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## Pair breaking in strong-coupling superconductors

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The tunneling density of states of superconducting lead films has been measured in parallel magnetic fields. The magnetic-field dependence of the typical phonon structure in lead is analyzed by incorporating strong-coupling effects into the well-known weak-coupling theory of superconductors under pair-breaking conditions.

In the past several years, strong-coupling effects in superconductivity as well as pair-breaking effects have been extensively studied and seem to be very well understood.<sup>1</sup> However, little is known about the combination of strong-coupling superconductivity under pair-breaking conditions. In view of the rapidly growing research in the electron-phonon interaction in metals, it seems to be of some interest to study a strong-coupling superconductor (strong electron-phonon interaction) in the pair-breaking situation, i.e., in a magnetic field. In this paper we present experimental results for the tunneling density of states of thin lead films in parallel magnetic fields, showing small field-dependent deviations from the usual weak-coupling behavior. These results can be analyzed theoretically by extending the usual Abrikosov-Gorkov theory of pair breaking in superconductors<sup>2</sup> to include the specific strong-coupling effects of the electron-phonon interaction.3,4 Similar studies for weak-coupling materials (Sn), however showing no anomalous behavior due to the phonon structure, have been presented by Levine<sup>5</sup> and by Millstein and Tinkham.6

The measurements were performed on aluminumlead tunnel junctions, consisting of a 50-nm-thick aluminum film and a lead film with varying thickness between 27 and 75 nm, separated by a natural grown aluminum oxide layer. Electrical contacts were made by pressing indium-coated leads directly on the films. The junctions were mounted in the vertical position in a superconducting split coil magnet; this enabled us to align the film with the magnetic field by simply rotating the junction holder from outside the cryostat. Accurate aligning within ±0.1° was obtained by monitoring the resistance transition of the lead film at its steepest point. Recordings of first and second derivative of the junction current-voltage characteristic (dV/dI and  $d^2V/dI^2$ ) were taken using a bridge circuit based on the original design of Adler and Jackson.7 A low noise transformer and a preamplifier were used for the  $d^2 V/dI^2$  measurements; this enabled us to study the phonon structure up to the critical field, where the signal level is very low. In all measurements, the aluminum film was in the normal state.

Figure 1 shows the tunneling density of states for various magnetic fields obtained from dV/dI measurements, together with theoretical weak-coupling curves. These theoretical curves have been calculated by smearing out the zero-temperature tunneling density of states of a weak-coupling superconductor under pair-breaking conditions in the limit of a small mean-free path  $/(1/\xi_0 \rightarrow 0; \xi_0;$  coherence length)<sup>8</sup> with a finite-temperature distribution function in the usual way. The overall features of the experiment are in reasonable agreement with this weak-coupling theory, the small deviations are probably due to effects of a finite mean-free path.9 Figure 2 shows the tunneling density of states of the AI-Pb junction including the region far above the gap. This plot shows the wellknown deviations from BCS theory at energies  $\omega_i$  and  $\omega_l$ , slightly below  $\omega_{ln} + \Delta_{00}$  and  $\omega_{ln} + \Delta_{00}$ , respectively.<sup>10</sup> Here,  $\omega_l$  and  $\omega_l$  denote the positions of the peaks in  $d^2 V/dI^2$ ,  $\Delta_{00} = \omega_s (H=0)$  is the energy gap in the absence of a magnetic field, and  $\omega_{ln}$  and  $\omega_{ln}$ are, respectively, the typical transverse and longitudinal phonon energies in lead. These deviations can be studied with higher resolution by measuring the second derivative  $(d^2 V/dI^2)$  of the junction characteristic. Figure 3 shows recorder tracings of this second derivative which have peaks at the energies  $\omega_t$ and  $\omega_i$ . (Note that there are peaks and not dips as one would expect from a derivative of the density of states, because  $d^2 V/dI^2$  is measured rather than  $dI^2/dV^2$ .)

To explain these experimental results, showing a small but clear shift of the phonon peaks as a function of the magnetic field, the usual well-known weak-coupling theory of superconductors under pairbreaking conditions has to be extended to include explicitly details of the electron-phonon interaction.

