Theory for the effects of impurities on the Raman spectra of superconductors. II. Temperature dependence and influence of final-state interactions

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The influence of final-state interactions on the electronic Raman scattering by pairs of quasiparticles in disordered superconductors is presented using the formalism developed in a previous paper [Phys. Rev. B **45**, 12 965 (1992)]. Below the gap, the effect of impurities on both the position and strength of the bound state is calculated, while additional modifications of the Raman spectrum above the gap due to final-state interactions are presented. Previous results for the position of the bound state by Maki and Tsuneto are obtained, while it is additionally shown that the strength of the collective mode diminishes rapidly with increasing disorder. While the Raman spectrum above the gap sharpens with the inclusion of final-state interactions for small disorder, for larger disorder the effects of final-state interactions become completely negligible. The temperature dependence of the theory is investigated and it is shown that a non-BCS temperature dependence of the peak in the Raman spectrum can be obtained for strong impurity scattering.

I. INTRODUCTION

It has been known for some time that when a Cooper pair is excited to create two single-particle excitations, residual interactions (final-state interactions, FSI) which do not contribute to Cooper pair formation mediate the binding of the two single-particle excitations to create a bound pair which is orthogonal to the Cooper pairs. The bound state would then appear at an energy below the gap. The theoretical work concerning the effects of final-state interactions (i.e., the residual phonon mediated electron-electron interactions in channels with nonzero angular momenta) in both clean[1,2] and dirty[3,4] superconductors has been well studied. It has been shown[3] that in the presence of impurities the energy of the bound state decreases but is undamped. Since there has been no experimental indication of a precursor excitation with a strong disorder dependence it is inferred that the energy of the bound state must be very small and thus the bound state must lie very close to the gap edge.[5] However, a bound state always exists for finite FSI. It was shown that for clean superconductors FSI alter the spectral weight distribution near the gap edge and thus can provide a mechanism to obtain symmetry dependent Raman spectra that make it difficult to distinguish between the effects of gap anisotropy and FSI.[2] It is the purpose of the present paper to discuss the role FSI have on the Raman spectrum of disordered superconductors.

Recently, a gauge-invariant description of the Raman spectrum for disordered superconductors has been given.[6] Special attention was given to the role of anisotropic mass fluctuations arising from a nonparabolic conduction band dispersion. It was shown that impurities dramatically affect the Raman spectrum, and symmetry dependent Raman spectra can be obtained for anisotropic impurity scattering. Further, the position of the peak of the Raman spectrum does not coincide with the gap edge for large impurity scattering, $2\Delta\tilde{\tau}_L < 1$, where $\tilde{\tau}_L$ is the mean free elastic lifetime in crystal harmonic channel L reduced by vertex corrections, i.e., $1/\tilde{\tau}_L = 1/\tau_{L=0} - 1/\tau_L$. The channel L is the symmetry channel corresponding to the point group of the crystal selected by the orientations of the incoming and outgoing polarization light vectors. Although Ref. 6 formally included FSI, for purposes of discussion the main emphasis concerned the role of impurities on the "pair approximation"[7] for the Raman response at zero temperature, i.e., FSI were neglected. The present paper completes the work of Ref. 6, and relies on the exact eigenstate formalism developed for correlation functions in arbitrarily disordered superconductors as presented in Refs. 6 and 8. Therefore, the main technical details of the calculations can be found in these references and thus we will be brief.

The plan of the paper is as follows: Section II concerns the disorder dependence of the Raman spectrum including final-state interactions with special detail given to the shape of the spectrum near 2Δ, while Sec. III concerns the temperature dependence of the spectrum. In particular it will be shown that a non-BCS temperature dependence for the peak of the Raman spectra can be obtained for large impurity scattering.

II. ROLE OF FINAL-STATE INTERACTIONS IN DISORDERED SUPERCONDUCTORS

A. $\omega < 2\Delta$: Bound states

The differential Raman cross section in channel L can be written as

$$\frac{d^2R_L}{d\omega\, d\Omega} = -\frac{r_0^2}{\pi}[1+n(\omega)]\tilde{\chi}_L''(\mathbf{q},\omega), \tag{1}$$

where $r_0 = \frac{e^2}{mc^2}$ is the Thompson radius, $\mathbf{q}$ and ω denote

the momentum and energy transferred from the light to the superconductor, and $n(\omega)$ is the Bose distribution. In this paper we will consider only small momentum transfers, $q\xi \ll 1$ where ξ is the dirty-limit coherence length. The response function $\tilde{\chi}$

$$\tilde{\chi}(\mathbf{q},\omega) = \langle[\tilde{\rho}(\mathbf{q}),\tilde{\rho}(-\mathbf{q})]\rangle_{(\omega)} \tag{2}$$

