make it possible to pump paramagnetic salts at low temperatures at the optically active phonon frequency, producing acoustic phonons at half the pump energy with anomalously large amplitudes. These phonons could then be detected in absorption as vibronic sidebands on the low-energy side of the n_0-phonon line.

*Work supported in part by the National Science Foundation and the U. S. Office of Naval Research under Contract No. NONR-233(88).

†Alfred P. Sloan Foundation Fellow 1965-1967.

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GEOMETRICAL RESONANCE AND BOUNDARY EFFECTS IN TUNNELING FROM SUPERCONDUCTING In†

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(Received 18 November 1965)

A new type of structure in the tunneling characteristics of Al-AlO_x-Pb film diodes (Al and Pb both superconducting) has been reported recently for diodes employing very thick Pb films (2.9-9.7 μ).[1] The structure consisted of damped oscillations in d^2V/dI^2 vs V which were periodic in V and reflected weak periodic structure in dI/dV. Evidence was presented indicating that the structural period $h\nu$ depended only on Pb film thickness d(Pb). Specifically, $h\nu$ was found to be proportional to $1/d$ over the range investigated. When interpreted in terms of a standing wave phenomenon, the slope of $h\nu$ vs $1/d$ yielded a characteristic velocity comparable to the Fermi velocity.

Similar oscillations have now been observed with thick In films (5.8-32.2 μ), demonstrating that the effect is not restricted to strong-coupling superconductors and hence may occur rather generally. Furthermore, preliminary measurements indicate that the structural amplitude can be greatly enhanced (factors of five or more in d^2V/dI^2) by deposition of a rather thin "overlay" of silver (~0.2 μ) on the exposed In or Pb film surface, even though the silver may be tens of microns away from the AlO_x barrier at which tunneling occurs. Preliminary indications are that $h\nu$ is insensitive to the presence of the silver overlay for the relatively thin silver films employed so far. Finally, measurements on very thick films (33 μ for In and 26 μ for Pb) suggest the absence of an energy gap in the excitation spectrum in the long-wavelength limit.

Diodes were prepared by standard methods[2] and exhibited room-temperature resistances in the range 2-100 Ω. Thin Al films (~300 Å) allowed the influence of longitudinal magnetic fields to be studied with both Al and In superconducting. Diode pairs (sisters) fabricated simultaneously, and differing only by one having a silver overlay, were employed in structural enhancement studies. All films were deposited on glass substrates nominally at room temperature. Preliminary x-ray studies have disclosed that the resulting polycrystalline In films exhibit a strong (101) texture, i.e., (101) crystallographic planes tend to be aligned parallel to the glass substrate. [Lead films behave similarly but with (111) planes involved.] Weighing techniques were utilized to determine In film thicknesses. Conventional modulation methods were employed to measure dV/dI and d^2V/dI^2.

Plots of dV/dI and d^2V/dI^2 for an In film 8.5 μ thick are presented in Fig. 1. These data are for a diode unit with a silver overlay. The structural amplitude (d^2V/dI^2) is approximately five times greater than for a sister unit without silver. To within experimental accuracy, $h\nu$ was the same for both units. The corresponding structure in dV/dI is much stronger than previously reported for Pb (without silver),[1] but is comparable to that observed with Al-AlO_x-Pb diodes having a silver overlay.

Matters now proceed much as for the case of Pb. Structural features in d^2V/dI^2 have been indexed by integers η with $\eta=1$ corresponding to the first and strongest peak of the series.

