Collective Excitations in Strong-Coupling Superconducting Alloys*

PETER FULDE AND SIGFRID STRASSLER†
Department of Physics, University of California, Berkeley, California
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The mean-free-path dependence of collective excitations in strong-coupling superconducting alloys is investigated by extension of the Fermi-liquid theory, which was developed by Larkin and Migdal for pure strong-coupling superconductors. Numerical results, which are presented, show that the dependence of the energy of transverse collective excitations on mean free path is large enough to suggest that the precursor in the infrared absorption spectrum of lead alloys is not due to collective excitations.

I. INTRODUCTION

THE onset of absorption of infrared radiation in superconducting lead and mercury at low temperatures below the energy which is required to break a pair (precursor)[1–3] has been the subject of several theoretical investigations. Absorption due to collective excitations was always considered as a possible explanation for the appearance of the precursor.

It has been known for some time that a weak-coupling theory of collective excitations disagrees with the infrared experiments in two respects: The intensity of absorption in the precursor region turns out an order of magnitude smaller than measured (see Ref. 4); and the onset of absorption is predicted to be a sensitive function of mean free path (see Ref. 5), again in disagreement with experiments. Since the precursor was, until now, found only in strong-coupling materials it is of interest to investigate whether or not a strong-coupling theory of collective excitations possesses the same shortcomings as the weak-coupling theory. In a recent investigation Larkin[6] was indeed able to derive absorption intensities, using a strong-coupling theory, which are in partial agreement with experiments. The aim of this communication is to investigate the influence of a finite mean free path on transverse collective excitations in a strong-coupling theory. This will be done by extending the theory of Larkin and Migdal[7] to the case of strong-coupling superconducting alloys.

The characteristic feature of Larkin and Migdal's approach to strong-coupling superconductors is that of a Fermi-liquid theory of Landau's type,[8] and therefore differs from the approach in Ref. 9 which starts from first principles. In a Fermi-liquid theory a number of experimentally determined functions of the direction in momentum space are introduced. They make it unnecessary to know the single-particle Green's function except near the Fermi surface, where it has a simple form. While for a normal metal it is sufficient to introduce one unknown function in order to characterize the two-particle excitation spectrum, Larkin and Migdal[7] have shown that we need two functions, which we call Γ^a, Γ^b, in order to do the same for a superconductor. Furthermore, Larkin's work shows that the onset of absorption of infrared radiation due to a collective state of quantum number l determines a certain combination of the $f_l{}^a$, $f_l{}^b$, which are the expansion coefficients of Γ^a, Γ^b in terms of spherical harmonics. While there is, in the weak-coupling case, an additional relation between $f_l{}^a$, $f_l{}^b$, which fixes $f_l{}^a$, $f_l{}^b$ as soon as the onset of absorption is fitted, such an additional relation does not exist in the strong-coupling case, and one is therefore left with one more degree of freedom. The mean-free-path dependence of the onset of absorption, which follows uniquely in the weak-coupling case, will in the strong-coupling theory depend on the specific choice of $f_l{}^a$. Nevertheless, one can show that for any physical choice of $f_l{}^a$ the mean-free-path dependence will be strong enough to suggest that the precursor in lead alloys is not due to collective excitations.

In the next section we will write down a formalism which applies to strong-coupling superconducting alloys, and in the last section the change of collective excitation energy with impurity concentration will be calculated and discussed.

II. STRONG COUPLING SUPERCONDUCTING ALLOYS

Owing to the work by Migdal[10] and Eliashberg[11] it is well known how one has to formulate the theory of superconductivity if the electron-phonon interaction is not weak. This work was later extended by Tsuneto[12] to include impurity scattering. In this section we will state some of the results of these authors. However, we

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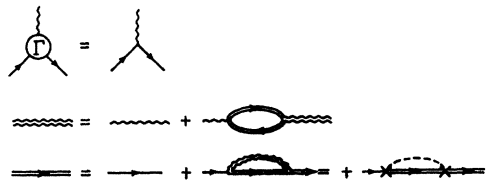

FIG. 1. Graphical representation of the equations for the vertex, phonon, and electron Green's function in the presence of impurities. Solid and wavy lines represent electrons and phonons, respectively. A double line represents the "dressed" particle. The dashed line connects two scattering events (crosses) taking place at the same impurity site.

shall rewrite the results in a form which will be more convenient in view of the following considerations.

