INVESTIGATION OF THE FINE DETAILS IN THE FERMI SURFACE OF COPPER BY THE METHOD OF ANGULAR CORRELATION OF ANNIHILATION QUANTUM FLIGHT PATHS

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The angular correlation curves of annihilation quanta in single-crystalline Cu samples of [100], [110] and [111] orientations have been measured. The measurements yielded the distributions of momentum components of conduction electrons in the examined crystallographic directions. These in turn are compared with distributions calculated theoretically on the basis of Fermi surface models for copper proposed by Pippard, and Zornberg and Mueller. Our results are in better agreement with the Zornberg-Mueller model of the Fermi surface of Cu which accounts for deviations from a sphere in its central part.

1. Introduction

It is well known that the angular distribution of the paths of annihilation quanta supplies information about the electronic structure of metals and in the case of oriented crystals some data on their Fermi surface. The origin of such information is the simple relationship between the angle α, which is a measure of the angular deviation from co-

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linearity of the flight paths of the two photons produced in annihilation, and the momentum p of the electron-positron pair.

$$\alpha = \frac{p}{mc} \tag{1}$$

where m is the mass of the electron. In turn, the probability of observing a specific angle, $N(\alpha)$, between two photons depends on the Fourier transform of the product on the electron and positron wave functions $\Psi_-(\vec{r})$ and $\Psi_+(\vec{r})$ [1],

$$N(\alpha) = |\int e^{-i\vec{p}\vec{r}} \Psi_-(\vec{r}) \Psi_+(\vec{r}) d\vec{r}|^2. \tag{2}$$

Hence, angular correlation measurements of the directions of annihilation quantum flight paths provides a means of finding electron momentum distributions in the examined crystals.

The method of positon annihilation applied to a number of single crystals of metals has revealed the distribution of conduction electron momenta to be anisotropic. Owing to the rather simple shape of the Fermi surface, which is almost spherical, copper has been the metal most frequently used in experiments. Berko and Plaskett [1] found there are slight differences in the angular correlation curves for the three principal directions [111], [110] and [100]. Fujiwara [2] obtained an angular correlation curve in the [111] direction which in its central part exhibited a departure from the parabolic shape, thereby confirming the non-sphericity of the Fermi surface of copper. Continuing his research with Sueoka [3] with the use of a very strong positron source, he determined the radius of the Fermi surface in the [111] direction; within experimental accuracy the result was identical with the distance from the center to the Brillouin zone boundary in this crystallographic direction. These researchers were also able to determine the radius of the neck which is a characteristic element of copper's Fermi surface. A somewhat different method of studying the topography of the Fermi surface of copper was applied by Sueoka [4] and Williams *et al.* [5]. They modified the conventional method of measurements, in which the single-crystalline sample is immobile an the rotating annihilation photon detector changes its angular position, into an arrangement in which the single crystal rotates about a chosen crystallographic axis and the detectors are at rest. This allowed the contour of the Fermi surface cross-section normal to the given direction to be investigated. Berko, Cushner and Erskine [6] compared the differences in angular correlation curves for several crystallographic orientations of copper with those predicted by theory; their measurements were found to be compatible with theory.

In all of the studies mentioned the scope of information about copper's Fermi surface was restricted by the number of crystallographic directions in which the angular correlation curves were measured. Mijnarends [7] proposed in 1967 a method of elaborating results which allows the distribution of electron momenta over any cross-section of the Fermi surface to be obtained with utilization of angular correlation curves from a finite number of crystallographic directions. It is obvious that the accuracy of the method is better when more correlation curves are available. With this method Mijnarends

[7] acquired fairly good results for a simple spherical distribution of momenta in a fictitious crystal assumed for the reasoning. As regards the case of copper, which has a non-spherical Fermi surface, the results were less satisfactory, doubtlessly being affected by the too small number of angular correlation curves used, together with the rather poor resolving power of the annihilation spectrometer employed in the measurements [8].

The goal of this work is to compare angular correlation curves obtained from original measurements with oriented single-crystalline samples of copper with curves calculated theoretically on the basis of the most up-to-date models of Fermi surface of copper which account for fine deformations of its central spherical section.