Most generally, the full (matrix) Green's function,

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FIG. 1. The normalized density of states of Pb obtained from tunneling measurements of an AI-Pb tunnel junction for various values of the parallel magnetic field. The dashed lines give the numerical results for a weak-coupling superconductor with pair breaking and thermal smearing in the limit of a small mean free path  $(1/\xi_0 \rightarrow 0)$ .

which describes electrons in a superconductor in the presence of randomly distributed impurities and pair breaking, is given in the limit of a short mean-free path by, using Maki's notation,<sup>11</sup>

$$G_{\omega}^{-1} = i \tilde{\omega} \underline{1} - \xi \rho_3 - \tilde{\Delta} \rho_1 \cdot \sigma_2 \quad , \tag{1}$$

where  $\tilde{\omega}$  and  $\tilde{\Delta}$  are the renormalized frequency and order parameter, and  $\rho_i$  and  $\sigma_i$  are Pauli matrices operating on different spaces ( $\sigma_i$  operates on the ordinary spin states, while  $\rho_i$  operates on the space composed of the electron and hole states). The Green's function, taking explicitly into account strong-coupling effects, is given in the absence of impurities by<sup>12</sup>

$$G_{\omega}^{0^{-1}} = i\omega Z - \xi \rho_3 - \Phi \rho_1 \cdot \sigma_2 \quad , \tag{2}$$

where Z and  $\Phi = Z \Delta$  are renormalization parameters due to strong-coupling effects. In the Born approximation and making use of Migdals theorem, impurity scattering in the presence of pair breaking leads to the following set of equations:

$$\overline{\omega} = \omega + \frac{1}{2Z} \left[ \frac{1}{\tau_1} + \frac{1}{\tau_2} \right] \frac{\overline{\omega}}{(\overline{\omega}^2 + \overline{\Delta}^2)^{1/2}} , \qquad (3)$$

$$\overline{\Delta} = \Delta + \frac{1}{2Z} \left( \frac{1}{\tau_1} - \frac{1}{\tau_2} \right) \frac{\overline{\Delta}}{(\overline{\omega}^2 + \overline{\Delta}^2)^{1/2}} \quad , \tag{4}$$

where the renormalized order parameter  $\overline{\Delta}$  and the frequency  $\overline{\omega}$  are given by

$$\overline{\Delta} = \overline{\Delta}/Z \quad , \tag{5}$$

$$\bar{\omega} = \tilde{\omega}/Z$$
 . (6)

Here,  $\tau_1$  is the normal impurity scattering lifetime while  $\tau_2$  can be related to the magnetic field.<sup>11</sup> Introducing the auxiliary parameter *u* defined by

$$u = \frac{\overline{\omega}}{\overline{\Delta}}$$
(7)

and performing an analytical continuation to real fre-



FIG. 2. The normalized density of states of Pb, for a wider energy range than Fig. 1, exhibiting strong-coupling effects. The dashed lines give the numerical results for a weak-coupling superconductor with pair breaking and thermal smearing in the limit of a small mean free path  $(l/\xi_0 - 0)$ .

quencies, one gets from Eqs. (3) and (4)

$$\frac{\omega}{\Delta} = u \left[ 1 - \zeta \frac{1}{(1 - u^2)^{1/2}} \right] .$$
 (8)

where

$$\zeta = 1/(\tau_2 \Phi) \quad . \tag{9}$$

The phonon part of the self-energy gives the following equations for the strong-coupling renormalization parameters in the limit of zero temperature<sup>4, 12</sup>:

$$\Delta(\omega) = \frac{1}{Z(\omega)} \int_0^\infty d\omega' f_0(\omega') [g_+(\omega, \omega') - \mu^*] \quad , \quad (10)$$

$$[1 - Z(\omega)]\omega = \int_0^\infty d\omega' f_1(\omega') \cdot g_+(\omega, \omega') \quad , \qquad (11)$$

with

$$f_s(\omega) = \operatorname{Re}\left\{\frac{u^s}{(u^2 - 1)^{1/2}}\right\}$$
(12)

and

$$g_{\pm}(\omega, \omega') = \int_{0}^{\infty} d\Omega \alpha^{2}(\Omega) F(\Omega) \\ \times \left[ \frac{1}{\omega' + \omega + \Omega} \pm \frac{1}{\omega' - \omega + \Omega} \right] . \quad (13)$$

Here  $\alpha^2(\Omega)F(\Omega)$  represents the energy-dependent electron-phonon interaction and  $\mu^*$  the Coulomb pseudopotential. In Eqs. (8), (10), (11), and (12)  $\omega$ should be understood as having an infinitesimal positive imaginary part. It follows from Eq. (8) that the

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FIG. 3. Recorder tracings of second derivative measurements for various values of the parallel magnetic field. The modulation voltage was 250  $\mu$ V, the temperature 1.74 K.

