Electromagnetic Power Absorption by Collective Modes in Unconventional Superconductors

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We calculate the gauge-invariant current response of an unconventional E_1 state superconductor to electromagnetic fields in the collisionless limit using a matrix kinetic equation approach. We show that, despite broadening by nodal quasiparticles and screening currents, excitonic modes with collective frequencies of order the superconducting gap frequency should be observable as large, well-defined peaks in the power absorption. Such an observation would be a definitive signal for unconventional superconductivity. We further argue that the effect of collision broadening in real samples should be small at low temperatures.

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Interest in the heavy fermion superconductors UPt_3, UBe_{13}, $CeCu_2Si_2$, and URu_2Si_2 has focused on the possibility that these compounds might represent the first known unconventional superconductors [1]. The order parameter in such a system, in contrast to a conventional BCS superconductor, transforms according to a nontrivial representation of the symmetry group of the normal state. Among such states, enumerated in group-theoretical classifications [1–4], are some which correspond to higher-dimensional representations, i.e., multicomponent order parameters such as those realized in the superfluid phases of ^{3}He [5]. The additional internal order-parameter degrees of freedom manifest themselves through a much richer spectrum of physical properties than that of a conventional superconductor. For example, the 2D representations $E_{1g,u}$ and $E_{2g,u}$ of the hexagonal group are predicted in Ginzburg-Landau theories [6–8] including coupling to small symmetry-breaking fields to give rise to a complex phase diagram in an applied magnetic field. These theories successfully reproduce many of the experimental features observed in the UPt_3 H-T phase diagram [9]. Certain aspects of the anisotropy of critical fields in UPt_3 appear to contradict predictions of the simplest theories of this sort, however, and the true symmetry of the ground state has yet to be definitively identified.

Unconventional states corresponding to higher-dimensional representations should support collective order-parameter oscillations which, if observed, e.g., in electromagnetic surface impedance measurements, would confirm the existence of unconventional superconductivity. Furthermore, the frequency, anisotropy, and temperature variation of such collective mode features should enable one to assign a definitive symmetry class to the equilibrium state. In this paper, we calculate, within the framework of the matrix kinetic equation derived by Betbedet-Matibet and Nozières [10] and Wölfle [11], the current response of a superconductor in a $(1,i)$ E_{1g} state to an electromagnetic field. On a phenomenological level, we analyze the role of impurities in damping collective oscillations. Our results indicate that a relatively sharp collective feature at $\Omega_{\rm coll} \simeq 1.2\Delta_0$ in a low-temperature far-infrared power absorption experiment should be observable in clean samples of UPt_3 (here Δ_0 is the gap maximum over the Fermi surface). We argue that such features should be observable and generic to all unconventional states, but also exhibit characteristic anisotropy and doping dependence.

Order-parameter collective modes in superconductors [12] are macroscopic quantum oscillations of the pair condensate which may be excited by external probes such as sound or electromagnetic waves. Although intensively studied in the late 1950s and early 1960s because of their intimate connection with gauge-invariance problems of the BCS theory, they are largely irrelevant for experiments on conventional superconductors [13]. This is because the Goldstone mode [14, 15] corresponding to the spontaneously broken U(1) gauge symmetry in a superconductor oscillates at the plasma frequency because of its coupling to the charge density. Furthermore, "excitonic" (optical) modes corresponding to excited bound states of Cooper pairs couple weakly and have collective frequencies $\Omega \gtrsim 2\Delta$ and are thus strongly damped by pair breaking.

More recently, in the context of the discussion of possible unconventional superconductivity in heavy fermion materials, several authors [16–20] recognized that excitonic modes similar to those seen in superfluid ^{3}He might be observable in unconventional superconductors as well. In both cases, excitonic modes below the pair-breaking edge are possible because the order parameter Δ_k transforms according to a higher-dimensional representation of the normal state, giving rise to a spectrum of pairing states. In the case of ^{3}He, observation of such excitonic modes in ultrasound measurements was instrumental in confirming the symmetry of the A- and B-phase order parameters. Although such modes must exist if the order parameter transforms according to a multidimensional representation, the question remains how best to observe them in an experiment. Early on it was realized that since the transition temperatures of heavy fermion superconductors are much higher than that of ^{3}He, electromagnetic absorption was a more promising probe than sound attenuation [18]. Early calculations [19] indicated that in the pseudoisotropic Balian-Werthamer model state, such modes should be observable as large sharp features in the power absorption or surface resistance below the gap edge in the clean, type-II limit. The mode was found to have a finite width even at T=0 caused by its coupling to screening currents $j(\vec{q}, \Omega_{\rm coll}(\vec{q}))$ at all wave vectors $0 \leq q \leq 1/\xi_0$. The question then arises whether such sharp absorption features can be observed in anisotropic states thought to be realized in heavy fermion superconductors, where additional broadening will be caused by excited quasiparticles in the neighborhood of the points or lines where the order parameter vanishes.