2. *Investigating the Fermi surface by the method of angular correlation of the directions of annihilation quanta*

Information about the distribution of momenta of conduction electrons is immediately available from angular correlation curves by using a simple model of pair annihilation in free collisions, which images reality pretty well. In the model, the positrons are entirely thermalized in the solid prior to annihilation (their mean thermal energy is much lower than the mean energy of annihilating electrons), the probability of annihilation is independent of electron momentum, and the momentum of the annihilating pair is not perturbed by the environment. Angular correlation curves obtained experimentally have to be corrected for annihilation processes between positrons and electrons bound in the atomic core and sometimes for the finite resolving power of the measuring device.

The counting rate of $\gamma-\gamma$ coincidences, $N(\Theta)$, within the angle interval Θ, $\Theta+d\Theta$ is proportional to the rate of annihilation, $N(p_z)$, of pairs having a z-component of momentum in the interval p_z, p_z+dp_z. This statement follows from relation (1) between the angle Θ and momentum component p_z,

$$\Theta = \frac{p_z}{mc}. \tag{3}$$

With the assumption of complete thermalization of positrons the rate of annihilation of pairs having a momentum component in the interval p_z, p_z+dp_z is, in turn, proportional to the number of conduction electrons having such momentum components. Hence, for any Fermi surface it is proportional to the number of occupied electron states contained in the volume of a slice dp_z thick cut at position p_z with section surfaces perpendicular to the direction of $\vec{p}_z$ (Fig. 1).

This geometrical interpretation is valid when the annihilation spectrometer has a resolution function $R(p_y)$ in the p_y direction broad enough to comprise the whole momentum component range in this direction and, at the same time, an adequately narrow resolution function $R(p_z)$ in the p_z direction (Fig. 1). This is an example of a spectrometer with narrow slits which was used in the measurements in the present work.

The rate of measured coincidences for any angle Θ is then related with the number of occupied states dS contained in the volume of the slice $dV(p_z)$ as

$$N(\Theta)d\Theta \sim N(p_z)dp_z = \text{const} \cdot ds \tag{4}$$

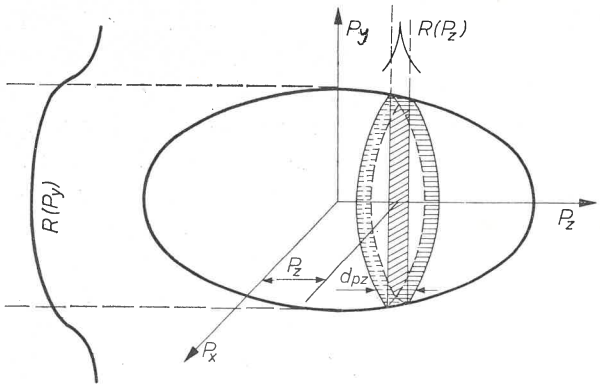

Fig. 1. Principle of measurent by the method of angular correlation of annihilation quantum directions. The measuring instrument cuts a slice (shaded section) out of the Fermi surface of thickness dp_z. The horizontal and vertical resolution functions are also shown

at a constant density of states within that slice

$$N(\Theta)d\Theta \sim N(p_z)dp_z = \text{const} \cdot dV(p_z) \tag{5}$$

or

$$N(\Theta)d\Theta \sim N(p_z)dp_z = \text{const}\, P(p_z)dp_z \tag{6}$$

where $P(p_z)$ is the area of the section of the Fermi surface normal to $\vec{p}_z$ at the point p_z. In particular, when the Fermi surface is a sphere of constant density of states (a gas of absolutely free electrons) and radius p_F, the expressions (5) and (6) take the form

$$N(\Theta)d\Theta = \text{const}\left(1-\frac{\Theta^2}{\Theta_F^2}\right) \quad \text{for} \quad (\Theta) \leqslant \Theta_F$$

or

$$N(p_z)dp_z = \text{const}\left(1-\frac{p_z^2}{p_F^2}\right) \quad \text{for} \quad (p_z) \leqslant p_F\,. \tag{7}$$

The shape of the angular correlation curve for a spherical Fermi surface is, in agreement with Eq. (7), a reversed parabola. For non-spherical Fermi surfaces there are deviations from the parabolic shape of the angular correlation curve which are a measure of the non-sphericity in the given crystallographic direction.

The shape of the angular correlation curve is affected by deviations from the simple annihilation model accepted at the beginning of this section, and by the interaction of positrons with electron gas and periodic potential of the lattice. When analyzing angular correlation curves it is of prime importance to make a proper division of the momentum distribution into parts corresponding to annihilation with conduction electrons and bound electrons in the atomic cores.