First, we consider a normal metal in the presence of impurities. The vertex Γ (see Fig. 1) is equal to $\Gamma_0=1$ up to terms of the order $(m/M)^{1/2}$ due to phonon corrections and of order $(v_F k_F \tau)^{-1}$ due to impurity scattering.[12] Here m and M denote the electron and ion mass, respectively, and τ is the mean free time of an electron between collisions with the impurities. The phonon Green's function can be calculated with the help of Fig. 1. It is unaffected by impurity scattering as long as $\tau_q v_F \gg 1$, where $\mathbf{q}$ is the phonon momentum.[12] Finally, the equation for the electron Green's function is depicted in Fig. 1. One can show that impurity scattering leads only to an additional term $isg\omega/2\tau$ in the denominator of the Green's function as compared with the pure case.

Using these simplifications we want to calculate the Green's function in a superconductor using Nambu's formalism.[13]

We start with the zero-order Green's function

$$\hat{G}_0^{-1}(p)=p_0\tau_0-(\epsilon_p-i\delta sg\epsilon_p)\tau_3, \tag{1}$$

where $\tau_0, \tau_1, \cdots \tau_3$ denote the unit matrix and the three Pauli matrices, respectively, and ϵ_p is the single-particle energy in the normal state calculated from the Fermi surface. The equation for the full Green's function $G(p)$ reads (again, see Fig. 1)

$$\hat{G}_0^{-1}(p)=\hat{G}_0^{-1}(p)+\Sigma_{\rm ph}(p)+\Sigma_{\rm im}(p), \tag{2}$$

where $\Sigma_{\rm ph}(p)$ and $\Sigma_{\rm im}(p)$ are the self-energy due to phonons and that due to impurities, respectively. We write $\Sigma_{\rm ph}(p)$ in the form

$$\Sigma_{\rm ph}(p)=[1-Z(p)]p_0\tau_0+[Z(p)\tilde{\epsilon}_p-\epsilon_p]\tau_3+\Delta(p)\tau_1. \tag{3}$$

The self-energy $\Sigma_{\rm im}(p)$ is given by

$$\begin{aligned}\sum_{\rm im}(p)&=n_i\int\frac{d^3p'}{(2\pi)^3}v(\mathbf{p}',\mathbf{p})\tau_3\hat{G}(p')\tau_3 v(\mathbf{p},\mathbf{p}')\\&=\sum_{\rm im}{}^0\tau_0+\sum_{\rm im}{}'\tau_1.\end{aligned} \tag{4}$$

Here n_i is the impurity concentration and $v(\mathbf{p},\mathbf{p}')$ is the impurity scattering potential. For simplicity we will assume S-wave scattering only. It is convenient to introduce two new quantities $\tilde{p}_0$ and $\tilde{\Delta}$ defined by

$$\begin{aligned}Z\tilde{p}_0&=Zp_0+\Sigma_{\rm im}{}^0,\\\tilde{\Delta}&=\Delta(p)-\Sigma_{\rm im}{}'.\end{aligned} \tag{5}$$