For definiteness, we study the 2D E_{1g} representation of the hexagonal group, with basis functions $\hat{k}_z\hat{k}_x, \hat{k}_z\hat{k}_y$ on the Fermi sphere, where $\hat{z}$ coincides with the hexagonal crystal axis $\hat{c}$. In fourth-order Ginzburg-Landau theories of the UPt_3 phase diagram, the $(1,i)$ state with symmetry $\hat{k}_z(\hat{k}_x + i\hat{k}_y)$ is the stable low-temperature phase if the quartic coefficient $\beta_2 s > 0$. This state is a popular candidate for the ground state of UPt_3 since it displays a line of nodes in the basal plane, consistent with anisotropic sound attenuation measurements [21, 22] and other transport properties [23, 24]. We then write the spin singlet equilibrium order parameter as $\Delta_k = \Delta_0 Y_{21}(\hat{k})$, where $Y_{2\pm1}(\hat{k}) \equiv 2\hat{k}_z(\hat{k}_x \pm i\hat{k}_y)$ is normalized such that Δ_0 is the gap maximum over the Fermi sphere. A time-dependent perturbationlike sound or an electromagnetic wave will, to leading order, excite all order-parameter components degenerate with $Y_{21}(\hat{k})$. In the case of a pair interaction transforming according to the $\ell = 2$ representation of the rotation group, one recovers the ^{3}He-like problem involving all five $Y_{2m}(\hat{k})$ analyzed by Hirashima and Namaizawa [25]. In a hexagonal crystal, however, rotational symmetry is lowered to D_{6h} symmetry, and only $Y_{2\pm1}$ are degenerate. We therefore write the order-parameter fluctuation $\delta\Delta_k = \sum_{m=\pm1} \delta\Delta_m Y_{2m}(\hat{k})$; by doing so, we exclude all possible spin-orbit Goldstone modes such as those found in Ref. [25].

We now calculate the order-parameter fluctuation and current response using the matrix kinetic equation [5]. We consider an electromagnetic wave characterized by potentials $\vec{A}, \phi$ normally incident on a type-II superconducting surface. Quasiparticles of momentum $\hat{k}$ on the Fermi surface then experience an energy shift of $\delta\epsilon_k(\vec{q}, \Omega) = e\phi + (ev_F/c)\hat{k} \cdot \vec{A} + v_q \delta n_0$, where δn_0 is the density fluctuation. We have neglected Fermi-liquid corrections, but included the average shift $v_q \delta n_0$ due to the long-range Coulomb interaction $v_q = 4\pi e^2/q^2$, which is necessary for the proper treatment of long-wavelength density fluctuations. The full solution of the kinetic equation requires the inversion of a 4×4 matrix of collective response functions and the construction of the scalar distribution function δn_k in terms of the self-consistently determined mean fields $\delta\Delta_k$ and $\delta\epsilon_k$. We reserve the details of the method of solution and its complete form for a longer work, and focus instead on the special case of maximum coupling to the collective mode, as well as qualitative features of the general case. We neglect particle-hole asymmetric terms in the kinetic equations, thereby neglecting some additional collective modes which couple weakly to electromagnetic fields. We note, however, that these terms have recently been claimed to be important for observations of circular dichroism in time-reversal-breaking unconventional states [20].