It follows, hence, that the method of angular correlation of the flight paths of annihilation quanta with the use of a spectrometer having narrow slits lets us reproduce the field

distribution in Fermi surface cross-sections normal to a given crystallographic direction, assuming that the density of states is constant, or the distribution of the number of states in a given direction integrated with respect to the two remaining directions.

3. The Fermi surface of copper

Copper crystallizes in the face-centered cubic system. The lattice parameter at 0 K is $3 \cdot 603 \times 10^{-10}$ m. Its Fermi surface, investigated for the first time by Pippard [9] by the anomalous skin-effect method, is spherical with eight necks stretching out in the direction of the hexagonal planes of the Brillouin zone (in the [111] direction, see Fig. 3). A lot of information about the shape of the Fermi surface became available thanks to

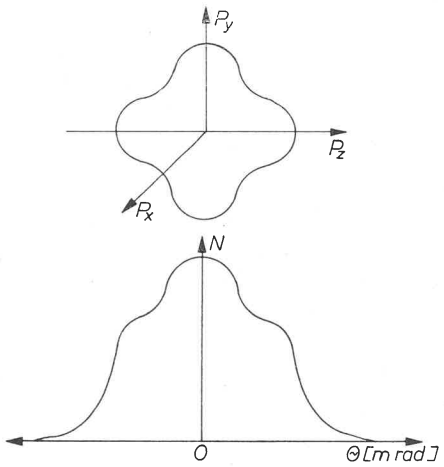

Fig. 2. Expected shape of angular correlation curve in the case of a non-spherical Fermi surface, shown at top

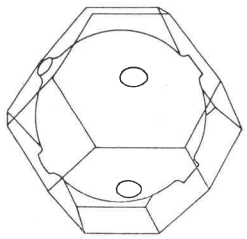

Fig. 3. Shape of Fermi surface of copper within first Brillouin zone

measurements carried out by the de Haas van Alphen method. They corroborated the general features of the model proposed by Pippard and added some new fine details in its shape.

Measurements by Shoenberg [10, 11] revealed some slight deviations from ideal sphericity in the central section of the Fermi surface. Analyzing the results of these measurements, Roaf [12] proposed a modified model incorporating a deformed sphere having protuberances in the [100] direction and identations in the [100] direction.

The fine details in the Fermi surface of copper were subsequently studied by Joseph *et al.* [13], who used a much improved experimental technique. On the basis of their results, Zornberg and Mueller [14] reproduced this surface, computing, among other things, the relative changes in radius k_F along various crystallographic directions. The outcome of their calculations are presented in Figs 4 and 5. The first figure illustrates the changes in the ratio k_F/k_s (where k_s is the radius of the spherical Fermi surface calculated

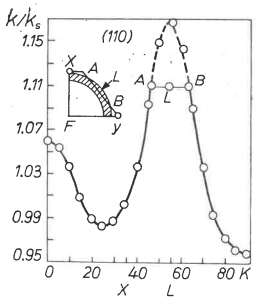

Fig. 4

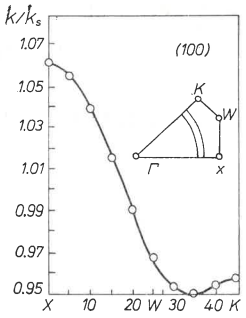

Fig. 5

Fig. 4. Changes in k_F/k_s ratio against angle measured from [100] direction in (110) plane (after Zornberg and Mueller [14])

Fig. 5. Changes in k_F/k_s ratio against angle measured from [100] direction in (100) plane (after Zornberg and Mueller [14])

in the approximation of almots free electrons) against the angle measured from the [100] direction in the (110) plane. The upper part of the figure shows the Brillouin zone cross-section with symmetry points indicated. In like fashion, Fig. 5 gives the dependence of k_F/k_s on angle measured from the [100] direction in the (100) plane. It follows

