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FERMI SURFACES OF GOLD AND SILVER FROM ULTRASONIC ATTENUATION*

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Recent experiments have confirmed that the Fermi surface in copper contacts the Brillouin zone boundary in the [111] direction as suggested by Pippard.[1-5] Using the low-temperature magnetic dependence of ultrasonic attenuation, we have found that the Fermi surfaces in gold and silver contact the zone boundary in a similar way. In gold the size of the contact area and the shape of the "neck" joining this to the main body of the surface have been obtained in a very direct way.

An oscillatory variation of ultrasonic attenuation in a magnetic field occurs because of geometrical coincidences between certain electron orbits and the periodic electric fields accompanying the sound wave. Liquid helium temperatures are needed to have the mean free path longer than the wavelength, λ. The effect, when $\vec{H}\perp\vec{q}$ (where $\vec{q}$ is the propagation vector of the wave), gives the dimensions and ultimately the shape of the Fermi surface because the period observed in $(H\lambda)^{-1}$ is determined by the Fermi momentum perpendicular to $\vec{H}$ and $\vec{q}$ at those parts of the surface where this momentum is extremal. If p is such a momentum, and P is the corresponding period in $(H\lambda)^{-1}$, then $p=e/(2cP)$.[6,7]

Magnetic field oscillations were observed in single crystals of zone-refined gold and silver at frequencies up to 154 Mc/sec at a temperature of 4.2°K. Longitudinal waves were propagated along both [001] and [011], and the direction of $\vec{H}$ varied while keeping $\vec{H}\perp\vec{q}$. Stronger oscillations were observed in gold than in silver, as many as 19 maxima and minima being found in certain orientations.

If the Fermi surface contacts a zone boundary, there will be a minimal cross section at the boundary; thus, if touching occurs in [111], there should be a long-period oscillation due to electrons circulating around this "neck" when $\vec{H}$ is along [111] and $\vec{q}$ is along [011]. Such a period has been observed in copper.[3] A pronounced period, which is shown in Fig. 1(a), is found also in gold and is the strongest evidence for the surface making contact. The momentum calculated from this period is 0.22, in units of 10^{-19} g cm sec^{-1}. (In the remainder of the paper all momenta will be expressed in these units.) This

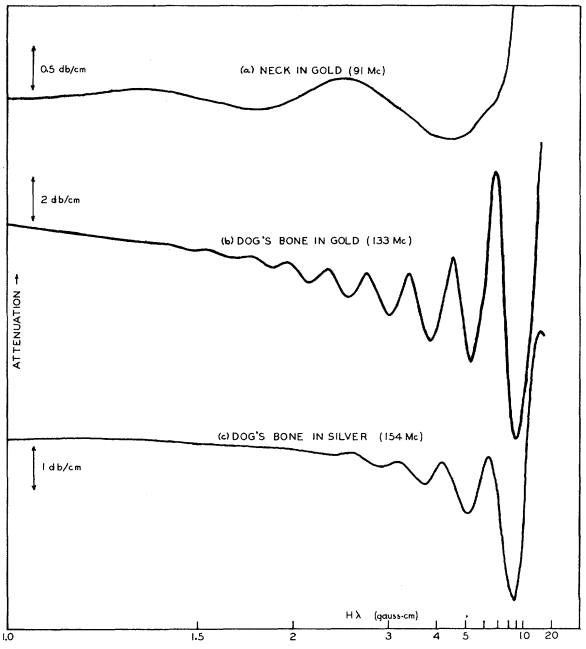

FIG. 1. Relative attenuation as a function of $H\lambda$ plotted on a reciprocal scale. (a) Attenuation in gold with $\vec{q}$ along [011] and $\vec{H}$ along [111]. (b) Attenuation in gold with $\vec{q}$ along [001] and $\vec{H}$ along [110]. (c) Attenuation in silver with $\vec{q}$ along [001] and $\vec{H}$ along [110]. (Note that attenuation scales are different for each curve.)

should be the radius of the touching area on the [111] zone boundary, which is 1.41 units away from the center of the zone. When $\vec{H}$ is moved away from [111], the neck period becomes shorter corresponding to the widening of the neck as it joins the main part of the surface. This angular dependence gives the detailed shape of the neck, the result being shown in Fig. 2.

The strongest period in gold appears with $\vec{q} \| [001]$ and $\vec{H} \| [110]$, as shown in Fig. 1(b); the corresponding momentum is 1.09 units. This period is interpreted as follows: Because contact occurs, the Fermi surface can be considered as a body-centered array of spheres (approximately) joined along [111], and corresponding directions, by the neck shown in Fig. 2; the central extremal cross section is a "dog's bone" shaped orbit when the field is along [110]. This is a hole-type motion involving four zone boundary reflections. The momentum of 1.09 units, therefore, is associated with half the length of this dog's bone. Since the distance from the center of the dog's bone to the center of the zone is 2.30 units, this period implies that the radius of the main surface along [110] is 1.21 units.[8]

For $\vec{q} \| [001]$ and $\vec{H}$ near [100], there is a much weaker period undoubtably due to electron orbits about the central body of the surface. From this period the radius of the main body along [100] is estimated as 1.26 units. The angular variation of this and the dog's bone period can be used to establish the shape of a principal cross section of the main body of the Fermi surface, the values of 1.26 units along [100] and 1.21 units along [110] being the maximum and minimum dimensions. If the surface in gold were a sphere that contained one electron per atom, its radius would be 1.27 units.