The current response is now given by the usual transport expression, $\vec{j} = -ev_F N_0 \langle \hat{k} \delta n_{\hat{k}} \rangle_{\hat{k}} - (ne^2/mc)\vec{A}$, where N_0 is the density of states at the Fermi surface, and $\langle \cdots \rangle_{\hat{k}}$ represents a Fermi surface average. If one performs a gauge transformation $\phi \to 0, \vec{A} \to \vec{A}' = c\vec{E}/i\Omega$, where $\vec{E}$ is the electric field, the transverse current response may be written in gauge-invariant form in the limit $\Omega << \Omega_{\rm pl} \equiv \sqrt{4\pi ne^2/m}$ as

$$\vec{j}_T = \left\{ \overleftrightarrow{K} - \frac{(\overleftrightarrow{K} \cdot \hat{q})(\hat{q} \cdot \overleftrightarrow{K})}{\hat{q} \cdot \overleftrightarrow{K} \cdot \hat{q}} \right\} \cdot \vec{A}' \equiv \overleftrightarrow{K}_{\rm tot} \cdot \vec{A}' \,, \qquad (1)$$

where $\overleftrightarrow{K}$ is the current response, to a vector potential, of the quasiparticle gas *in the absence of the long-range Coulomb interaction* [26]. The symmetries of this tensor have recently been discussed by Yip and Sauls [20], who calculated the current response for an axial-type p-wave state using methods similar to ours. If we take the electric field $\vec{E}$ at the surface to be polarized along $\hat{\epsilon} \perp \hat{q}$ and the gap axis $\hat{\ell} = \hat{z} = \hat{\epsilon} \times \hat{q}$, the coupling to the mode may be shown to be maximal. Furthermore, the kinetic equation decouples into two 2×2 blocks, and its solution simplifies considerably. For this configuration, the response $\overleftrightarrow{K}$ may be written $\overleftrightarrow{K}_{\rm ex} + \overleftrightarrow{K}_{\rm coll}$, with single-particle excitation response

$$\overleftrightarrow{K}_{\rm ex} \equiv (-3ne^2/mc)\langle \hat{k}\hat{k}(\Omega^2 - \eta^2\lambda_k)/(\Omega^2 - \eta^2) \rangle_{\hat{k}}$$

describing particle-hole excitations of energy $\eta = \vec{q} \cdot \nabla_k \epsilon_k$. The collective response $\overleftrightarrow{K}_{\rm coll}$ describes resonant excitations of the order parameter at finite frequency Ω into a state involving an admixture of a $Y_{2,-1}$ component into the ground state. This is equivalent to an oscillation of the preferred directions $\hat{n}$ and $\hat{m}$ of the nonequilibrium order parameter $\Delta_k = 2\Delta_0 \hat{k}_z(\hat{n} + i\hat{m}) \cdot \hat{k}$ about their equilibrium positions $\hat{n} = \hat{x}$ and $\hat{m} = \hat{y}$ (a corresponding mode in superfluid ^{3}He-A has been termed the "clapping mode"). We find

$$\overleftrightarrow{K}_{\text{coll}} = \left(\frac{-3ne^2}{2mc}\right)(v_F q)^2 \left(\frac{\alpha}{D^+}\right) \times [(\overleftrightarrow{\lambda}_1 \cdot \hat{q})(\hat{q} \cdot \overleftrightarrow{\lambda}_{-1}) + (\overleftrightarrow{\lambda}_{-1} \cdot \hat{q})(\hat{q} \cdot \overleftrightarrow{\lambda}_1)], \quad (2)$$

where $\alpha \equiv \Omega^2\lambda - 4\lambda_2\Delta_0^2 - (v_F q)^2 \hat{q} \cdot \overleftrightarrow{\lambda} \cdot \hat{q}$, and the resonance denominator D^+ is given by

$$D^+ = \alpha[\alpha + 2\Delta_0^2(\lambda_2 - \lambda_{4,1})] - [(v_F q)^2 \hat{q} \cdot \overleftrightarrow{\lambda}_1 \cdot \hat{q} - \lambda_1\Omega^2 + 4\Delta_0^2\lambda_{2,1}]^2. \quad (3)$$