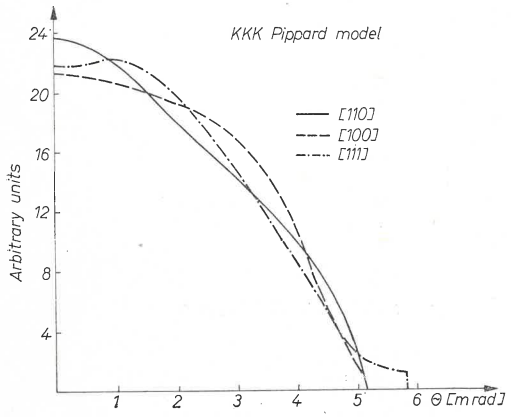

Fig. 6

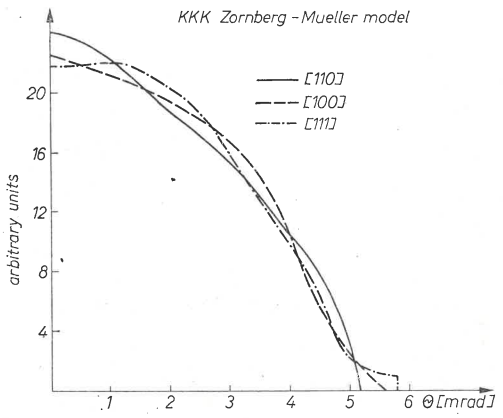

Fig. 7

Fig. 6. Angular correlation curves calculated on the basis of the Fermi surface model for copper proposed by Pippard

Fig. 7. Angular correlation curves calculated on the basis of the Fermi surface model for copper proposed by Zornberg and Mueller

from Figs 4 and 5 that the radius of the Fermi surface in the [100] direction is larger by about 6 per cent than k_s. In Fig. 4 a small indentation of about two per cent is observed in the (100) plane for an angle near 25°, whereas in the (100) plane there is an indentation of approx. 5 per cent at an angle of 35°.

The results of our measurements carried out by the method of angular correlation of the flight paths of annihilation quanta are compared in subsequent sections of this paper with the spherical model of Pippard and the model of Zornberg and Mueller which introduces the aforementioned slight deviations from a sphere. For this purpose these models were used for theoretically computing the expected angular correlation curves for the [100], [110] and [111] directions. They are then compared with the experimental curves measured in this work. Fig. 6 depicts the computed distributions from the Pippard model, whereas Fig. 7 from the model of Zornberg and Mueller.

4. Measuring arrangement

The angular correlation curves were measured by means of a two-channel scintillation spectrometer designed for analyzing the gamma radiation emitted during two-quantum annihilation of electron-positron pairs. The block diagram of the spectrometer is shown in Fig. 8. The radiation detectors were NaI(Tl) scintillators operating together with FEU-29

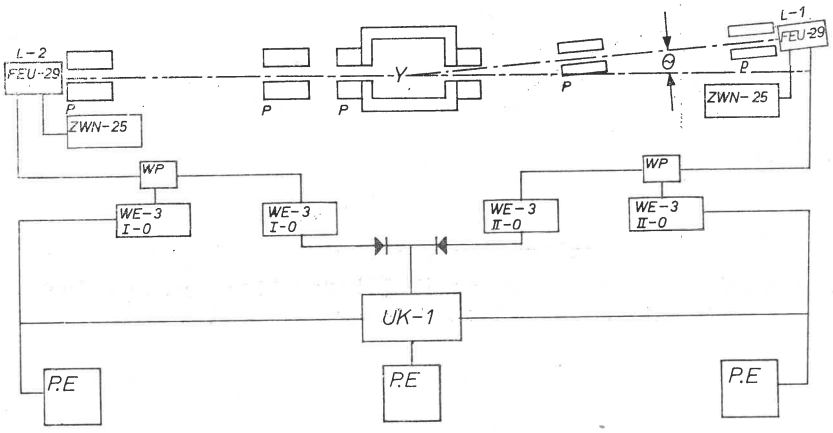

Fig. 8. Block diagram of two-channel scintillation spectrometer

photomultipliers. The pulses from the counters, after being amplified current-wise, were fed to four WE-3 universal input units. The units labelled in Fig. 8 as ID and IID formed the lower threshold of the amplitude analyzer gates. The standard pulses from the outputs of these units were then transferred to the coincidence inputs of a UK-1 coincidence-anticoincidence circuit. The input units IG and IIG, on the other hand, were the upper gate thesholds, and the pulses from them were led to the anticoincidence inputs of the UK-1 circuit. Such co-operation of the various elements of the arrangement ensured that only those pulses arrived at the UK-1 output which originated from gamma radiation falling within the energy interval of the 0.51 MeV annihilation line in which we were interested.

