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Measurement of Microwave-Enhanced Energy Gap in Superconducting Aluminum by Tunneling*

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Al-Al_2O_3-Al tunnel junctions were used to measure large increases in the energy gap of superconducting aluminum films in the presence of 10-GHz microwave radiation. When the microwave power was increased at constant temperature, enchancement occurred only for temperatures at which twice the equilibrium energy gap exceeded the photon energy. When the temperature was increased at constant microwave power, enhancement was observed at higher temperatures.

Eliashberg and co-workers[1] have predicted that the energy gap, Δ, of a superconducting thin film may be enhanced by microwave irradiation. In their model, photons of frequency $\nu < 2\Delta/h$ excite quasiparticles from states near the bottom of the excitation spectrum to states of higher energy. Thus, additional pair states near the Fermi wave vector, k_F, become available for occupancy. Since the pair states near k_F contribute most strongly to the pairing interaction, this redistribution of pair-state occupancy increases the condensation energy, and leads to an increase in Δ. Eliashberg and co-workers[1] have suggested that this increase would account for the microwave enhancement of the critical current of superconducting microbridges.[2] Subsequently, several experiments have supported the concept of gap enhancement: phonon-induced enhancement of the critical current of superconducting microbridges and point contacts,[3] microwave-induced enhancement of the critical currents and transition temperatures of aluminum strips,[4] and microwave-induced enhancement of the voltage at which gap structure occurs in superconducting point contacts.[5] Recently, Chang and Scalapino[6] have performed detailed computer calculations of gap enhancement in which the energy dependence of the quasiparticle recombination time, and the effects of the nonequilibrium phonon distribution are included.

We have used Al-Al_2O_3-Al tunnel junctions to measure large increases in the energy gap of superconducting aluminum films in the presence of X-band radiation.[7] We define $\langle\Delta_e\rangle$ as the average equilibrium gap for the two films of a given junction, and T_ν as the temperature at which $h\nu = 2\langle\Delta_e\rangle$. When the microwave power was increased from zero with the junction at fixed temperature, T, $\langle\Delta\rangle$ increased for $T<T_\nu$, and decreased for $T \gtrsim T_\nu$. Alternatively, when we maintained constant microwave power and increased the temperature from below T_ν, gap enhancement was observed to a temperature $T_\downarrow$, where $T_\downarrow > T_\nu$. $T_\downarrow$ increased with microwave power. When the temperature was subsequently lowered at constant microwave power, the gap abruptly reappeared at a temperature significantly lower than $T_\downarrow$.

The Al-Al_2O_3-Al junctions were fabricated in a cross-film geometry with a film width of about 300 μm, and film thicknesses in the range 80 to 300 nm. Normal-state resistances ranged from 1 to 10 Ω. The transition temperatures of the films were from 1.20 to 1.26 K. The substrate was usually BaF_2, which has good acoustical matching to aluminum, and a low absorption at microwave frequencies.[8] Each substrate was mounted in an X-band wave guide near an adjustable short-circuit plunger, with the plane of the films parallel to the axis of the wave guide. The junction and wave guide were immersed in a temperature-regulated helium bath. The temperature of the helium was measured to a relative accuracy of 0.3 mK with an Allen-Bradley carbon resistor that was carefully shielded to eliminate any discernible interaction with the microwaves. Two concentric Mu-metal cans around the cryostat reduced the ambient field to below 10^{-6} T. The differential resistance, dV/dI, of the junctions was measured using a standard technique.[9]