energy gap  $\omega_g$  is given, formally as in the case of a weak-coupling superconductor, by the maximum value of  $\omega$  in the region 0 < u < 1, if the imaginary part of  $\Delta$  is negligible. If the  $\omega$  dependence of  $\Delta$  can be neglected as well, one gets

$$\omega_{\rm g} = \Delta_0 (1 - \zeta_{\rm g}^{2/3})^{3/2} \ . \tag{14}$$

Here

$$\zeta_g = \frac{1}{\tau_2 Z_0 \Delta_0} = \frac{\Gamma}{\Delta_{00}} \frac{\Delta_{00}}{\Delta_0} , \qquad (15)$$

where  $\Delta_0$  is the order parameter at the gap frequency,  $\Delta_0 = \Delta(\omega_g)$ , and similarly  $Z_0 = Z(\omega_g)$ .  $\Delta_0$  and  $Z_0$ can in principle be calculated by a simultaneous solution of Eqs. (10) and (11). The pair-breaking parameter  $\Gamma$  can be related to the ratio of the applied parallel magnetic field  $H_{\rm fl}$  to the critical field  $H_{\rm cll}$  at which  $\Delta \rightarrow 0$ ,

$$\Gamma = \frac{1}{\tau_2(1+\lambda)} = \frac{1}{2} \left( \frac{H_{\parallel}}{H_{e\parallel}} \right)^2 \frac{\Delta_{00}}{\gamma} \quad , \tag{16}$$

where  $\Delta_{00}$  is the superconducting order parameter in the absence of magnetic fields,  $\Delta_{00} = \Delta_0 (\Gamma = 0)$ ,  $1 + \lambda = Z (\omega = 0, T = 0)$  in the normal state  $(\Delta = 0)$ , and  $\gamma \neq 1$  accounts for strong-coupling effects. Note that the assumptions leading to Eq. (14) are satisfied if  $\Gamma \ll \omega_I$ ,  $\omega_I$ . The parameter  $\gamma$  must be obtained from Eqs. (8), (10), and (11) in the limit

$$H_{\parallel} = H_{c\parallel}, \Delta = 0$$
, i.e., with  $f_1 = 1$  and

$$f_0(\omega) = \operatorname{Re}\left(\frac{\Delta(\omega)}{\omega + i[\Delta_{00}(1+\lambda)/Z(\omega)](1/2\gamma)}\right) . (17)$$

The equation for  $T_c$  in the limit  $H_{\parallel}=0$  is again given by Eqs. (10) and (11) with  $f_1=1$ , but

$$f_0(\omega) = \operatorname{Re}[\Delta(\omega)] \frac{1}{\omega} \tanh \frac{\omega}{2T_c}$$
 (18)

If the  $\omega$  dependence and the imaginary parts of  $\Delta(\omega)$ and  $Z(\omega)$  are ignored, we note that both expressions for  $f_0(\omega)$  acquire the same asymptotic  $\omega^{-1}$  dependence with low-frequency cutoffs proportional to  $\Delta_{00}/\gamma$  and  $T_c$ , respectively. Neglecting terms of order  $(\Delta_{00}/\omega_{t,l})^2$  or  $(T_c/\omega_{t,l})^2$  one finds that Eqs. (10) and (11) then lead to the relation

$$\gamma = \frac{2\Delta_{00}}{3.52\,T_c} \quad . \tag{19}$$

In the case of lead, where  $2\Delta_{00}/T_c = 4.3$ ,<sup>12</sup> this gives  $\gamma \approx 1.22$ . However,

$$\frac{\Gamma}{T_c} \approx 0.88 \left( \frac{H_{\parallel}}{H_{e\parallel}} \right)^2 , \qquad (20)$$

just as in the weak-coupling limit. A more accurate result can be obtained from the calculations of Rainer and Bergmann<sup>13</sup> of strong-coupling effects on the bulk nucleation field  $H_{c2}$ . Substituting  $(H_{\parallel}/H_{c\parallel})^2$  for  $H/H_{c2}$ ,<sup>11</sup> one obtains, using their notation,

$$\Gamma_{sc} = \eta_{c2} \left( \frac{H_{\parallel}}{H_{c\parallel}} \right)^2 T_c h_{c2}^{BCS} \quad . \tag{21}$$

With  $h_{c2}^{BCS} = 0.88$  and  $\eta_{c2} = 1.24$  for Pb at T = 0, this leads to

$$\frac{\Gamma}{T_c} = 1.09 \left( \frac{H_{\parallel}}{H_{c\parallel}} \right)^2 . \tag{22}$$

Therefore, one gets

$$\frac{\Gamma}{\Delta_{00}} = \left(\frac{\Gamma}{T_c}\right) \left(\frac{T_c}{\Delta_{00}}\right) = 0.507 \left(\frac{H_{\parallel}}{H_{c\parallel}}\right)^2 , \qquad (23)$$

leading to  $\gamma \approx 0.99$ . Thus it appears that  $\Gamma/\Delta_{00}$  remains close to its weak-coupling value thanks to a remarkable cancellation of strong-coupling effects.