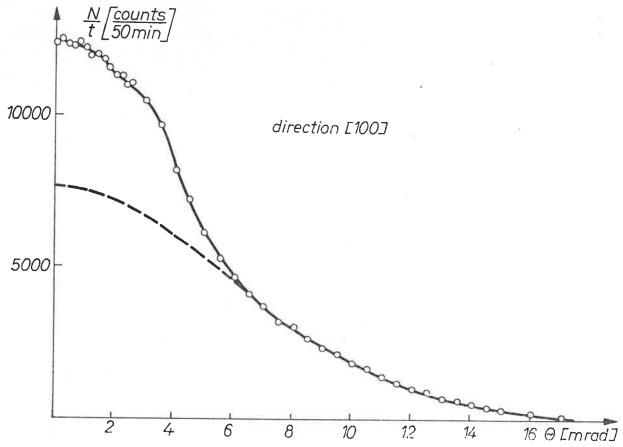

Fig. 9. Experimental angular correlation curve in [100] direction. The dashed line is the Gaussian curve fitted to the tail end of the experimental curve

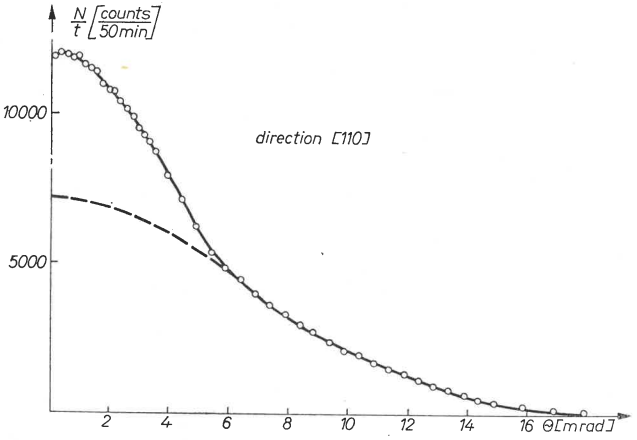

Fig. 10. Experimental angular correlation curve in [110] direction. The dashed line is the Gaussian curve fitted to the tail end of the experimental curve

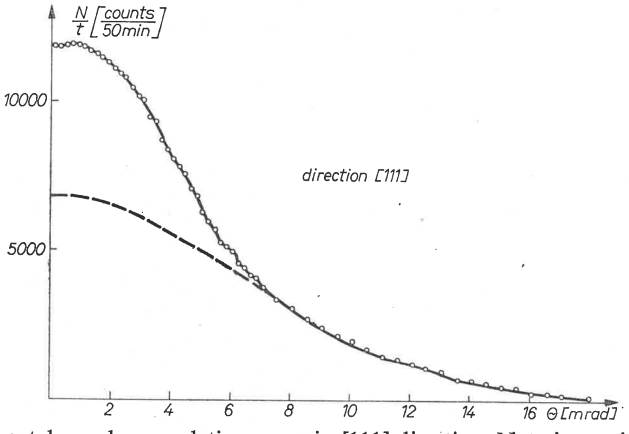

Fig. 11. Experimental angular correlation curve in [111] direction. Notation as in Figs 9 and 10

A system of lead shields cut out a beam of annihilation radiation of definite width. The distance from the center to each of the detectors was 2 m. The angle was altered by rotating the counter L-1 together with its shields about an axis passing through the center of the sample and perpendicular to the plane of rotation of the counter. Counter L-2 was immobile. The position of the counter L-2 with respect to the main spectrometer axis was read by means of accurate angle-gauges with an error of less than 0.02 mrad. The single-crystalline copper samples of [100], [110] and [111] orientations were produced at the Special Experimental Apparatus Shop of the Institute of Nuclear Research in Świerk near Warsaw. The purity of the samples was not worse than 99.9%, and the accuracy of their crystallographic orientation was 0.5°. They were in the form of cuboidal strips of the dimensions $1 \times 10 \times 15$ mm^3. The positron source was about 10 mCi of the isotope ^{22}Na. The half-width of the horizontal resolution function $R(p_z)$ was 0.84 mrad.