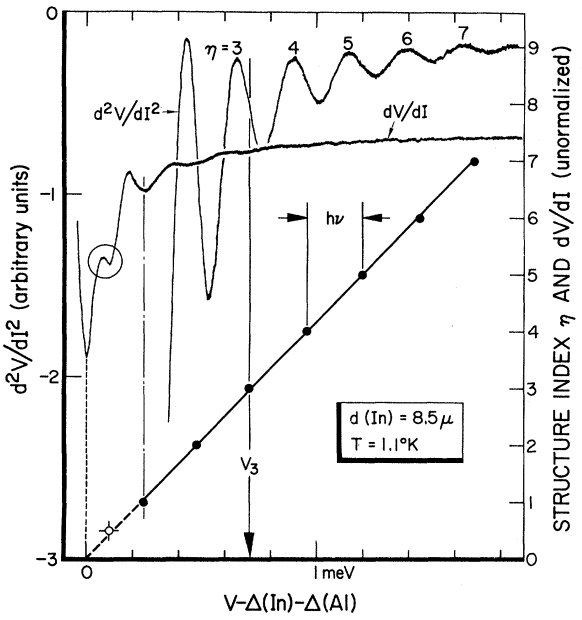

FIG. 1. Voltage dependence of dV/dI and d^2V/dI^2 for an Al-AlO_x-In tunnel diode. Modulation levels were 14 μV (rms) and 70 μV (rms), respectively. The In film had one surface covered with a relatively thin (~0.2 μ) film of silver. The structural amplitude in d^2V/dI^2 was approximately 5 times greater than for a very similar diode without silver. Integers η index peaks in d^2V/dI^2 with $\eta = 1$ corresponding to the first peak of the series (not shown). Voltages V_η denote points of maximum negative slope in d^2V/dI^2 and correspond to local maxima in dI/dV. For the relatively thin films of silver employed, $h\nu$ did not appear sensitive to the presence of silver. The relationship between the encircled feature and the main structural sequence is not clear at this point.

(Only oscillations corresponding to $2 \leq \eta \leq 7$ have been included for clarity. Structure corresponding to $\eta = 9$ was discernible in the original d^2V/dI^2 data.) Under conditions of the experiment, points of maximum negative slope correspond to local maxima in dI/dV and are labeled V_η. A plot of $V_\eta(\eta)$ yields a straight line of slope $h\nu$ and intercept $V_0 = \Delta(\text{In}) + \Delta(\text{Al})$. This is taken as evidence for the presence of assisted tunneling processes corresponding to quantum emission.[1,3] A new feature of the present data is the sharp dip in dV/dI (encircled in Fig. 1) occurring between V_0 and V_1. Preliminary observations suggest that the strength and form of such features are sensitive to the presence of silver, while the voltage at which they occur depends primarily on

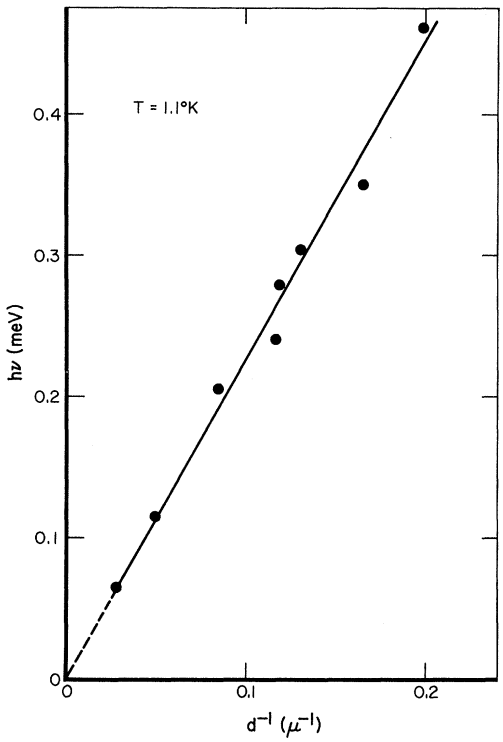

FIG. 2. Variation of oscillation period $h\nu$ with inverse In film thickness d(In).

d(In).[4] The relationship between such features and the main structural sequence is not clear at this point.[5] Figure 2 depicts the proportionality between $h\nu$ and $1/d$ for the range covered and indicates that any energy gap in the excitation spectrum (as $1/d$ tends to zero) must be less than 0.07 meV. (For Pb, the gap must be less than 0.05 meV.)

There appears to be a strong analogy between the general behavior of Al-AlO_x-In and Al-AlO_x-Pb diodes. Both In and Pb have $h\nu$ insensitive to voltage polarity, specific junction impedance, and the presence of longitudinal magnetic fields less than the bulk critical value. Both behave similarly with respect to thermal smearing and annealing effects. Both show structural enhancement with silver. (Structure of the type under consideration is observed only when both diode members (Al/In or Al/Pb) are superconducting.)

