

COMMENTS AND ADDENDA

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Evidence for Splitting of Phonon Structures in Single-Crystal Pb Tunneling*

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Experimental evidence is presented which shows that the two energy gaps appearing in the tunneling characteristics of superconducting Pb crystals have a discernible splitting effect on some of the transverse phonon peaks. This is in agreement with our earlier observations but disagrees with the recent results of Leger and Klein.

INTRODUCTION

In a previous paper¹ dealing with a tunneling investigation of energy-gap anisotropy in bulk single-crystal Pb it was mentioned that some of the second-derivative curves (d^2V/dI^2 vs V) showed evidence of splitting of some of the phonon structures. In particular, two of the transverse phonon structures (labeled as ω_3 and ω_4 in Fig. 3 of Ref. 1) showed knees on the low-voltage sides of the peaks. Since the peak-to-knee separation was the same as $\Delta_2 - \Delta_1$, it was suggested that the effect was due to the presence of the two gaps Δ_1 and Δ_2 in the crystal.

In those experiments the crystals were probed with Pb films having an energy gap Δ_F nearly coincident with Δ_2 . Under these conditions it could be possible to get an enhancement² of the phonon structure associated with Δ_2 . This was offered as a tentative reason why the splitting was not more pronounced.

Recently Leger and Klein³ have reported a somewhat similar tunneling experiment in which thick Pb films ($\sim 5 \mu$) are probed by Al films. It was assumed, following Tomasch,⁴ that the Pb films were predominantly single crystalline with the [111] direction oriented normal to the film surface. They observed two energy gaps, $\Delta_1 = 1.25$ meV and $\Delta_2 = 1.40$ meV, in the tunneling characteristics, but did not find any evidence for splitting of the phonon structures. In their experiments the energy gap of the probing film, Δ_{Al} , was far re-

moved from either Δ_1 or Δ_2 , so that there was no question of gap coincidence. Also, the Al film produces no phonon structure of its own.

Using the Eliashberg gap equation and a number of assumptions, Leger and Klein further showed that such a splitting of the longitudinal phonon structures should not be expected. However, they did not consider explicitly the transverse phonon structures for which the evidence of splitting was claimed.

Closer examination of our d^2V/dI^2 curves (including recent ones) gives strong evidence that the knees appearing on some of the transverse structures actually are associated with the two energy gaps of the crystal. Furthermore, there is often an enhancement of the phonon structure associated with Δ_2 , but this does not appear to be caused by gap coincidence, as was previously suggested. The various pieces of evidence are enumerated below.

EXPERIMENTAL

- (i) The peak-to-knee separation agrees with that expected from the energy-gap separation to within the measurement accuracy of about 10%.
- (ii) The splitting is not present in all of the single-crystal specimens. In the cases for which the splitting does occur the tunneling characteristics have two distinctive features. First, the energy gaps Δ_1 and Δ_2 are well resolved in the tunneling characteristics and, second, the tunneling current contribution at Δ_2 is greater than the contribution

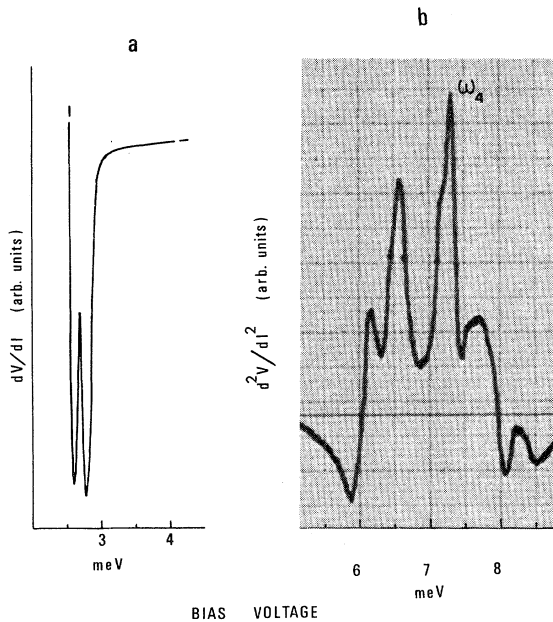


FIG. 1. (a) First-derivative curve for which the two energy gaps are well resolved. (b) Second-derivative curve (photographically reproduced from the X-Y recorder tracing) from the specimen of (a), showing the transverse phonon structures. Note the well-developed knee on the ω_4 peak. The direction normal to the tunnel barrier is $\theta = 53^\circ$, $\phi = 38^\circ$, where θ and ϕ are the usual spherical polar angles (Ref. 1). $T = 1.2^\circ\text{K}$.

at Δ_1 . That is, the ratio I_2/I_1 , defined in Fig. 1 of Ref. 1, is considerably greater than unity. Compare Figs. 1 and 2 below.