If we solve Eq. (2), making use of the relations (3) and (4), we see that we can express $\tilde{p}_0$ and $\tilde{\Delta}$ as $\tilde{p}_0=\eta p_0$, $\tilde{\Delta}=\eta\Delta$, where η is given by

$$\eta=1+i/2\tau Z(p_0^2-\Delta_0^2)^{1/2}. \tag{6}$$

Here we have defined τ as usual by $(2\tau)^{-1}=n_i\pi|v|^2N(0)$, where $N(0)=m^*k_F/2\pi^2$ and m^* is the effective mass of the electron. Δ_0 is defined by $\Delta=\Delta_0 Z$. The quantity η resembles very much the corresponding quantity in the weak-coupling limit except for the appearance of the renormalization constant Z. Near the Fermi surface the Green's function has the simple form

$$\hat{G}(p)=\frac{1}{Z}\frac{(\eta p_0\tau_0+\tilde{\epsilon}_p\tau_3+\eta\Delta_0\tau_1)}{(\eta p_0)^2-\tilde{\epsilon}_p{}^2-(\eta\Delta_0)^2}, \tag{7}$$

where $\tilde{\epsilon}_p=p_F/m^*(|\mathbf{p}|-p_F)$. Therefore it seems appealing to apply Landau's concept of the Fermi liquid[8] which requires only the knowledge of the Green's function near the Fermi surface.

III. COLLECTIVE EXCITATIONS

The collective excitations can be viewed as exciton-like bound states between particle-particle and particle-hole. The eigenfunctions and eigenvalues of the excitons can be found by solving an equation which corresponds to the diagram drawn in Fig. 2. In Nambu's notation[13] we obtain

$$\chi(p,q)=i\int\tau_3 V(\mathbf{p}-\mathbf{k})\hat{G}(k_+)\chi(k,q)\hat{G}(k_-)\tau_3\frac{d^4k}{(2\pi)^4}. \tag{8}$$

$V(\mathbf{p}\text{-}\mathbf{k})$ stands for the potential which scatters two particles; and in diagram language corresponds to the sum of a phonon line and an impurity line, i.e., a line connecting two scattering events at the same impurity atom. The Coulomb interaction has been taken into account only insofar as it enters into the derivation of Fröhlich's Hamiltonian describing the electron-phonon interaction. Transverse collective excitations do not couple to the Coulomb field and therefore the introduction of the screened Coulomb interaction between electrons would complicate the calculations unneces-

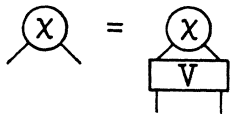

FIG. 2. Graphical representation of the exciton-like collective state χ. V represents the electron-electron interaction which in our model consists of the phonon interaction and the impurity scattering.

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sarily. Neglecting it does not change any of the results of this section. By writing out Eq. (8) explicitly, we see that in all terms which contain the product of the two diagonal elements of $\hat{G}$ the integration extends over regions far from the Fermi surface. On the other hand, the elements of $\hat{G}$ do have a simple form only near the Fermi surface where Eq. (7) holds. We therefore introduce two functions Γ^a and Γ^b, which depend on the direction in momentum space, as experimentally determinable parameters which will restrict all integrations to regions close to the Fermi surface. Γ^a, Γ^b are defined by

$$\begin{aligned}\Gamma^a&=V-iV(GG+FF)^0\Gamma^a,\\ \Gamma^b&=V-iV(G\bar{G}-FF)^0\Gamma^b.\end{aligned}\tag{9}$$

An integration over $d^4p/(2\pi)^4$ is always understood to be included. G, $\bar{G}$, and F denote the functions $G(p)=[\hat{G}(p)]_{11}$, $\bar{G}(p)=-[\hat{G}(p)]_{22}$ and $F(p)=i[\hat{G}(p)]_{12}$. The index 0 indicates that $\Omega=|\mathbf{q}|=0$. Details of how Γ^a and Γ^b can be used to eliminate regions far from the Fermi surface in integrations may be found in Refs. 6 and 7. Our function Γ^a differs slightly from the corresponding function in Ref. 6 since our definition is more appropriate in the case of finite mean free path. Since we will be interested only in how the lowest eigenvalue of the collective excitations changes, we can restrict our further considerations to the case $\mathbf{q}=0$. Furthermore, we set $\chi_1=-\chi_4$ and $\chi_2=-\chi_3$, thus losing only the unphysical eigenstate (see Ref. 5). In this way we obtain