In view of the foregoing evidence, it appears reasonable to ascribe a common origin to the structure associated with In and Pb. It has been tentatively suggested that the effect in

Pb may be attributable to collective electronic excitations involving Cooper pairs.[1] A key point underlying this view was that if one assumed a standing wave model, the observed slope of $h\nu$ vs $1/d$ yielded a velocity v comparable to the Fermi velocity u_F. Table I indicates a similar result for In. Values v_1 and v_2 for In were calculated from the data of Fig. 2 on the basis that the wavelength equaled either d or $2d$. Three values of u_F have been included for comparison: u_{F1} corresponds to the free-electron value with $m^*=m$, u_{F2} to the free-electron value with calorimetric values of m^* ($1.35m$ for In and $2.1m$ for Pb), and u_{F3} to experimental values obtained from anomalous skin-effect data. (Valences of three and four were assumed for In and Pb, respectively.) It is interesting to note that In and Pb have ratios v_2/u_{F2} approximately equal to $\sqrt{3}/2$ (0.85 and 0.86, respectively).

It is difficult to assess the importance of crystalline texture at this juncture since all tunneling experiments have involved strongly textured In and Pb films. Repetition of such measurements with single-crystal laminae (perhaps epitaxial films or recrystallized foils) as a function of orientation would be of considerable interest, as would measurements with randomly oriented polycrystalline laminae. Both approaches pose challenges with regard to experimental technique.

Generally speaking, the present results continue to support the view that thick films of certain superconductors exhibit geometrical resonances (standing-wave modes) to which tunneling electrons may lose energy in quantum-assisted transitions. Since the effect has now been observed in both strong- and weak-coupling superconductors, there may be increased reason to anticipate its general occurrence.

Perhaps the most interesting and potentially useful aspect of the present investigation stems from the apparent sensitivity of the structural amplitude to the presence of an overlay. At best, this could mean that two new controllable variables—overlay species and overlay thickness—have become available. With regard to the former, the normal metal (Ag) could be replaced with a superconductor, ferromagnet, semiconductor, insulator, or even a composite laminar structure. If the overlay does indeed produce its effect by changing the constraints imposed on an excitation wave, then investigations of this type might shed light on the generalized displacement associated with the excitation. The actual merit of such an approach remains rather problematical for the moment. Returning to silver, whether the thin overlay produces structural enhancement by providing unpaired electrons at the thick superconductor's boundary or whether other factors are involved cannot be decided on the basis of the limited information currently available. It is clear, however, that a number of possible experimental continuations are available.

Table I. Comparison of velocities v_1 and v_2 (calculated from $h\nu$ vs $1/d$ plots) with various values of the Fermi velocity.[a]

Film	v_1[b]	v_2[c]	u_{F1}[d]	u_{F2}[e]	u_{F3}[f]
In	5.5	10.9	17.3	12.8	12[g]
Pb	3.7	7.5	18.2	8.7	5.0[h]

[a]All velocities are in units of 10^7 cm/sec.
[b]Wavelength taken equal to d.
[c]Wavelength taken equal to $2d$.
[d]Free-electron prediction assuming m^*/m_e to be unity.
[e]Free-electron prediction using m^*/m_e ratios from specific-heat data (1.35 for In and 2.1 for Pb).
[f]Values obtained from anomalous skin-effect data.
[g]P. N. Dheer, Proc. Roy. Soc. (London) A260, 333 (1961).
[h]J. Bardeen and J. R. Schrieffer, in Progress in Low Temperature Physics, edited by C. J. Gorter (Interscience Publishers, Inc., New York, 1961), Vol. III, Chap, VI, p. 243.

The author is indebted to T. Wolfram for valuable discussions, and to R. R. Hargrove for preparing the diodes studied.

Note added in proof.—It has recently been brought to the author's attention that W. L. McMillan and P. W. Anderson have formulated an alternative explanation for the origin of the observed structure including the enhancement effect. Their explanation involves the birefringent propagation of single-particle excitations, and appears capable of accounting for the additional structural feature encircled in Fig. 1.

†Based on work sponsored by The Division of Research, Metallurgy and Materials Programs, U. S. Atomic Energy Commission, under Contract No. AT-(11-1)-GEN-8.

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[2]I. Giaever and K. Megerle, Phys. Rev. 122, 110 (1961).