Figure 1(a) shows a dV/dI curve for a specimen in which the energy gaps are well resolved. The d^2V/dI^2 curve for the same specimen is shown in Fig. 1(b), where it can be seen that the splitting of the ω_4 structure is pronounced. On the other hand, Fig. 2(a) shows a dV/dI curve in which the energy gaps are not well resolved, and in the corresponding d^2V/dI^2 curve of Fig. 2(b) there is no evidence of splitting. The ratio I_2/I_1 was 1.5 ± 0.1 for the specimen of Fig. 1, while for the specimen of Fig. 2 it was 0.9 ± 0.1 .

The degree of resolution of the gaps in dV/dI and the magnitude of I_2/I_1 both depend on the orientation of the tunnel junctions.^{1,5} Junctions oriented near the [111] direction have particularly well-resolved gaps and high values of I_2/I_1 . The resolution of the peaks in the dV/dI curves is determined by the intrinsic widths of the peaks and also by their separation. Narrow peaks usually give good resolution but the peak separation, by itself, is not enough to say whether a splitting of the phonon structures will occur. For example, the maximum observed value of $\Delta_2 - \Delta_1$ was 0.22 ± 0.01 meV, yet there was no evidence of splitting in the d^2V/dI^2

curve of this specimen. In this case, the energy gaps were not well resolved in the dV/dI curve, and $I_2/I_1 = 0.8 \pm 0.1$.

Out of a total of ten specimens, five showed evidence of knees at ω_4 and, in general, there was good correlation with well-resolved gaps. Furthermore, the average value of I_2/I_1 was 1.51 ± 0.11 rms for the specimens showing a splitting, while for those with no splitting the average was 1.08 ± 0.16 rms.

(iii) A curve similar to the ω_4 structure of Fig. 1(b) can be reconstructed by adding together two peaks of different amplitude and displaced in energy by the appropriate $\Delta_2 - \Delta_1$. This is shown in Fig. 3(a), where the width of the major component peak was estimated to be the same as that of the major ω_4 peak in Fig. 1(b). The width of the minor peak is scaled down by the ratio of the gap widths in the dV/dI curve of Fig. 1(a). The amplitudes of the component peaks are in the ratio 2:2:1.

Figure 3(b) shows a similar reconstruction for the specimen of Fig. 2(b), where there was no splitting of the phonon structures. The component peaks are somewhat wider in this case and their separation $\Delta_2 - \Delta_1$ is less. The amplitude ratio of about 1:1 gives the best fit to the shape of the ω_4 peak in Fig. 2(b).

The relative amplitude of the component peaks of Fig. 3 is a matter of some interest. This ratio definitely depends on the tunneling current ratio I_2/I_1 . When I_2/I_1 was large, the amplitude ratio

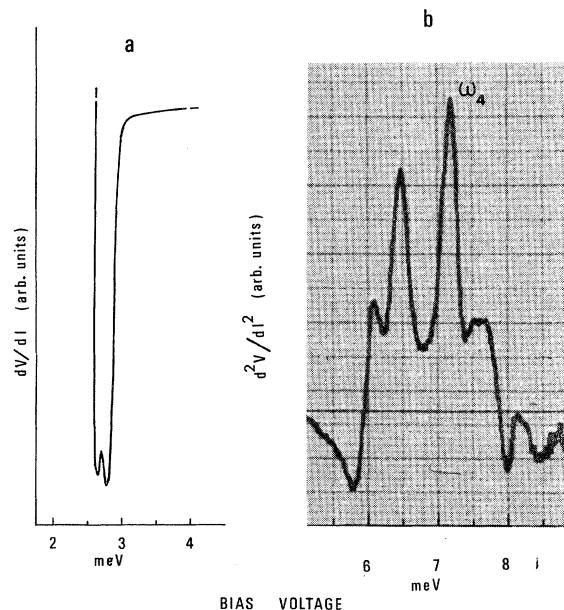


FIG. 2. (a) First-derivative curve for which the two energy gaps are not well resolved. (b) Second-derivative curve from the specimen of (a). Note the absence of a knee on the ω_4 peak. $\theta = 12^\circ$, $\phi = 42^\circ$, $T = 1.3^\circ\text{K}$.

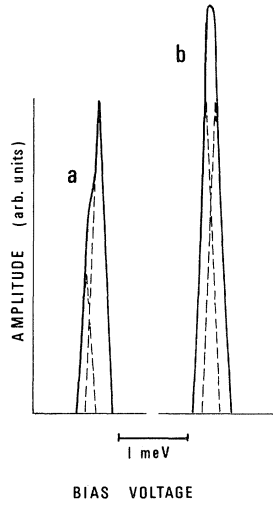


FIG. 3. Reconstructed phonon structure peaks.

necessary to reconstruct an acceptable curve was also large. In fact, in the majority of cases, the best-fitting curves were obtained when the amplitude ratio was made equal to $(I_2/I_1)^2$. However, it should be emphasized that this relation is tentative, as the data in this respect are not as good as one would like. It is hoped that further experiments could resolve the matter.