For a detailed theory of a strong-coupling superconductor under pair-breaking conditions, the set of Eqs. (8) to (13) has to be solved numerically, just as in the case without pair breaking.<sup>4,12</sup> However, even without elaborate computer calculations, one is able to understand the main features of the experimental results, i.e., the shift of the phonon peaks as a function of the magnetic field, by considering some simple bounds in the weak-coupling limit. Such a procedure seems to be justified by the fact that  $\gamma$  is not

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different from the weak-coupling value ( $\gamma = 1$ ). Furthermore, as shown in Figs. 1 and 2 and discussed above, the overall features of the experimentally measured tunneling density of states are in qualitative agreement with weak-coupling theory, i.e.,  $\Delta_0$ as a function of the magnetic field appears to scale according to the usual pair-breaking theory. The field dependence of the typical phonon anomalies in the tunneling density of states (Fig. 3) can be discussed by using Eq. (10). The normalized density of states is given by

$$\frac{N(\omega)}{N_0} = \operatorname{Re}\left[\frac{\overline{\omega}}{(\overline{\omega}^2 - \overline{\Delta}^2)^{1/2}}\right] = f_1(\omega) \quad . \tag{24}$$

In the vicinity of singularities in the phonon density of states, the electronic density of states will deviate significantly from the weak-coupling result. This can qualitatively be understood by expanding Eqs. (8) and (24) in powers of  $u^{-1}$  and  $\Delta/\omega$  and  $\Gamma/\omega$ , which is justified for energies  $\omega \sim \omega_t$  or  $\omega_t$ . To lowest order in  $\Gamma/\omega$  one finds

$$\frac{N(\omega)}{N_0} \simeq 1 + \frac{(\text{Re}\Delta)^2}{2\omega^2} - \frac{(\text{Im}\Delta)^2}{2\omega^2} \quad . \tag{25}$$

A peak in the imaginary part of  $\Delta$  will cause a sharp decrease in the density of states, giving a sharp dip in the derivative of Eq. (25), or, equivalently, a peak in the experimentally measured  $d^2 V/dI^2$  curve. If the electron-phonon interaction  $\alpha^2 F$  is approximated by a delta function  $\delta(\omega - \omega_0)$ , it follows from Eq. (10) that Im $\Delta$  starts being nonzero due to strong-coupling effects at the frequency  $\omega_g + \omega_0$ , where  $\omega_0$  is the relevant phonon energy and  $\omega_g$  the lowest energy state available for an electron in the superconductor (energy gap). The actual structure will peak at a somewhat higher energy, related to the position of



FIG. 4. The measured energy shift of the longitudinal and transverse phonon peaks in Pb plotted as a function of  $(H_{\parallel}/H_{cll})^2$ . The solid line gives the lower limit (energy gap), while the dashed line gives the upper limit, as inferred from a simplified strong-coupling theory.

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the maximum of

$$\operatorname{Im}\Delta(\omega) \simeq \frac{f_0(\omega - \omega_0)}{\operatorname{Re}Z(\omega)} - \frac{\operatorname{Re}\Delta(\omega)}{\omega} f_1(\omega - \omega_0) \quad . \tag{26}$$

The behavior of  $\Delta(\omega)$  near  $\omega_0$  is dominated by  $f_0$ . The frequency  $\omega_m$  where  $f_0$  has its maximum value can easily be calculated in the weak-coupling limit [defined by  $g_+(\omega, \omega') \cong g_+(0,0) = \lambda$  in Eq. (10)]. The peak positions obtained in this way will be shifted to lower energies by incorporating strong-coupling effects through  $g_+(\omega, \omega')$ . Therefore, the frequency  $\omega_g + \omega_0$  must give a lower limit for the experimentally observed shift of the peaks with magnetic field, while an upperbound for the shift of the strong-coupling structure is given by  $\omega_m + \omega_0$ .

In Fig. 4 the measured energy shift of both the longitudinal and transverse peaks is plotted as a function of  $(H_{\parallel}/H_{e\parallel})^2$  together with the numerical results

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for the lower and upper limit from Eqs. (14) and (26). The ratio  $\Delta_0/\Delta_{00}$  has been taken over from the Abrikosov-Gorkov theory.<sup>2</sup> The figure shows that the experimental data are between the upper and lower limiting cases. Our analysis indicates that pairbreaking effects in strong-coupling superconductors can qualitatively be understood on the basis of standard theory.

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