$$\begin{aligned}\chi_1=i\Gamma^a[G_+G_-+F_+F_-&-(GG+FF)^0]\chi_1\\ &+\Gamma^a[G_+F_--G_-F_+]\chi_2,\end{aligned}\tag{10}$$

$$\begin{aligned}\chi_2=-\Gamma^b[G_+F_-+F_+\bar{G}_-]&\chi_1\\ +i\Gamma^b[G_+\bar{G}_-&-F_+F_--(G\bar{G}-FF)^0]\chi_2.\end{aligned}$$

Since the quantities in the square brackets approach zero fast enough as functions of their arguments, we may interchange the order of integrations and use for G and F the simple expressions given by Eq. (7).

First let us find the eigenvalue of Eq. (10) in the absence of impurity scattering. It is advantageous to introduce the quantities $f^a=N(0)\Gamma^a/Z^2$ and $f^b=N(0)\Gamma^b/Z^2$. The expansion coefficients in spherical harmonics we call $f_l{}^a$ and $f_l{}^\xi$ (the latter notation was used in Ref. 6 and we will adopt it here). Furthermore, we set $f_l{}^\omega=f_l{}^a/(1-f_l{}^a)$. Using the results of Refs. 5 and 6, where the integrals in Eqs. (10) were calculated, we obtain the following equation for the determination of the eigenvalue Ω:

$$1=(f_l{}^\xi\alpha^2-f_l{}^\omega)g(\alpha)\,.\tag{11}$$

Here $g(\alpha)=\arcsin x/x(1-x^2)^{1/2}$ and $\alpha=\Omega/2\Delta_0$. It is clear that the knowledge of Ω from experiments determines a certain combination of $f_l{}^\xi$ and $f_l{}^\omega$ but not the quantities themselves. The situation is therefore different from the weak coupling case where

$$f_l{}^\omega=V_l,\quad f_l{}^\xi=V_0V_l/(V_0-V_l)\,,\tag{12}$$

and V_0, V_l are the zeroth and lth harmonic of the interaction potential V multiplied by the density of states. In that case Ω determines V_l, and since V_0 is determined by the gap, $f_l{}^\omega$ and $f_l{}^\xi$ are known. For the following, we assume that we have only collective excitations with $l=2$ and want to investigate how Ω changes as impurity scattering is introduced. We distinguish two cases.

A. Low Impurity Concentrations

As long as the impurity concentration is low enough so that $2Z\tau\omega_D\gg1$, where ω_D is the Debye energy[1] we can calculate explicitly the change in the collective excitation energy as a function of mean free path. In that case inspection of Eqs. (10) shows that the integration extends over regions near the Fermi surface even if we set $(Z\tau\Delta_0)^{-1}=0$ in $(GG+FF)^0$ and $(G\bar{G}-FF)^0$. Therefore, in defining Eqs. (9), we may use the G and F functions in the absence of impurity scattering. Consequently, in Eqs. (10) the $G_\pm$, $F_\pm$ will be τ dependent while $(GG+FF)^0$ and $(G\bar{G}-FF)^0$ will not. For the τ values under consideration here this method is accurate. Only if $2Z\tau\omega_D$ becomes of the order of 1 will the integrations in Eqs. (10) extend over regions in which Eq. (7) is no longer a good approximation for $\hat{G}$. Furthermore, since we assume S-wave scattering only, there will be no τ dependence of $f_2{}^\xi$, $f_2{}^\omega$ coming from $V(p\text{-}p')$ and it follows that $f_2{}^\xi$, $f_2{}^\omega$ are identical to $f_{2,\mathrm{pure}}{}^\xi$, $f_{2,\mathrm{pure}}{}^\omega$. This allows us to calculate the mean-free-path dependence of the collective excitations for given values of $f_{2,\mathrm{pure}}{}^\xi$, $f_{2,\mathrm{pure}}{}^\omega$. For this purpose, with the help of Eqs. (10), we write the eigenvalue equation as