5. *Results, their interpretation and discussion*

The difference in angular distributions obtained for copper in different crystallographic directions stem above all from the non-sphericity of the Fermi surface. There is also a slight effect giving rise to such differences due to interactions between a positron and lattice potential, and electron-electron and electron-positron interactions. The data on the non-sphericity of copper's Fermi surface which will be later compared with its newest models had been obtained by measuring the angular correlation curves $N(\Theta)$ in three crystallographic directions, [100], [110] and [111]. The curves obtained in the measurements are shown respectively in Figs 9, 10 and 11. The time for each measurement at one angle Θ was 50 min. The number of coincidences for the angle $\Theta = 0$ was about 12000 against a background of about 400 coincidences in 50 min. After subtracting the background, the angular correlation curves were symmetrized by the "center of gravity" method. The vital operation in annihilation analyses by the angular correlation method is the proper discrimination of acts of annihilation with conduction electrons from the space within the Fermi surface having momenta not greater than p_F from acts of annihilation with electrons bound in the atomic cores. It may be accepted after the authors of Refs [1, 11, 14] that the responsibility of annihilation with momenta greater than the Fermi momentum primarily falls on atomic core electrons and to a much smaller extent on free electrons having high momentum components due to electron-phonon interactions. The participation of the latter was extensively discussed in Refs [1, 8], whereas the experimental distribution of these components for copper can be found in Ref. [15]. An important effect of the momentum components on the shape of angular correlation curves has been observed for all directions within the angle range from 4 mrad to 8 mrad, but it is largest for the [110] direction. In measurements in which the statistical error is large it is impossible to ascertain whether or not there is anisotropy in the distribution of high momenta, because the amplitude of oscillations does not exceed 2% [15]. The conditions under which our experiments were carried out allow us to omit the effect of anisotropy in the high momentum component distribution on the shape of the "tails" in the angular correlation curves obtained experimentally. This in turn lets us accept the Gaussian

curve best fitted to the tail end of the angular correlation curve as a good approximation of the annihilation momentum distribution of atomic core electrons. Such an approximation is used very often in research on the electronic structure of metals in the liquid and solid phases. It is particularly well substantiated in the papers by Arias-Limenta and Varlashkin [16], who compared the angular distributions for Rb and Cs in the solid and liquid states with respective distributions for liquid Kr and Xe. It was found that at room temperature the angular distributions for liquid Kr and Xe excellently coincide with the Gaussian curves fitted to the tail ends of the angular correlation curves of solid Rb and Cs. Atoms of liquid Kr and Xe are analogues of the atomic cores of Rb and Cs.

In accordance with the procedure described, the parameters of the Gaussian curves qest fitted to the tail ends of each angular correlation curve obtained from the measurements were determined. Figures 9, 10 and 11 show the Gaussian curves inscribed to the distribution curves found experimentally in the three crystallographic directions. The Gaussian curves, therefore, represent the acts of annihilation with the bound electrons of atomic cores. The angular correlation distributions corresponding solely to annihilation

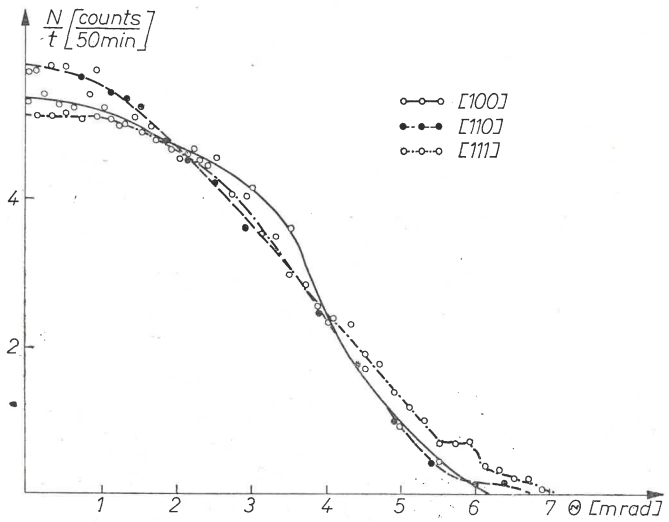

Fig. 12. Experimental angular correlation curves in the [100], [110] and [111] directions after subtraction of Gaussian curves and normalization to a common area

with conduction electrons were obtained by subtracting the Gaussian curves from the measured angular correlation curves. All of the curves corresponding to annihilation with conduction electrons were normalized to a single area and are presented in Fig. 12.

A comparison of the angular correlation curves, Fig. 12, obtained thus with curves computed according to Pippard's model (Fig. 6) and that of Zornberg and Mueller (Fig. 7) corroborates the overall topography of the Fermi surface of copper, *i.e.* it consists of a spheroidal central part with eight proturbances extending along the directions of the hexagonal planes of the first Brillouin zone. The course of the curves is particularly concordant with the Zornberg-Mueller model in the range of angles up to 2 mrad. All curves

intersect in the vicinity of 4 mrad, and in the [111] direction there is a characteristic indentation corresponding to the neck in the Fermi surface tangent with the Brillouin zone boundary in the neighbourhood of an angle of 5.8 mrad.