(iv) The experimental data support the fact that, on the average, the peaks in d^2V/dI^2 have Δ_2 , rather than Δ_1 , as their reference gap in the crystal.⁶ This becomes apparent when either $(\Delta_1 + \Delta_F)$ or $(\Delta_2 + \Delta_F)$ is subtracted from the raw data and the resulting numbers compared. Table I shows the averages from nine single-crystal specimens along with the corresponding data for a thin Pb film specimen of this work. First, when $(\Delta_2 + \Delta_F)$ is used as the reference point, the single-crystal data are in much closer agreement with the thin-film data. Actually, the agreement with the thin-Pb-film data of Rowell and Kopf² is even closer. Second, and perhaps more important, the single-crystal data show less scatter from the mean when referred to $(\Delta_2 + \Delta_F)$. That is to say, the positions

of the phonon structures in the d^2V/dI^2 curves are much more responsive to changes of Δ_2 than to changes of Δ_1 .

It is perhaps worth noting that two recent single-crystal junctions show considerably increased energy values of the phonon structures, as much as 0.39 meV for the longitudinal peaks. This is in sharp contrast with the majority of the specimens, which showed rather insignificant variation with tunneling direction.⁵ These particular two junctions had different orientations than those sampled previously. It is possible that the effect is similar to that predicted by Carbotte and Dynes,⁷ but more data are required before a definite conclusion can be reached. The data from these two specimens are not included in Table I.

DISCUSSION

Taken together, the various data represent good evidence that the two gaps do produce a discernible splitting effect on at least one of the phonon peaks. It is true that we see no evidence for splitting of the longitudinal peaks, but in these cases the observed peak widths are almost twice as large as the transverse ones. Thus, in the longitudinal case there is no disagreement with the prediction of Leger and Klein,³ but in the general case it appears to be wrong to say that no splitting effects should be observed.

The absence of splitting in the experimental curves of Leger and Klein is not surprising, considering the rather poor resolution in comparison with the single-crystal results. For example, the transverse peaks in their Fig. 1(b) are considerably wider than those of the single crystals. The reason for the rather poor resolution is not clear, although it could be connected with the use of an Al probing film.² The fact that the two energy gaps in their dV/dI curve are well resolved does, however, suggest that the energy gap in the Al film is adequately sharp.

It is also worth noting that the energy-gap values reported by Leger and Klein³ ($2\Delta_1 = 2.50$ meV and $2\Delta_2 = 2.80$ meV) do not agree with the bulk single-crystal results¹ for the [111] direction ($2\Delta_1 = 2.36$

TABLE I. The first two rows give the average phonon structure energies in meV with $(\Delta_2 + \Delta_F)$ and $(\Delta_1 + \Delta_F)$, respectively, subtracted from the values measured in the d^2V/dI^2 curves. The rms deviations from the averages are also given. The third row gives the phonon structure energies from a Pb film (~ 3000 Å) specimen with $2\Delta_F = 2.78$ meV subtracted (this work).

Reference energy	ω_1	ω_2	ω_3	ω_4	ω_5	ω_6	ω_7
$\Delta_2 + \Delta_F$	3.10 ± 0.03	3.40 ± 0.01	3.79 ± 0.01	4.51 ± 0.02	5.28 ± 0.02	8.52 ± 0.02	9.05 ± 0.02
$\Delta_1 + \Delta_F$	3.26 ± 0.04	3.57 ± 0.03	3.96 ± 0.03	4.68 ± 0.03	5.45 ± 0.03	8.69 ± 0.02	9.22 ± 0.04
$2\Delta_F$	3.01		3.70	4.37	5.22	8.34	9.02

meV and $2\Delta_2 = 2.76$ meV), nor with the recent results of Lykken *et al.*⁸ on single-crystal Pb films. This perhaps means that the films of Leger and Klein³ were not single crystalline.

The two gaps Δ_1 and Δ_2 observed in tunneling on single-crystal Pb^{1,5} do not vary greatly with orientation ($\sim 5\%$) and Leger and Klein³ suggested that this is experimental evidence for a rather nonselective tunneling process. However, the fact that (i)

the current ratio I_2/I_1 ,^{1,5} (ii) the resolution of the gaps in dV/dI , and (iii) the detailed shapes and amplitudes of the transverse phonon peaks in d^2V/dI^2 all vary significantly with orientation⁵ would seem to suggest that the tunneling is selective.

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³A. Leger and J. Klein, Phys. Rev. B **3**, 3968 (1971).

