of the phonon wind on EHD in our experiment by linearly scaling the experimental results of Bagaev et al.<sup>2</sup> for this force to our excitation power  $P \sim 2 \text{ mW cm}^{-2}$  from Figs. 3(a) and 3(b). This estimate includes the effects of phonons due to the thermalization of the excited carriers and phonons produced in the Auger decay of drops.<sup>2</sup> Using<sup>8</sup>  $\tau_{p} = 1.6$  nsec, and<sup>4,9</sup>  $R \simeq 0.2 \ \mu$ m, we find  $F_{b} \sim 10^{-14}$  dyn for the force on an entire drop due to the phonon wind at 2.0 K. This is a factor of ~  $10^5$  smaller than the force  $F_i \sim B/d \sim 6 \times 10^{-9}$ dyn necessary to remove a drop from an impurity atom, where  $d \sim 20$  nm is the EHD surface thickness. The force on EHD under the same conditions due to the FE wind<sup>11</sup> is  $F_{\rm FE} \sim 3 \times 10^{-16}$ dyne, a factor of  $\sim 10^2$  smaller than  $F_{p}$  above. Thus we expect EHD to remain rigidly pinned to impurities at the excitation levels used in this experiment, in agreement with our data. At higher T the phonon-wind force  $F_p$  increases dramatically as a result of increases<sup>4,8,9</sup> in the threshold excitation  $P_+$ , in the drop radius R, and in EHD coupling with phonons; at 3.5 K,  $F_p$  is comparable to  $F_i$  above and sufficient to remove EHD from their pinning sites. This result is consistent with the experimental disappearance<sup>4</sup> of hysteresis effects at temperatures  $T \gtrsim 3$  K.

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## Measurement of Characteristic Time Constants of Microwave-Enhanced Superconductivity in Aluminum Films

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Time constants of the processes causing the microwave enhancement of the critical current in superconducting Al films have been determined by measuring the response to an amplitudemodulated microwave signal. The dominant time constant corresponds to the quasiparticlerecombination time, while for  $T/T_c < 0.9$  a second time constant similar to the inelastic scattering time is also found. These observations confirm the results of Chang and Scalapino that increased recombination is more important than redistribution in enhancing the energy gap.

Nonequilibrium phenomena in superconductors have been the subject of an appreciable number of papers in the last few years. An important property is the reported microwave-enhancement of the critical current<sup>1-5</sup> and the energy gap.<sup>6</sup> The first microscopic explanation of the enhancement effect is given by Ivlev, Lisitsyn, and Eliashberg.<sup>7</sup> In their model the microwave photons cause a redistribution of the quasiparticles to higher energies, where they are less effective in depressing the order parameter. Numerical calculations by Chang and Scalapino<sup>8</sup> show that it is not only this redistribution that causes an enhancement of the gap. An even larger enhancement is caused by the resulting net decrease of the total number of quasiparticles because of their increasing recombination rate into Cooper pairs with increasing energy.

In this Letter we report measurements of the response of the critical current in Al strips to an amplitude-modulated microwave signal with a varying modulation frequency. From these measurements time constants related to the enhancement effect can be obtained. We first give a short description of the measurement method, then present a number of results of the measurements and finally a brief discussion of the results.

The sample on which the measurements are made is a  $0.1-\mu$ m-thick,  $2-\mu$ m-wide, and  $40-\mu$ m-long Al film evaporated on a silicon slice.<sup>4</sup> It is immersed in a temperature-regulated He bath.

The microwave signal which is supplied to the sample is obtained by superposition of the signals from two generators, the main one at a fixed frequency f (8 GHz for the reported results), the second one at a frequency  $f + \Delta f$ . This results in an amplitude-modulated microwave signal with modulation frequency  $\Delta f$  and with a modulation depth depending on the power ratio of the two generators. Now the measurement consists in recording the resulting modulation amplitude of the critical current as a function of  $\Delta f$  by sweeping the frequency of the second generator while keeping its delivered power to the sample constant. We now briefly describe how this is realized.