[3]Transitions corresponding to quantum absorption $[0 \leq |V| \leq |\Delta(\text{In}) + \Delta(\text{Al})|]$ have not as yet been observed. As in the case of Pb, this is thought to reflect the low equilibrium excitation of the energy reservoir (at 1.1°K) to which tunneling electrons couple.

[4]Similar effects are observed in Pb although there the situation is further complicated by the apparent multiple-gap behavior associated with thick Pb films.

[5]It is conceivable that such features may be related to the type of infrared-absorption structure observed for single crystals of Sn. P. L. Richards, Phys. Rev. Letters 7, 412 (1961).

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de HAAS–van ALPHEN EFFECT, MAGNETIC BREAKDOWN, AND THE FERMI SURFACE OF CADMIUM*

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(Received 6 December 1965)

The accepted model for the Fermi surface of cadmium is the modified single-orthogonalized-plane-wave (OPW) surface first proposed by Harrison.[1] This surface must be described in the single-zone scheme since the degeneracy of the energy bands across the AHL symmetry plane of the hexagonal-close-packed Brillouin zone is removed when spin-orbit interaction effects are considered.[2] The results of a number of previous experiments[3] pertinent to the electronic band structure of cadmium have been interpreted in terms of this model. Our de Haas–van Alphen (dHvA) and magnetoresistance investigations in cadmium indicate that this description of its Fermi surface in incorrect in certain essential details. The purpose of this Letter is to describe the model which fits our data (as well as data previously published) and, in addition, to discuss some interesting magnetic breakdown[4] effects which we have observed.

The dHvA measurements were taken in magnetic field strengths extending to 38 kG utilizing low-frequency field-modulation techniques.[5] The crystals used in these investigations were purified by zone refining and had residual resistance ratios ($R_{300°K}/R_{1.1°K}$) of 39000. The extremal cross-sectional area branches which were derived from the observed dHvA frequencies[6] and which are pertinent for this discussion are shown for the major crystallographic planes on a semilog plot in Fig. 1. The angular variation of each of these branches was determined from field-rotation diagrams.[7] The data are accurate to 0.1%.

The extremal-area branches α and β_1 in Fig. 1 have been observed in previous experiments. However, they were thought to be degenerate along (0001) and hence consistent with a six-fold set of cylinders whose major axes were tilted 28.5° from (0001) toward (11$\bar{2}$0). These were assigned to diagonal arm modifications of the hole sheet in the second zone of the sinfle OPW model.[1] Our measurements show that these two branches are not degenerate at (0001) and that in fact they arise on sheets of the Fermi surface which have an axis of symmetry along (0001). Branch α arises on a closed sheet of the Fermi surface. We have assigned it to the hole sheet in the first zone of the single-OPW

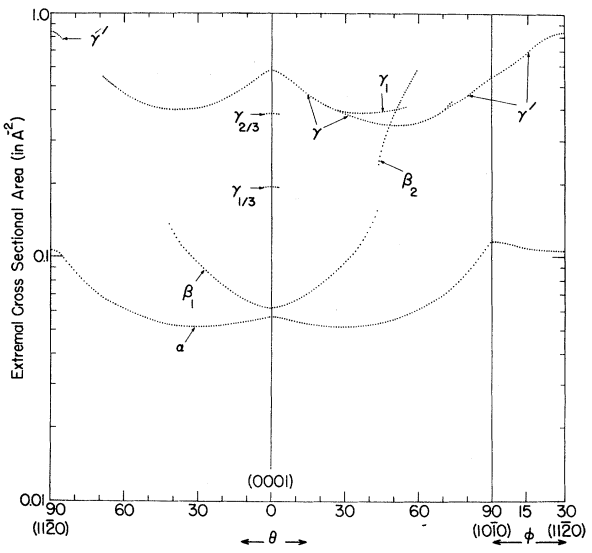

FIG. 1. The extremal dHvA cross-sectional area branches associated with the sheets of the Fermi surface in the first and second Brillouin zones of cadmium. The polar angle, θ, measures the angle between (0001) and the magnetic field direction. The azimuthal angle, φ, measures the angle between (10$\bar{1}$0) and the direction of the magnetic field when the latter is in the basal plane. The unit of area is reciprocal square angstroms.