Here we have defined various moments of the dynamical pair response function $\lambda_k(\vec{q}, \Omega)$ first derived by Tsuneto [27], as given, e.g., in Ref. [19]. These include $\lambda = \langle\lambda_k\rangle_{\hat{k}}$, $\overleftrightarrow{\lambda} = \langle\hat{k}\hat{k}\lambda_k\rangle_{\hat{k}}$, $\lambda_1 = \langle\lambda_k Y_{21}^2/|Y_{21}|^2\rangle_{\hat{k}}$, $\lambda_2 = \langle\lambda_k|Y_{21}|^2\rangle_{\hat{k}}$, $\overleftrightarrow{\lambda}_m = \langle\hat{k}\hat{k}\lambda_k Y_{2m}^2/|Y_{21}|^2\rangle_{\hat{k}}$, $\lambda_{2,1} = \langle\lambda_k Y_{21}^2\rangle_{\hat{k}}$, and $\lambda_{4,1} = \langle\lambda_k Y_{21}^4/|Y_{21}|^2\rangle_{\hat{k}}$. An intuitive feeling for these quantities may be gained by examining the static homogeneous limit at $T = 0$. In this case $\lambda_k \to 1$, so that $\overleftrightarrow{\lambda} \to \overleftrightarrow{1}/3$, $\lambda_2 \to 8/15$, and $\lambda \to 1$. Furthermore, it is easy to check that λ_1, $\lambda_{2,1}$, and $\lambda_{4,1}$ are of order $(v_F q/\Delta_0)^2$, so that the position of the collective pole in the response, corresponding to $\text{Re}D_+ = 0$, is given roughly at $q \to 0$ by $\Omega^2 = 2\Delta_0^2 \text{Re}[\lambda_2(\Omega)/\lambda(\Omega)] \simeq 1.42\Delta_0^2$. The full numerically determined dispersion of the mode is shown in Fig. 1.

We now solve the boundary value problem for an electromagnetic wave normally incident on a half space, leading to an expression for the power absorption at fixed frequency:

$$P(\Omega) = \frac{-\Omega|B_0(\Omega)|^2}{\pi c}\hat{\epsilon}\cdot\text{Im}\int_0^\infty dq\left\{q^2 - \frac{4\pi}{c}\overleftrightarrow{K}_{\text{tot}}\right\}^{-1}\cdot\hat{\epsilon}, \quad (4)$$

where B_0 is the magnetic field amplitude at the surface. For the highly symmetric configuration considered, we note that the coupling to the density δn_0 vanishes, so that $\overleftrightarrow{K}_{\text{tot}} = \overleftrightarrow{K}$ in (1). We have evaluated (4) numerically, with results illustrated in Fig. 2 for various values of the Ginzburg-Landau parameter $\kappa \equiv \Lambda_0/\xi_0$, where Λ_0 and ξ_0 are the T=0 penetration depth and coherence length, respectively. For highly nonlocal electrodynamics $\kappa \ll 1$ the single-particle excitations dominate the power spectrum, while the collective mode dominates in the local (London) limit appropriate for the heavy fermion superconductors. In fact, the weight in the collective mode is found to scale with κ. This follows from (4) after expanding the denominator in terms of the small quantity $\overleftrightarrow{K}_{\text{coll}}$, taking account of the lower cutoff Λ_0^{-1} in the q-integral provided by $\overleftrightarrow{K}_{\text{ex}}$. We conclude that, while the peak is broader than in the pseudoisotropic case, in the clean limit broadening due to excited nodal quasiparticles should not prevent the observation of the mode.

Since relaxation times at T_c in good samples of UPt$_3$ are of order $\tau_N \gtrsim 1/T_c$, the clean or anomalous skin effect limit discussed above is clearly not appropriate. One might at first glance conclude that the chances of observing such a mode at frequencies $\Omega_{\text{coll}} \sim \Delta_0 \sim T_c$ are therefore marginal. As pointed out by Wölfle [18], however, at temperatures $T \ll T_c$ the actual damping of the collective oscillations can be considerably smaller. When collisions are properly accounted for, the order parameter obeys an effective damped harmonic oscillator equation

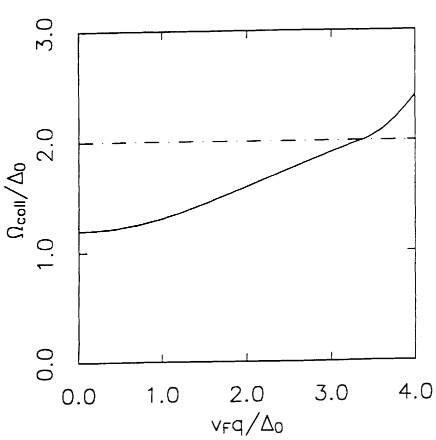

FIG. 1. Dispersion relation $\Omega_{\text{coll}}(q)$ vs $v_f q$ for E_{1g} "clapping" mode. Dot-dashed line represents "pair-breaking edge," i.e., $\Omega > 2\Delta_k\ \forall\hat{k}$.