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⁵B. L. Blackford, Physica (to be published).

⁶This is presumably due to the fact that, for the majority of specimens, the tunneling current contribution at Δ_2 is greater than at Δ_1 .

⁷J. Carbotte and R. C. Dynes, Physica (to be published). Carbotte and Dynes predict that the energies of observed phonon structure peaks should vary with tunneling direction, giving the possibility of measuring phonon anisotropy by single-crystal tunneling.

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High-Temperature Magnetic Susceptibility of Dilute Alloys in the Mean-Random-Molecular-Field Approximation

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An examination of the high-temperature magnetic susceptibility in the mean-random-field approximation gives new information on the interaction mechanism between the impurities in a dilute-magnetic-alloy system. More specifically, the experimental "high"-temperature susceptibilities of Au-Fe, Au-Cr, and Au-Mn are analyzed and found to be in good semiquantitative agreement with theory. A theoretical connection between the high- and low-temperature properties of the alloy system agrees well with experiments on Au-Fe and Au-Cr.

Dilute magnetic impurities in nonmagnetic metals exhibit a maximum^{1,2} in their temperature-dependent susceptibility $\chi(T)$, and the temperature of the maximum T_{\max} is approximately proportional to the impurity concentration c for impurity concentration of the order of 1%. Whereas a reasonable amount of theoretical and experimental work has been done to understand the behavior of these alloys at temperatures³ much below T_{\max} , very little attention has been paid to their properties at temperatures greater than T_{\max} .

The purpose of this note is to point out that new and useful information on the interaction mechanism between the magnetic impurities can be obtained from intermediate- and high-temperature magnetic susceptibility of the dilute-magnetic-alloy system using a mean-random-field (MRF) approximation recently developed by one of the authors.⁴

A brief summary of the theory is as follows: The magnetic impurities are assumed to be randomly and uniformly distributed throughout the solid, and the statistical model of Margenau⁵ is used to derive the probability distribution $P(H, T)$ of the random internal field H at temperature T . In order to obtain an analytical expression for $P(H, T)$, an approximation is used in which, when calculating the field distribution about a particular impurity site, functions of the random fields at all other impurity sites are replaced by their mean values. This approximation is called the mean-random-field approximation. One thus obtains a self-consistent integral equation for the probability distribution $P(H, T)$. The result is⁴

$$P(H, T) = (1/\pi) \{ \Delta(T) / [\Delta(T)^2 + H^2] \}, \quad (1)$$

where Δ is the width of the probability distribution

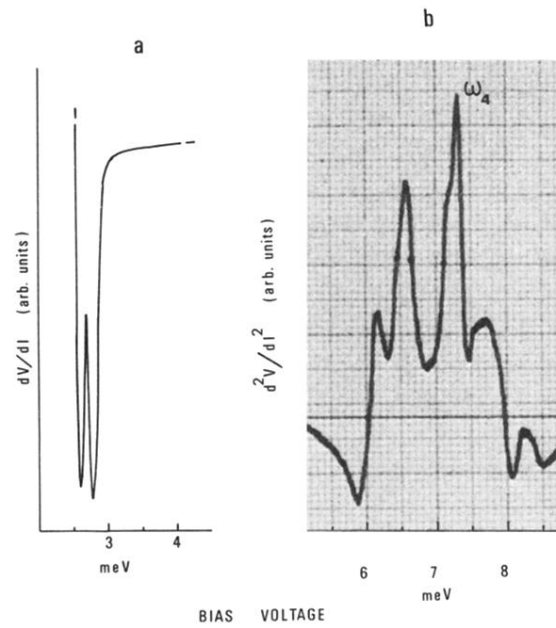


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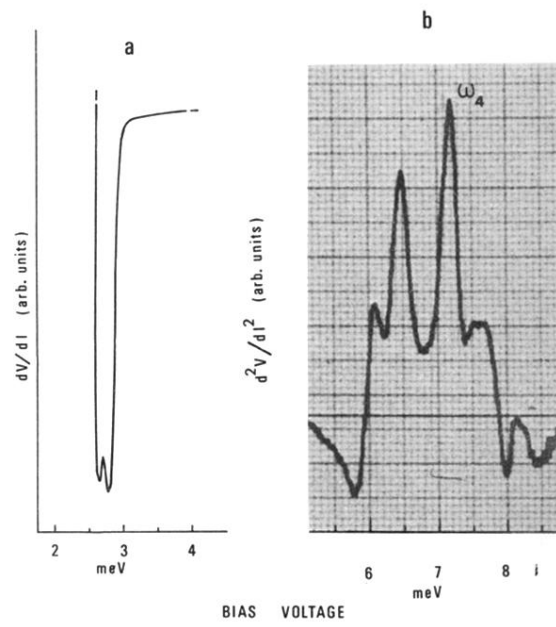


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