The upper curve in Fig. 1 shows dV/dI vs voltage, V, for a representative junction on a BaF_2 substrate in the absence of microwave power. The thickness of each Al film was about 300 nm. There are two sharp minima in dV/dI, a and b, that we assume occur at voltages[10] $(\Delta_> + \Delta_<)/e$ and $(\Delta_> - \Delta_<)/e$, where $\Delta_>$ and $\Delta_<$ are the larger and smaller energy gaps of the two films. Each gap was in good agreement with the BCS prediction[11] over the temperature range studied. $T_{c>}$ and $T_{c<}$ were the temperatures at which $\Delta_>$ and $\Delta_<$ extrapolated to zero. We use the higher transition temperature, $T_{c>}$, in the definition of the reduced temperature, t. The remaining curves in Fig. 1 show the additional structure that was induced as the 10-GHz (41.4-μV) microwave power was increased. The power levels shown in the figure refer to the power delivered to the wave guide. At a reduced temperature of 0.99, $h\nu/2\Delta_<$ was equal to 0.83 for this junction. Part of the structure arose from photon-assisted tunneling.[10] The minima c, d, e, and f occurred at voltages $(\Delta_> + \Delta_< + \Delta_< + h\nu)/e$, $(\Delta_> + \Delta_< - h\nu)/e$, $(\Delta_> - \Delta_< + h\nu)/e$, and $[-(\Delta_> - \Delta_<) + h\nu]/e$, respectively. As the microwave power was increased from zero, the minimum at a $(\Delta_> + \Delta_<)$ moved to higher voltages. At each power level, the photon-assisted tunneling minima c and d $(\Delta_> + \Delta_< \pm h\nu)$ moved by the same voltage as the minimum at a, so that c and d were separated from a by a constant voltage $\pm h\nu/e$. Thus, the average gap, $\langle\Delta\rangle = \frac{1}{2}(\Delta_> + \Delta_<)$, was enhanced by the microwaves. The voltage at which b $(\Delta_> - \Delta_<)$ occurred was relatively independent of microwave power, as were the photon-assisted tunneling minima e and f $[\pm(\Delta_> - \Delta_<) + h\nu]$. Additional structure indicated by arrows at the bottom of the figure was due to microwave-induced Josephson current steps[12] at voltages $nh\nu/2e = 20.7n$ μV ($n = 0, \pm 1, \pm 2, \ldots$). The voltages at which these steps occurred were independent of power. To within the experimental accuracy (± 1 μV), each type of structure occurred at the same voltage for positive and negative polarities. The amplitude of both the photon-assisted tunneling minima and the Josephson minima oscillated as the power was increased to 20 mW.

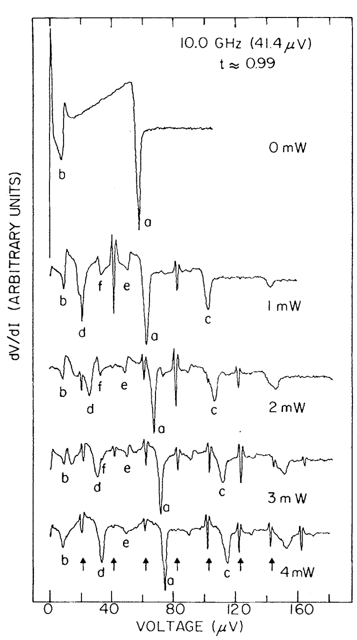

FIG. 1. dV/dI vs voltage for Al-Al_2O_3-Al tunnel junction on BaF_2 substrate.

In Fig. 2 we plot values of $2\Delta_>$, $2\Delta_<$, and $2\langle\Delta\rangle$ vs microwave power at $t \approx 0.99$ for the junction referred to in Fig. 1. As the power was increased from 0 to 20 mW, $\Delta_>$, $\Delta_<$, and $2\langle\Delta\rangle$ increased by about 80%, while $(\Delta_> - \Delta_<)/e$ varied by no more than about 3 μV. The variation of $\langle\Delta\rangle$ with microwave power qualitatively resembled the behavior predicted by Chang and Scalapino.[6] At the higher power levels, the enhancement began to saturate, but, at this temperature, did not begin to decrease again at the highest power available.

Figure 3 shows the temperature dependence of $2\langle\Delta\rangle$ for several power levels for the junction on BaF_2 referred to in Figs. 1 and 2. These curves were obtained using two different methods.