Even though the original silver was nominally purer than the gold, the oscillations for a given frequency were much weaker. The dog's bone orbit gave the most distinct period, this being shown in Fig. 1(c) for a frequency of 154 Mc/sec. A momentum of 1.05 units is found, which implies a main body radius of 1.25 units along [110] for silver. The zone and the ideal Fermi sphere are almost identical in silver and gold. There was an indication of a very weak neck period in silver, enough to give a rough estimate of 0.18 unit for the radius of contact. Even though the neck period is not clear, the strong dog's bone period, when $\vec{H} \| [110]$, leaves little doubt that the Fermi surface of silver makes zone boundary contact in a way similar to the other noble metals.

Our results agree with very recent de Haas–van Alphen measurements of Shoenberg.[9] He has observed the dog's bone oscillation in gold (the name is his) and the neck period in all three metals. The two methods are complementary in that the de Haas–van Alphen effect determines the area of an orbit, while the ultrasonic effect determines its length. Moreover, it seems that orbits involving zone boundary reflections give stronger oscillations than the main surface orbits with the ultrasonic effect, but that the opposite is true for the de Haas–van Alphen effect.

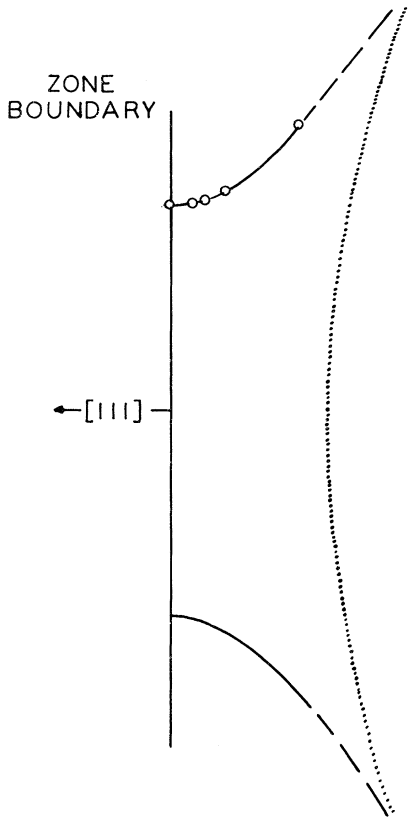

FIG. 2. Inferred shape of the touching "neck" in gold at the [111] zone boundary. The measured radius at the boundary is 0.22×10^{-19} g cm sec^{-1}. The dotted curve is a circle of radius 1.25×10^{-19} g cm sec^{-1} drawn from the center of the zone.

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EFFECTS OF POLYTYPISM ON THE ELECTRON PARAMAGNETIC RESONANCE OF $K_3Co(CN)_6$ AND OTHER SPECTROSCOPIC IMPLICATIONS*

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The electron paramagnetic resonance (EPR) spectra of Cr-doped $K_3Co(CN)_6$ crystals show unexpected splittings.[1] In every crystal investigated, at least four prominent lines, occurring in sets of two, were detected for each $\Delta M = \pm 1$ Cr^{+++} (in Co^{+++} sites) transition. In several crystals, at least two additional pair-wise equivalent resonances were noticed. The resonance geometry for the prominent lines is indicated in Fig. 1; the room temperature spectral data obtained at a frequency of 9.3 kMc/sec are given in Table I.

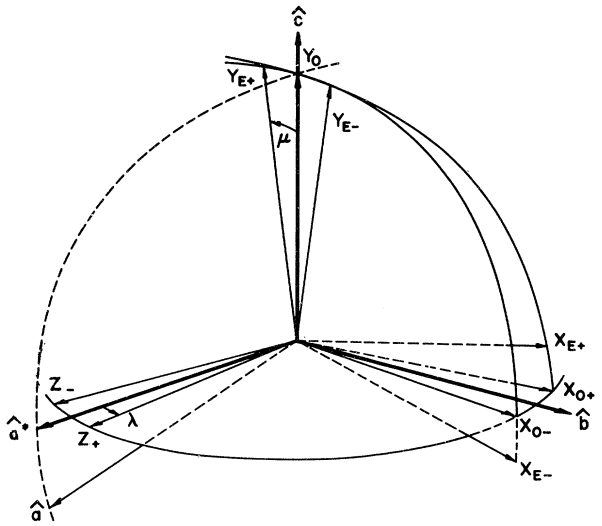

FIG. 1. Geometry of the four prominent Cr^{+++} resonances in polytypic K_3Cr-$Co(CN)_6$.

The existence of different well-defined stacking periodicities in crystals of a given chemical species is termed polytypism. Each layering (stacking) sequence corresponds to a specific polytype and generates a crystal (or region in a crystal) which is singularly described by a given unit cell and crystal structure. In general, several distinct polytypes may coexist in a visually homogeneous crystal.

Recently, pure and doped (0.1% Cr) crystals of $K_3Co(CN)_6$ were found to be polytypic by single-crystal x-ray diffraction.[2] The unit cells of the various polytypes are derived by rotation of (100) one-layer monoclinic lamellae about c and their stacking on (100). The geometry of the two most prevalent polytypes, $1M$ (one-layer monoclinic) and $2Or$ (two-layer orthorhombic) is illustrated

Table I. Room temperature spectral data at 9.3 kMc/sec.

Ordinary lines	Extraordinary lines
$\lambda = \pm 6.5°$	$\lambda = 6.5°$, $\mu = 8.25°$
$\mu = 0$	$\lambda = -6.5°$, $\mu = -8.25°$
$D = 0.07832$ cm^{-1}	$D = 0.07866$ cm^{-1}
$E = 0.00902_6$ cm^{-1}	$E = 0.00973_0$ cm^{-1}
$g_y = 1.991_2$	$g_y = 1.992_3$
$g_x = 1.993_6$	$g_x \simeq 1.993_3$