is formed with an effective density operator

$$\tilde{\rho}(\mathbf{q}) = \sum_{\mathbf{k},\sigma} \gamma(\mathbf{k}) c_\sigma^\dagger(\mathbf{k}+\mathbf{q}) c_\sigma(\mathbf{k}) \tag{3}$$

with electron creation, annihilation operators $c^\dagger, c$. Here $\gamma(\mathbf{k})$ are the Raman tensor elements selected by the incident and scattered polarization vectors $\mathbf{e}^i, \mathbf{e}^s$:

$$\gamma(\mathbf{k}) = \sum_{\alpha,\beta} e_\alpha^s \gamma_{\alpha,\beta}(\mathbf{k}) e_\beta^i. \tag{4}$$

Besides the field terms, the Hamiltonian is written as

$$\begin{aligned} H = H_{e\text{-}m} &+ \sum_{\mathbf{k},\sigma} \epsilon(\mathbf{k}) c_\sigma^\dagger(\mathbf{k}) c_\sigma(\mathbf{k}) \\ &+ \sum_{\mathbf{k},\mathbf{k}',\sigma} V_{\mathbf{k},\mathbf{k}'}^{\mathrm{imp}} c_\sigma^\dagger(\mathbf{k}) c_\sigma(\mathbf{k}') \\ &+ \sum_{\mathbf{k},\mathbf{k}'} V_{\mathbf{k}',\mathbf{k}} c_\uparrow^\dagger(\mathbf{k}') c_\downarrow^\dagger(-\mathbf{k}') c_\downarrow(-\mathbf{k}) c_\uparrow(\mathbf{k}). \end{aligned} \tag{5}$$

Here V^{imp} and V are the impurity and effective phonon-mediated electron-electron interactions, respectively, and the interaction only acts in the Cooper channel. Additional terms of the Hamiltonian correspond to electron-electron interaction in the electron-hole (zero-sound) channel, which have been considered for clean superconductors in Ref. 2, and Coulomb interactions. Since Coulomb forces only couple to isotropic charge fluctuations, we only need to consider them in the $L = 0$ channel. We shall for the time being neglect the zero-sound channel as well, and return to it at the end of the subsection.

The response function $\tilde{\chi}_L$ is given by integral equations depicted in Figs. 1 and 2 of Ref. 8. The integral equations are solved by first expanding the vertices and interactions in crystal harmonics, i.e.,

$$V_{\mathbf{k}',\mathbf{k}} = \sum_{L,L'} V_{L,L'} \phi_L^*(\mathbf{k}) \phi_{L'}(\mathbf{k}'),$$

and then expanding the renormalized vertex in quaternions. It was found in Ref. 6 that $\tilde{\chi}_L$ can be given in terms of three response functions of a superconductor,

$$\tilde{\chi}_L(\mathbf{q},i\omega) = 2C_L(\mathbf{q},i\omega)/[1 + V_L C_L(\mathbf{q},i\omega)], \tag{6}$$

$$\begin{aligned} C_L(\mathbf{q},i\omega) = B_{+,L}(\mathbf{q},i\omega) \\ + V_L A_L^2(\mathbf{q},i\omega)/[1 - V_L B_{-,L}(\mathbf{q},i\omega)]. \end{aligned} \tag{7}$$

$B_{+,L}$ is the "pair approximation" response, i.e., neglecting FSI, while A_L and $B_{-,L}$ represent collective effects. The position of the collective mode is given by the zeros of the denominator in Eq. (6). For energies below the gap, this always has a solution for nonzero V_L. In the $L = 0$ channel, the collective mode is the Anderson-Bogoliubov sound mode,[9] which is soft and lies in the gap for neutral superconductors but is lifted to plasmon energies by long-range Coulomb interactions. The influence of disorder on the position and residue of the collective mode in the $L = 0$ channel has been investigated in Ref. 8. It was found that the only difference from clean superconductors is that the speed of the sound mode is simply renormalized by the disorder. For large q, the mode in neutral superconductors moves closer to the gap edge and decreases in strength.

In the case of $L \neq 0$ channels, long-range Coulomb forces do not couple to the anisotropic mass fluctuations and thus the mode remains in the gap. Further, since the quasiparticles are not screened (and thus the Raman spectrum does not vanish for zero momentum), in the limit of small momentum transfers the role of the wave number q as the particle-hole width is replaced by the inverse of the impurity lifetime $1/\tilde{\tau}_L$, which does not drop out due to the nonconservation of anisotropic mass tensor in channels $L \neq 0$. Consequently, Maki and Tsuneto[3] have shown that the mode position strongly depends on disorder, moving closer to the gap edge for increasing disorder. Since no precursor excitation has been experimentally seen to have such a strong disorder dependence, it was inferred that the collective mode must have a very small binding energy, with broadening making it indistinguishable from the gap edge.[5] In this subsection, we check these results and also calculate the residue of the pole.