Method 1.—The temperature was incrementally varied from the lowest temperature shown, keeping the microwave power constant. In the case of the 1-mW curve, at the highest temperature indicated, the structure in dV/dI began to smear

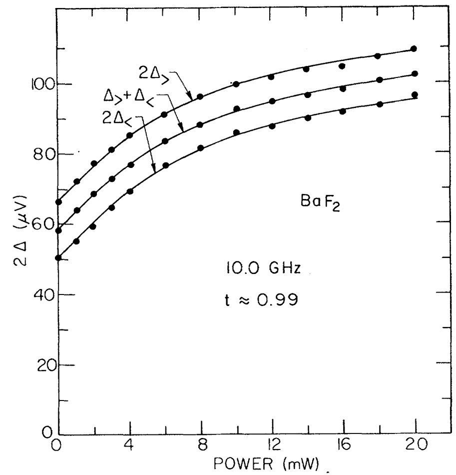

FIG. 2. $2\Delta_>$, $\Delta_>+\Delta_<$, and $2\Delta_<$ vs microwave power for Al-Al_2O_3-Al junction.

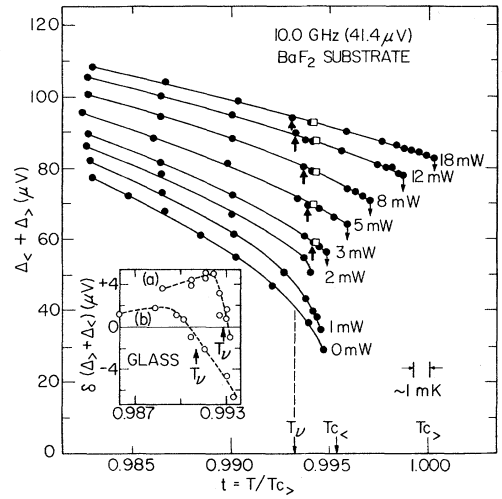

FIG. 3. $\Delta_>+\Delta_<$ vs reduced temperature at several levels of microwave power for Al-Al_2O_3-Al junction on BaF_2 substrate. Inset shows gap enhancement, $\delta(\Delta_> + \Delta_<)$, vs temperature for Al-$Al_2O_3$-Al junction on a glass substrate at (a) 9.9 GHz and (b) 11.9 GHz. The appropriate values of T_ν are indicated.

out; at higher temperatures, the structure became so heavily smeared that we were unable to obtain a value for $\Delta_>+\Delta_<$. Similar behavior was observed at 2 mW. At the higher power levels, however, the structure in dV/dI *abruptly* disappeared when we exceeded the temperature $T_\downarrow$. $T_\downarrow$ was substantially higher than T_ν, and increased with increasing microwave power. The curve labeled 18 mW gave a $T_\downarrow \approx T_{c>}$ within experimental error. Further increases in power did not result in any further increase in $T_\downarrow$. Thus, as the temperature was increased at constant power ($\geqslant 3$ mW), $2\langle\Delta\rangle$ was maintained at a value greater than $h\nu$ until an instability occurred at $T_\downarrow$, and there was a first-order transition to zero gap. When the temperature was reduced (keeping the power constant) there was no structure in dV/dI until we reached a temperature $T_\uparrow$, where the gap abruptly reappeared. $T_\uparrow$ decreased with increasing power. Thus we observed hysteresis in the plot of $2\langle\Delta\rangle$ vs temperature at constant microwave power. This hysteresis is reminiscent of that observed by Klapwijk and co-workers[4] in the resistive transition between the normal and superconducting states of Al strips in the presence of 3-GHz microwaves.