The critical current is determined by the ap-



FIG. 1. (a) Current through the sample as function of time, (b) on-off pulse of main generator, and (c) onoff pulse of sweep generator.

plication of a linearly increasing current through the sample and by measuring its value when this current causes the film to switch to its normal state. Now the linearly increasing current ramp is repeated periodically as illustrated in Fig. 1(a) while both generators are periodically switched on and off according to Figs. 1(b) and 1(c).<sup>9</sup> In this way the critical-current values measured during the successive current ramps 1-4 (Fig. 1) are (1) the critical current without microwaves.  $I_{co}$ ; (2) the enhanced critical current  $I_c$  caused by the microwave power  $P_m$  of the main generator; (3) the enhanced critical current  $I_c'$  caused by the power  $P_s$  of the sweep generator; and (4) the minimum value  $I_c - |\Delta I_c|$  caused by the amplitudemodulated microwave power  $P_m + P_s$ . This fourth value needs some clarification. As a result of the modulation of the microwave power with frequency  $\Delta f$  the enhanced critical current of the sample will also be modulated:  $I_c + \operatorname{Re}(\Delta I_c e^{j\Delta\omega t})$ . The strip switches to its normal state when the linearly increasing current through the sample reaches this modulated critical current. This occurs practically at  $I_c - |\Delta I_c|$  because we have chosen the increase of the current through the sample during one period of the modulation to be very small compared to  $|\Delta I_c|$  for all relevant modulation frequencies. Now with one synchronous detector the difference  $I_c' - I_{c0}$  is determined. With a feedback loop to the power regulation of



FIG. 2. Recorded response curve of  $|\Delta I_c(\Delta f)|$  at T = 1.06 K with  $P_s/P_m = 0.1$  and with attenuation factor  $\alpha = 40$  dB. The upper curve is the central peak of the lower curve on an extended frequency scale. The dashed curve is  $|a_1/(1+j\Delta\omega\tau_1)+a_2/(1+j\Delta\omega\tau_2)|$  with  $\tau_1 = 190$  nsec and  $\tau_2 = 2.5$  nsec.

the sweep generator this difference and consequently the power from this generator reaching the strip are kept constant when the frequency is swept. A second synchronous detector measures the difference  $|\Delta I_c|$  in the critical-current values during periods 2 and 4 and it is recorded as a function of  $\Delta f$ .

A result of a recorded response curve is shown in Fig. 2, measured at an attenuation factor  $\alpha$  of the microwave power of 40 dB, which means a relatively small deviation from equilibrium (see Fig. 3). It is shown in the figure that the response curve reasonably fits with the modulus of the superposition of two responses of the form a/(1 $+j\Delta\omega\tau)$  with time constants  $\tau_1 = 190$  nsec and  $\tau_2$ = 2.5 nsec, respectively, and an amplitude ratio  $a_2/a_1 = 1.1$ . With increasing temperature and with increasing microwave power  $a_2/a_1$  is found to decrease. For  $T/T_c > 0.9$  and  $\alpha < 35$  dB the ratio has become so small that the second response is no longer detectable.

We have measured the time constant  $\tau_1$  as a function of  $\alpha$ . The results are plotted in Fig. 3, together with the critical-current values. It proved that for  $\alpha \ge 23$  dB the response could well be described with this time constant  $\tau_1$ . For higher microwave powers the response became more complex. For  $\alpha = 44$  dB and so in the lowpower region where  $\tau_1$  is independent of  $\alpha$  we determined  $\tau_1$ ,  $\tau_2$ , and  $a_2/a_1$  at different temperatures. The results are given in Fig. 4 as a func-



FIG. 3. Critical current  $I_c$  and characteristic time  $\tau_1$  as a function of the attenuation factor  $\alpha$  of the microwave power for T = 1.0, 1.1, and 1.2 K. The main microwave frequency is 8 GHz.