$$(1/f_2{}^\omega+1+I_3)(1/f_2{}^\xi+I_1)+(I_2)^2=0\,,\tag{13}$$

where the quantities $I_{1,2,3}$ are defined as

$$\begin{aligned}I_1&=-\frac{iZ^2}{N(0)}\int\frac{d^4p}{(2\pi)^4}[G_+\bar{G}_--F_+F_--(G\bar{G}-FF)^0]\\ &=\frac{1}{2}\int d\omega\left\{\frac{1+(\omega_+\omega_--\Delta_0^2)/\rho_+\rho_-}{\rho_++\rho_-+i/\tau Z}-\frac{1}{\rho}\right\},\\ I_2&=\frac{Z^2}{N(0)}\int\frac{d^4p}{(2\pi)^4}(G_+F_-+F_+\bar{G}_-)\\ &=\frac{1}{2}\int d\omega\frac{\Delta_0(\omega_+-\omega_-)/\rho_+\rho_-}{\rho_++\rho_-+i/\tau Z},\\ I_3&=-\frac{iZ^2}{N(0)}\int\frac{d^4p}{(2\pi)^4}[G_+G_-+F_+F_--(GG+FF)^0]\\ &=\frac{1}{2}\int d\omega\frac{1-(\omega_+\omega_--\Delta_0^2)/\rho_+\rho_-}{\rho_++\rho_-+i/\tau Z}.\end{aligned}\tag{14}$$

Here $\rho_\pm$ are given by $\rho_\pm=(\omega_\pm{}^2-\Delta_0^2)^{1/2}$. Numerical results for different values of $(Z2\tau\Delta_0)^{-1}$ are shown in

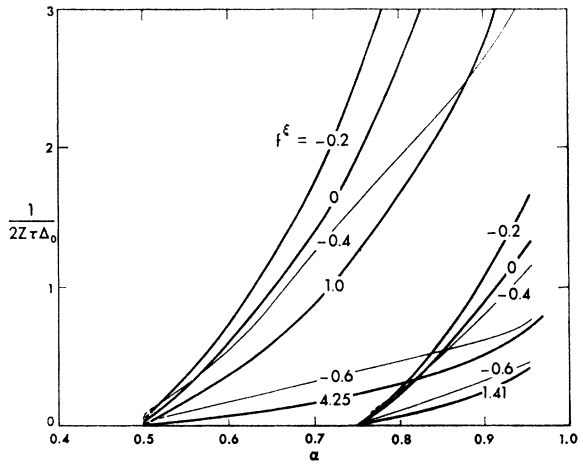

FIG. 3. Change of the energy Ω of the exciton with $\mathbf{q}=0$ as a function of mean free time τ. $\alpha=\Omega/2\Delta_0$. Z denotes the renormalization constant and different curves correspond to different values of $f_2{}^\xi$, all of which lead to an exciton energy $\alpha=0.5$ or 0.75 in the pure case. $f_2{}^\omega$ ranges from 0.23 ($f_2{}^\xi=4.25$) to -0.98 ($f_2{}^\xi=-0.6$) and from 0.22 ($f_2{}^\xi=1.41$) to -0.92 ($f_2{}^\xi=-0.6$).

