this result is in good agreement with most of the previous experimental data, except for the positron-annihilation experiment done with the long-slit geometry and for a recent Compton profile experiment done by Schülke, Langer, and Lanzki.[13] The experimental long-slit data of Donaghy and Stewart[1] yield to a FS anisotropy of 5%, but the measurement was subject to some uncertainty in the determination of the anisotropy, due to the HMC. The same remark applied to the work of Basinski, Douglas, and Stewart.[3] The 2D-ACPAR geometry is better suited than the long-slit geometry because less HMC enter in the central part of the distribution $(p < p_F)$ and the slope at the Fermi radius is steeper. Our results are in good agreement with the calculation of MacDonald[12] in which a nonlocal exchange and correlations term are included in the crystal potential. This calculation indicates that the nonlocal corrections in the potential used in the band theory reduce the largest anisotropies calculated with a local potential ($\sim 5\%$) as was first mentioned by Rasolt, Nickerson, and Vosko.[15]

V. DISCUSSION

The method of analysis has also been tested by similar 2D-ACPAR measurements in sodium and potassium single crystals for which the FS is nearly spherical. The FS distortions $(k_F - k_{F0})/k_{F0}$ are less than 0.3% in these metals.[16] The FS radius is found to be very near the free-electron FS radius with a relative error of 0.3% for sodium and of 0.6% for potassium. This estimate lends support to the validity of the results for lithium.

With the chosen orientation for the experiment, the projection of the relevant HMC on the $(1\bar{1}0)$ plane has only a small overlap with the central part $(p < p_F)$, as shown in the inset in Fig. 1. The overlap between the (101) and the (011) HMC with the central part concerns only the FS radius in the direction of overlap and affects only few points in the curve of the FS distortions. These points are indicated by full circles on Fig. 3. From our model of the 2D-ACPAR distribution, we have estimated that the effect of the HMC could give an error of $\pm 0.2\%$ on the Fermi radius. We would like to point out that the maximum anisotropy of the FS is not affected by the effect of the HMC but could be affected by the slow fall in momentum density.

An implicit assumption in the analysis is that the two-photon momentum density $\rho^{2\gamma}(\mathbf{p})$ is constant up to the Fermi momentum. The two-photon momentum density decreases near the Fermi momentum as shown by the calculation of Rabou and Mijnarends.[17] The analysis of the FS anisotropy in Li could be affected by the slow fall in momentum density, especially in the [110] direction. Such an effect can reduce the determination of the FS radius by at most 0.2%. However, it is important to observe that the effect of positron-electron correlations could eliminate and perhaps reverse the decrease of the momentum density.

The effect of positron-electron correlations has been included in the model by multiplying the momentum density of the free-electron gas by the enhancement factor described in the form used by Kahana:[18]

$$\rho^{2\gamma}(\mathbf{p}) = \text{const} \times \sum_{k \leq k_F} \delta(\mathbf{p} - \mathbf{k}) \epsilon(p) \ ,$$

where

$$\epsilon(p) = a + b(p/p_F)^2 + c(p/p_F)^4 \ .$$

The coefficients a,b,c were obtained by interpolating Kahana's data for the proper electron density. Another possibility is to analyze the data with $\rho^{2\gamma}(\mathbf{p})$ calculated by a band-structure method in order to obtain an experimental value for the enhancement factor.[19] The value found for the enhancement factor (see Ref. 19) is lower than that calculated by Kahana but the momentum enhancement could be masked by a compensating effect due to the large anisotropy of the FS ($\sim 5\%$) resulting from the band structure calculated with a local potential. The conclusion of this work[19] is that the Kahana-like theories describe reasonably well the position-electron many-body effects in the alkali metals. Nearly no effect has been observed on the position of the Fermi radius obtained with the analysis described in Sec. IV, after having taken into account the enhancement factor. The criterion still remains unchanged.

The core-electron contribution is also affected by the positron-electron many-body interactions. We have tried to determine the many-body enhancement factor for core electrons in comparing the experimental core contribution with the once-integrated core momentum density calculated using IPM and renormalized free-atom core orbital wave function. This calculation produces a too broad distribution in comparison with the experimental core distribution. Therefore, a constant enhancement for core electrons like that used in aluminium[20] is not sufficient to describe the core distribution.

VI. CONCLUSIONS

The development of 2D-ACPAR has enabled new positron annihilation measurements to be made on lithium single crystals and has allowed us to measure the FS geometry. Our results are the first continuous determination of the FS anisotropies in one crystallographic plane. From the analysis, we concluded that the maximum anisotropy of the FS is $2.8 \pm 0.6\%$, which is in very good agreement with the calculation of MacDonald[12] and the de Haas–van Alphen measurements of Randles and Springford.[4]

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