FIG. 4. Measured values of  $\tau_1$ ,  $\tau_2$ , and  $a_2/a_1$  as a function of  $\Delta/kT$ . The full drawn line has the theoretically expected  $T^{-1/2}e^{\Delta/kT}$  behavior for  $\tau_R$ .

tion of  $\Delta/kT$ , where the energy gap  $\Delta$  is determined by the BCS formula  $\Delta/\Delta_0 = 1.74(1 - T/T_c)^{1/2}$  with  $T_c = 1.3$  K for our sample.

For high power levels  $\alpha < 23$  dB a dip occurs in the top of the response curve as illustrated in Fig. 5 for the curve at  $\alpha = 20$  dB. With decreasing  $\alpha$  the dip becomes more important and at  $\alpha$ 



FIG. 5. Recorded response curves of  $|\Delta I_c(\Delta f)|$  at T = 1.0 K with  $P_s/P_m = 0.03$  and at  $\alpha = 20$ , 17, and 13 dB. At  $\alpha = 17$  dB the enhanced critical current is maximal.

= 17 dB, where the maximum value of  $I_c$  is reached, the dip has become so large that  $|\Delta I_c|$ is 0 at  $\Delta f = 0$ . For still higher values the dip turns upwards again as illustrated by the curve at  $\alpha = 13$  dB.

Taking an overall view of these results we would make the following observations:

1. For small deviations from the equilibrium value of the critical current the time constant  $\tau_1$  which is found turns out to be practically equal to the recombination time  $\tau_R$  of the quasiparticles. This is illustrated in Fig. 4, where the full-drawn line has the theoretically expected T dependence<sup>10</sup>  $\tau_R = \Gamma_R T^{-1/2} e^{\Delta/kT}$  with  $\Gamma_R = 4.7 \times 10^{-8}$  sec K<sup>1/2</sup>, a value in good agreement with reported results.<sup>11</sup> These observations support the calculations of Chang and Scalapino,<sup>8</sup> who found that the increased recombination rate made a greater contribution to the enhancement effect than the redistribution effect on its own.

2. The interpretation of  $\tau_2$  is less clear. It fits nicely in Chang and Scalapino's model, if it is the inelastic scattering time for quasiparticles. Previously reported values for this time<sup>12</sup> are indeed within the same order of magnitude as  $\tau_2$ . The experimental trend for  $\tau_2$  to increase with increasing T is not understood, but this trend is not very clear as a result of the experimental uncertainty in  $\tau_2$  at low values of  $a_2/a_1$ . At  $\Delta/kT$ = 1.2 we estimate  $\tau_2$  to be correct within a factor of 2, whereas the uncertainty at  $\Delta/kT = 2$  is  $\pm 10\%$ .

3. We found  $\tau_1$  to be larger than  $\tau_2$ , whereas calculations<sup>13</sup> predict the intrinsic recombination time to be smaller than the scattering time. This can be understood by realizing that the experimental recombination time is larger than the intrinsic one because of photon trapping.<sup>14</sup>

4. To understand the relative importance of the effects with time constants  $\tau_1$  and  $\tau_2$  as expressed by  $a_2/a_1$  as a function of *T*, more calculations are needed. The only calculated point is at  $T/T_c = 0.9$  or  $\Delta/kT \sim 1$  where a value  $a_2/a_1 \sim 0.3$  is found.<sup>8</sup> This value is reasonably close to our experimental results.

5. For increasing microwave power the time constant  $\tau_1$  decreases. This also fits qualitatively in Chang and Scalapino's model because of the increasing average recombination rate of quasiparticles when they are excited to higher energies.