Fig. 3. We have chosen sets of $f_2{}^\xi, f_2{}^\omega$ values such that in the absence of impurities the collective excitation energy Ω is $\Omega=\Delta_0$ and $1.5\Delta_0$, respectively. As a finite mean free path is introduced Ω will shift toward higher values depending on the size of f^ξ. In the weak-coupling case with $V_0=0.25$, $f^\xi=4.25$ and 1.41. As Larkin[6] has discussed, f^ξ will be comparatively small for a strong-coupling superconductor. Figure 3 shows that as f^ξ decreases, Ω becomes somewhat less sensitive to mean free path as compared to the weak-coupling case. But as the values of f^ξ decrease further, the sensitivity of Ω to mean free path increases again. In the cases under consideration, $f^\xi\simeq-0.20$ and $\simeq-0.25$ are the values of f^ξ which give least mean-free-path dependence for Ω. We want to remark that curves corresponding to $f^\xi<-0.2$ have rapidly varying slopes near the points $(\alpha,1/2Z\tau\Delta_0)=(0.5;0)$; $(0{,}75;0)$. For practical purposes an expansion in terms of $1/2Z\tau\Delta_0$ is therefore not very useful and we omit it here. Finally, if f^ξ becomes smaller than $f_{\text{crit}}{}^\xi$, which is defined as having a corresponding $f^\omega=-1$, the curves are bent in the opposite direction i.e., toward smaller α values, and become complex as $(2Z\tau\Delta_0)^{-1}$ increases. This is not surprising if one looks at Eq. (13). From the discussion in Ref. 6 it is clear that these f^ξ values are unphysical and we will not consider them any further. $f_{\text{crit}}{}^\xi$ is, for the two cases under consideration, approximately $f_{\text{crit}}{}^\xi=-0.7$ and -0.75. For $(1/2Z\tau\Delta_0)\to\infty$ all curves drawn in Fig. 3 approach $\alpha=1$ asymptotically. Figure 3 thus shows that it is improbable that collective excitations are responsible for the occurrence of a precursor found in infrared absorption experiments on lead alloys. In those experiments[3] no appreciable mean-free-path dependence of the precursor was found for τ values as low as $\tau\Delta_0\simeq1/13$ or $2Z\tau\Delta\simeq\frac{1}{3}$ if Z is assumed to be approximately equal to 2.

Since the onset of absorption in pure lead takes place roughly at $\alpha=0.5$, one sees that the theory leads to a reduction of the precursor width of at least 50% for $2Z\tau\Delta_0=\frac{1}{3}$ which has not been observed.

B. High-Impurity-Concentration Limit

If $2Z\tau\omega_D\lesssim1$ the calculations have to be carried out with the τ dependent quantities $f_2{}^\omega$ and $f_2{}^\xi$. In that case no relation between $f_2{}^\omega$, $f_2{}^\xi$ and $f_{2,\text{pure}}{}^\omega$, $f_{2,\text{pure}}{}^\xi$ can be given. Only in the weak-coupling case can we show that $f_2{}^\xi\to0$ in the high-impurity-concentration limit while $f_2{}^\omega$ is τ-independent so that $f_2{}^\omega=f_{2,\text{pure}}{}^\omega$. For high impurity concentrations one can derive by expansion a simplified equation for the eigenvalue of the form

$$(1/f_2{}^\xi-(2Z\tau\Delta_0)[F(\alpha)-E(\alpha)])(1/f_2{}^\omega+1+(2Z\tau\Delta_0)[F(\alpha)-E(\alpha)])+(2Z\tau\Delta_0)^2\alpha^2F^2(\alpha)=0. \quad (15)$$

Here $F(\alpha)$ and $E(\alpha)$ are the complete elliptic integrals of the first and second kind. The equation is of the same form as in the weak-coupling case and can be discussed in the same way as in Ref. 5.

IV. SUMMARY

By using a Fermi-liquid-theory approach for the description of strong-coupling superconductors, we have investigated the mean-free-path dependence of the excition-like collective states with total momentum $\mathbf{q}=0$. We have shown that the mean-free-path dependence is large enough so that it should have been detected in lead alloys if the precursor in the infrared absorption spectrum of those alloys were due to those collective states, as recently proposed by Larkin. The Landau type of approach is very useful in this context since it keeps calculations relatively simple and allows strong enough general statements. Despite this, it would be interesting to calculate the parameters introduced into such a theory from first principles rather than having them determined from experiments. An attempt in this direction is planned.

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