The height ratios were calculated for the various pairs of angular correlation curves found for two different crystallographic directions for an angle $\Theta = 0$, *viz.*, $\frac{h\,[110]}{h\,[100]}$, $\frac{h\,[110]}{h\,[111]}$ and $\frac{h\,[100]}{h\,[111]}$. This was done both for the curves acquired experimentally and those theoretically from both of the models under consideration here. The data obtained are given in Table I.

TABLE I

Height ratio	Theor. curves from Z–M model	Theor. curves from Pippard model	Experimental curves
$\frac{h\,[110]}{h\,[100]}$	1.07	1.11	1.07
$\frac{h\,[110]}{h\,[111]}$	1.11	1.08	1.11
$\frac{h\,[100]}{h\,[111]}$	1.03	0.97	1.04

The data in Table I demonstrate excellent agreement of results with the Zornberg-Mueller model, at the same time exposing the rather large deviations from data obtained with the Pippard model. This is the first, very important circumstance which speaks in favour of the Fermi surface model of copper calculated by Zornberg and Mueller.

In order to have a closer look at the fine structure suggested by this model in the shape of the Fermi surface, every angular correlation curve for a given crystallographic direction was compared with the analogous curves obtained from both models theoretically for the same direction (Figs 13, 14, 15).

For the [100] and [110] directions the curves from both models differ only slightly and approximate the experimental curves very well. Notwithstanding, as regards the character of their shape, the experimental curves appear to be more like those obtained by means of the Zornberg-Mueller model. Likewise for the [111] direction, for which the experimental curves differ more from the model curves, the comparison distinctly speaks in favour of the same model. The conclusions reached from the comparison of the curves and the analysis of the height ratios for the various pairs of angular correlation curves point to the Zornberg-Mueller model, which features the fine deformations in the spherical section of the Fermi surface, as the one corresponding better to reality.

All of the experimental curves (Figs 13, 14 and 15) become diffuse beyond the Fermi momentum, whereas for lower values of momentum they generally run under the theoretical curves. This may stem above all from the way in which curves are presented, for both the

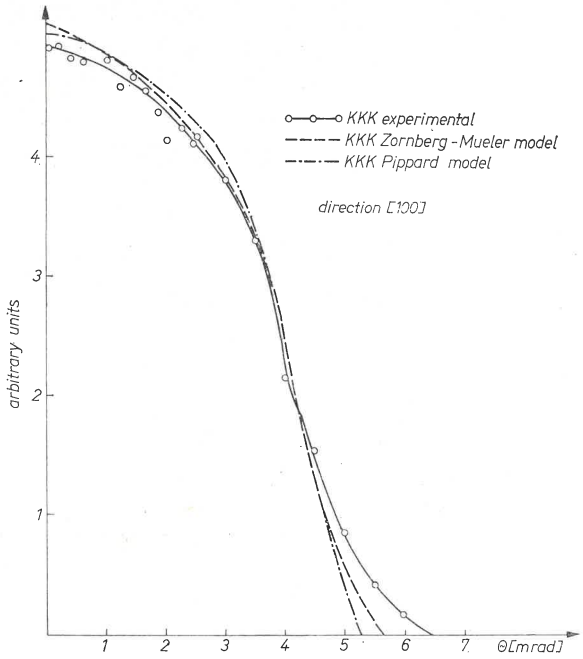

Fig. 13. Comparison of angular correlation curves found theoretically with experimental curve in [100 direction

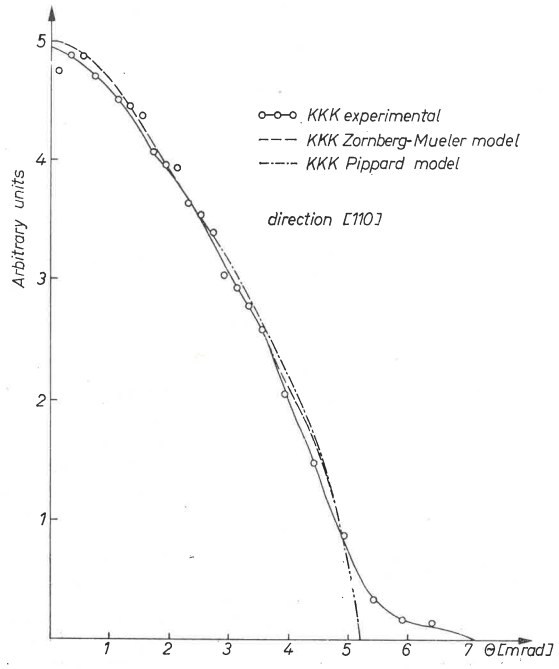

Fig. 14. Comparison of angular correlation curves found theoretically with experimetal curve in [100] direction