6. For microwave energies near the maximum value of the enhanced critical current a new process is seen to occur that has a decreasing effect on the critical current and time constant  $\sim 400$  nsec causing a dip in the response curves. This

effect is probably due to heating of the sample by the microwaves and the corresponding time constant should then be equal to the thermal relaxation time of the sample. The importance of heating near the maximum of  $I_c$  is confirmed by our observations that an increase in the thermal resistance of the sample to the He bath causes a decrease in the enhanced critical current  $I_c$  at the same microwave power, an effect also recently reported on by Klapwijk, van Linden van den Heuvel, and Mooy.<sup>15</sup>

In conclusion, we feel that we have shown that frequency-response measurements of the microwave enhancement of critical currents of superconducting films can give much information about the microscopic processes that take place in the superconductor. More specifically, our measurements support the calculations of Chang and Scalapino. It would appear that the increased recombination of quasiparticles into pairs under microwave irradiation is of major importance for understanding of the enhancement effect. A more complete understanding of the measured temperature and microwave power dependences of the frequency response with the corresponding time constants requires further calculations to be carried out.

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## Oxidation-Induced Breakdown of the Conservation of Perpendicular Momentum in the Angle-Resolved Photoelectron Spectra of Cu(111)

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It is shown that changes in the angle-resolved spectra of a Cu(111) surface brought about by oxidation can be largely explained in terms of an increase in the breakdown of the conservation of the component of the momentum perpendicular to the surface in the photoionization matrix element of the wave function of the bulk.

Several workers have investigated the changes in the photoelectron spectra of solids brought about by chemisorption of atoms on the surface of clean metals.<sup>1-4</sup> In spite of the large amount of work done in this area, the mechanisms that change the photoelectron spectra are not well understood. Changes are usually attributed to the removal of surface states and to the addition of peaks associated with the atomic orbitals of the adsorbate. Anderson and Lapeyre<sup>4</sup> have shown that alteration of the size of the surface Brillouin zone by absorbates can modify the probability of emission from states associated with bulk states.

In this Letter we report the results of angleresolved photoemission experiments performed with polarized light on the Cu(111) surface. We find that the pronounced changes occurring in the spectra when the surface is oxidized can be explained by an increase in the relaxation of the conservation of the component of momentum perpendicular to the surface,  $k_{\perp}$ . This is a new mechanism, which, as far as the authors know, has not been suggested before to account for changes in the angle-resolved photoelectron spectra of clean metals induced by chemisorption.

The spectra were obtained in a commercial angle-resolved photoelectron spectrometer, for which we have constructed and added a polarizer for our discharge lamp. This polarizer allows the electric field of the incident photons to be oriented at any azimuthal direction about the Poynting vector. The sample was a Cu(111) surface which was cleaned by several cycles of ion bombardment and electron-beam heating. The angular and energy resolutions of the spectrometer were  $\pm 2^{\circ}$  and 0.15 eV, respectively. The degree of polarization of the light was ~90%, while typical peak intensities of  $2 \times 10^3$  counts/sec were achieved. The oxidized surface was produced by exposing the clean surface to 1000 L (1L =  $10^{-6}$ Torr sec) of oxygen. Low-energy electron-diffraction (LEED) measurements showed that the oxidized surface had the same symmetry and surface spacing as the clean surface. Only the energy dependence of the diffraction peaks was altered upon oxidation.

We show in Fig. 1 representative spectra from the  $\Gamma LU$  and  $\Gamma LK$  planes<sup>5</sup> of clean and oxidized Cu taken with *p*-polarized Ne I and He I radiation, for polar angles of emission  $\theta = 45^{\circ}$  and  $\theta = 55^{\circ}$ , respectively. The corresponding spectra taken with *s*-polarized light are shown in Fig. 2. The spectra for clean Cu taken with Ne I and He I radiation exhibit a strong dependence on the polarization direction, which can be explained by a theoretical model proposed earlier.<sup>6</sup> The initial states contributing to the spectra obtained with *s*- and *p*-polarized light have odd and even reflection symmetry with respect to the plane of emission, respectively.

The angle-resolved spectra obtained with ppolarized light shown here exhibit a much greater sensitivity to oxidation than previously reported angle-averaged spectra.<sup>1</sup> The most dramatic change induced by oxidation is the addition of a peak at 3-eV binding energy in the spectra ob-