Our results are qualitatively consistent with the predictions of Eliashberg and co-workers,[1] who find that, at higher temperatures, Δ becomes a double-valued function of temperature in the presence of microwave power (see, for example, Ivlev and co-workers,[1] Fig. 1). At a given power level there is a maximum temperature, T_{max}, at which superconductivity exists: At this temperature, Δ is nonzero. The theory predicts that there will be a first-order transition to the normal state as the temperature is increased above T_{max}. As the temperature is lowered from above T_{max}, the theory predicts a first-order transition back to the superconducting state somewhere in the temperature interval over which Δ is double-valued, thereby allowing the possibility of hysteresis.

Method 2.—We increased the microwave power from zero, keeping the temperature constant. At temperatures below $T_\square$, marked (□) in Fig. 3, we obtained the same enhanced values of the gap as in Method 1, whereas, at temperatures $T_\square$ and above, the behavior was markedly different. At $T_\square$, there was a small enhancement at low power levels, but, as the power was increased above 3 mW, the structure in dV/dI disappeared. At higher temperature, no enhancement at all was observed; rather, the gap became zero as the power was increased. For the junction represented in Fig. 3, $T_\square$ was roughly 1 mK above T_ν, the temperature at which $h\nu = 2\langle\Delta_e\rangle$. Evidently, when the temperature was raised to $T_\square$, the pair breaking effects of the microwaves and of the phonons emitted by quasiparticle relaxation began to reduce the enhancement; as the temperature was increased further, no enhancement of the gap was possible by Method 2. The frequency dependence of this effect is shown in the inset of Fig. 3, where the change in $2\langle\Delta\rangle$, $\delta(\Delta_> + \Delta_<)$, is plotted versus temperature for a junction on a glass substrate. These data were taken in the low-power regime where Methods 1 and 2 were indistinguishable. At each of the two frequencies (9.9 and 11.9 GHz), there was an enhancement for $T \lesssim T_\nu$ while $\langle\Delta\rangle$ was depressed for $T \gtrsim T_\nu$. This result is consistent with the observation by Tredwell and Jacobsen[3] that the critical current of aluminum microbridges was enhanced by 10-GHz phonons only when $h\nu < 2\Delta_e$. The depression of $\langle\Delta\rangle$ for $T \gtrsim T_\nu$ suggests that 10-GHz microwaves do not enhance the transition temperature of aluminum by Method 2. Since Klapwijk and co-workers[4] found that T_c was enhanced by 3-GHz microwaves, the degree of enhancement is evidently strongly frequency dependent.

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Observation of a Charge Transfer During Bi Oxidation as Noted from the Final-State Changes*†

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By monitoring the change in the conduction-band optical density of states (CBODOS) and the bonding shifts of the core levels, we observe an electronic transfer from Bi to oxygen atoms during Bi oxidation. Similar analysis of the CBODOS of other Bi and Pb chacogenides indicates a partial ionicity in these compounds which correlates with electronegativities and the chemical shifts of the core levels.

The observation of changes in electronic structure during chemisorption and/or oxidation has been confined primarily to monitoring the changes of the occupied, i.e., valence-band, density of states (VBDOS) and core-level shifts. However, a charge redistribution of valence electrons, reflecting formation of new bonds, can also result in changes in the conduction band. We report here the first observation of changes in the (empty) conduction-band density of states (CBDOS), noted during the oxidation of Bi. Because of a strong spin-orbit (SO) splitting of the conduction band (CB) of bismuth oxide, we can determine the predominant total-angular-momentum component of Bi valence electrons which are transferred to oxygen atoms during formation of Bi_2O_3. Specifically, during formation of Bi_2O_3, we observe in the CBDOS the onset of a dominant peak due to states of mainly $6p_{1/2}$ character which in the atomic ground state are fully occupied. These empty $6p_{1/2}$ states in Bi_2O_3, reflecting the redistribution of Bi valence electrons, are strongly localized at Bi atoms as evidenced by their very small width [~ 0.5 eV full width at half-maximum (FWHM)]. The bonding shifts (1.9 eV) of the core levels, observed simultaneously with the onset of