theoretical and experimental curves had been normalized to a common area under them. In the individual cases other factors may also tend to shape the curves as they are. For the curves in the [110] and [100] directions it is seen that the experimental ones run distinctly below the theoretical ones within the angle range from 3.5 to 4.5 mrad. This, together with their diffuseness, seems to indicate the occurrence of strong electron-phonon

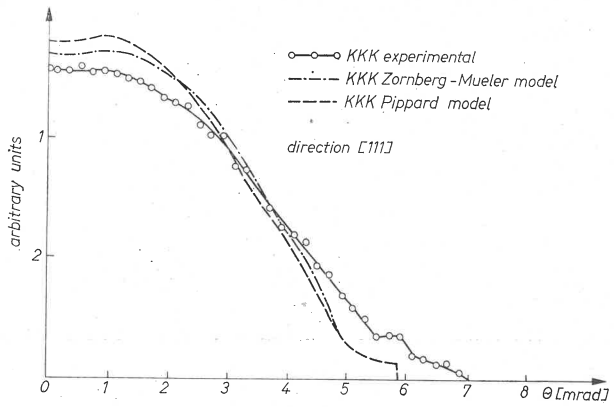

Fig. 15. Comparison of angular correlation curves found theoretically with experimental curve in [111] direction

interactions for the electrons having momenta corresponding to this angle range. In consequence, this would decrease the Fermi surface cross-section and the participation of these electrons in the high momentum component effect.

The relatively high diffuseness of the curve in the [110] direction may be due to, after the authors of Ref. [15], the existence of $s-d$ electron hybridization. For the [111] direction the exceptional run of the experimental curve above the theoretical ones for angles less than Θ_F stems primarily from the finite resolving power of the equipment used, while for angles bigger than Θ_F from the character of the region it describes. Namely, this is the region of tangency between the Fermi surface and the boundary of the Brillouin zone.

6. Conclusions

The presented results of investigations on the Fermi surface of copper lead to the conclusion that the method of angular correlation of the flight path directions of annihilation quanta permits to rather accurately verify the models of such surfaces, check their shapes, dimensions and position with respect to the Brillouin zones. The comparison of the angular correlation curves for copper with those obtained from the Pippard and Zornberg-Mueller models speaks in favour of the latter model and at the same time indicates the far-reaching possibilities of the applied method for investigating the fine details occurring in the topography of these surfaces.

The mode of separating the angular correlation curves into parts corresponding to annihilation with conduction electrons and that with electrons bound in atomic cores used here is the only method at present which leads to satisfactory results. Nevertheless, it should be kept in mind that the Gaussian distribution ascribed to annihilation with the bound electrons is an approximate formula which may prove to be better or worse for different metals. Theoretical attempts to compute these distributions, initiated in 1956 by Ferrel [18], still lead to results which are not better than the Gaussian approximation employed in this study.

The subtraction of the Gaussian distributions from the experimentally obtained angular correlation curves was done before the distributions were normalized to a common area. The latter operation was performed after the angular correlation curves for annihilation with conduction electrons were obtained. The sequence of operations accepted thus is at the same time equivalent to resigning from the very frequent assumption about the isotropy of a crystal as regards annihilation with atomic core electrons. The results obtained here show a distinct anisotropy in this annihilation, as had been observed earlier in Refs [4, 15]. Numerically it is expressed for copper as given in Table II.

TABLE II

Crystallographic direction	Share of bound electrons in the annihilation, %
(100)	73
(110)	77
(111)	72

Investigations of the fine details in the Fermi surface by the angular correlation method applied to annihilation directions require annihilation spectrometers of especially excellent resolving powers. In our scientific centers researches are now being carried out on the Fermi, surfaces of a number of metals, including the remaining two noble metals, with spectrometers of much improved resolving powers.

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