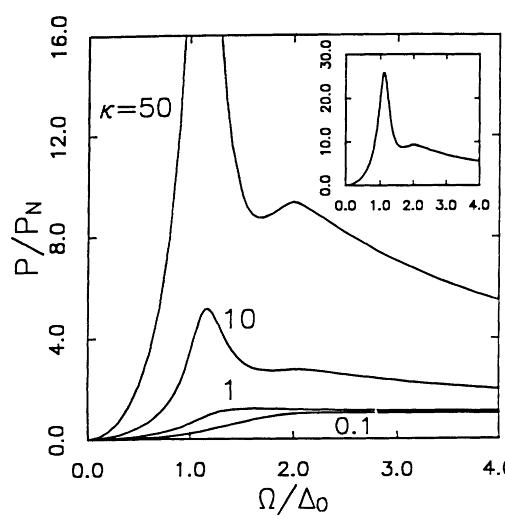

FIG. 2. Normalized power absorption P/P_N in E_{1g} state for various $\kappa \equiv \Lambda_0/\xi_0$. P_N is power absorption in anomalous skin effect regime.

for $q \to 0$:

$$\left[\Omega^2 + i\Omega\frac{\chi_0}{\tau} - \Omega_{\rm coll}^2\left(1 - \frac{\chi_1}{1 - i\Omega\tau}\right)\right]\delta\Delta_j = 0, \qquad (5)$$

where $\delta\Delta_j$ are any of the order-parameter components with the symmetry of the exciton, and χ_0 and χ_1 are combinations of moments of the Tsuneto function $\lambda_k(\Omega)$ and an analogous function of $\Omega + i/\tau$. The term χ_0/τ represents a relaxation of the order parameter due to quasiparticle collisions, while χ_1 represents a quasiparticle correction to the oscillator potential itself. Thus both χ_0 and χ_1 are found to vanish as $T \to 0$ roughly as the normal-fluid density ρ_n [we neglect gapless effects leading to $\rho_n(T = 0) > 0$]. For a (1,i) E_{1g} state and a "clapping"-type mode [5] of the type considered here, we estimate $\chi_0 \sim (1/\Delta_0)(T\tau)^2 \sim T^4$ and $\chi_1 \sim \chi_0$ for $T \ll T_c$. Thus the mode should always be well defined even for $\Omega\tau_N \simeq 1$ at sufficiently low temperatures.

We note that an odd-parity state with symmetry $\hat{d}(\hat{k}_x + i\hat{k}_y)$ belonging to the E_{1u} representation will give rise to a current response of identical structure if the spin axis $\hat{d}$ is taken to be homogeneous and time independent. While the damping coefficients χ_0 and χ_1 will be quite different, the position and anisotropy of the mode feature in the power absorption in the clean limit will differ from the (1,i) E_{1g} state considered here only through qualitatively insignificant angular averages. However, other possible states will be distinguishable through quite different anisotropies, e.g., dependence on electric field polarization.

We have shown that excitonic collective modes, characteristic of all multidimensional unconventional order parameters, should be observable in electromagnetic absorption experiments. Our best estimate of the maximum linear frequency at which such a mode might be detected is $\nu_{\rm max} \simeq 26$ GHz. If the mode is well defined at intermediate temperatures, the temperature variation of the gap should allow for its detection at fixed $\nu < \nu_{\rm max}$ as temperature is swept, but for frequencies too small compared to $\nu_{\rm max}$ the absorption peak temperature will lie too close to T_c, most probably smearing out the predicted feature beyond recognition. We are not aware of any published experiments which have been performed on UPt_3 involving frequencies in the required range. The measurement we propose requires cavities with resonant frequencies in the far-infrared operated at low temperatures, a difficult but not impracticable experiment. Observation of such a collective feature would provide definitive evidence of unconventional superconductivity and aid in the identification of the pair state of such systems.

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