To obtain information on the collective mode, we need the response functions A_L and $B_{\pm,L}$. The functions A_L and $B_{\pm,L}$ are given in terms of integrals in Eqs. (18)–(20) in Ref. 6. The imaginary parts can be obtained analytically for all frequencies and are expressed in terms of complete elliptical integrals in Eqs. (28)–(30) in Ref. 6. The corresponding real parts for frequencies below 2Δ can only be obtained analytically in the limits of small and large disorder, and for frequencies $\omega = 0$ and $\omega \simeq 2\Delta$. For arbitrary frequencies the real parts of the response functions must be first obtained numerically and then the position of the divergence of Eq. (6) must be found. However, we are only interested in the physically interesting case of collective modes lying near the gap edge, and thus we first present analytic results for this energy region and then present numerical results for other regions later in this section. Analytic expressions for small (large) disorder, $1/2\Delta\tilde{\tau}_L \ll (\gg) 1$, and for $\omega = 0$ can be found in the Appendix.

Asymptotically close to the gap edge such that $\sqrt{1-(\omega/2\Delta)^2} \ll \frac{1}{2\Delta\tilde{\tau}_L}$, the response functions can be written as

$$\begin{aligned} &\frac{B_{-,L}(\mathbf{q},\omega)}{\gamma_L^2} \\ &\quad = 1/V_{L=0} - Y(2\Delta\tilde{\tau}_L) - N_F 2\Delta\tilde{\tau}_L[E(\alpha) - F(\alpha)], \end{aligned} \tag{8}$$

$$\frac{B_{+,L}(\mathbf{q},\omega)}{\gamma_L^2} = N_F - N_F 2\Delta\tilde{\tau}_L[E(\alpha) - F(\alpha)], \tag{9}$$

$$\frac{A_L(\mathbf{q},\omega)}{\gamma_L^2} = 2\Delta\tilde{\tau}_L \alpha N_F F(\alpha), \tag{10}$$

with

$$Y(x) = \gamma_L^2 N_F \frac{1}{\sqrt{1-x^2}} \text{arccosh}(1/x) \quad \text{for } x < 1,$$
$$Y(x) = \gamma_L^2 N_F \frac{1}{\sqrt{x^2-1}} [\pi/2 - \arcsin(1/x)] \quad \text{for } x > 1. \tag{11}$$

Here $\alpha = \omega/2\Delta, \gamma_L$ is the Lth component of the Raman vertex expanded in either crystal or spherical harmonics, F and E are complete elliptical integrals of the first and second kinds, respectively, and N_F denotes the density of states per spin at the Fermi level. For $\omega < 2\Delta$ the response functions have no imaginary part. Similar expressions for $B_{\pm,L}$ have been obtained previously,[3] and a brief derivation of these results is given in the Appendix.[10] The real parts can also be numerically obtained from the imaginary parts via a Kramers-Kronig transform. This procedure is spelled out in Ref. 8.

Using Eqs. (8)–(11) we now give analytic results for the effect of disorder on the position and residue of the collective mode near 2Δ. We write the collective mode contribution to the Raman spectrum as $\tilde{\chi}_L''(\mathbf{q},\omega < 2\Delta) = Z_L N_F \gamma_L^2 \delta(\omega - \omega_0)$, and define the dimensionless FSI parameter as $g_L = V_L N_F$. Expanding Eqs. (8)–(10) for frequencies near 2Δ we find that for $g_0/g_L \gg 1$ the position of the collective mode is

$$\omega_0 = 2\Delta\sqrt{1 - 16e^{-2\rho_L}}, \tag{12}$$

while the residue obtained by linearizing the denominator in Eq. (6) around ω_0 is

$$Z_L = \frac{32\pi\rho_L e^{-2\rho_L}}{\sqrt{1-16e^{-2\rho_L}}}. \tag{13}$$

Here

$$\rho_L = 1 + \frac{g_0^*}{2\Delta\tilde{\tau}_L} \frac{1}{1 + g_0^*(1 - 4\Delta\tilde{\tau}_L)} \times \left[(1/g_L - 1/g_0^*)(1 + 1/g_L) - (2\Delta\tilde{\tau}_L)^2\right], \tag{14}$$

with

$$1/g_0^* = 1/g_{L=0} - Y(2\Delta\tilde{\tau}_L). \tag{15}$$

Since g_0^* and subsequently ρ_L increases with disorder, we see that the effect of disorder is to exponentially push the bound state closer to the gap and to decrease its strength. This is similar to the behavior of the bound state in the clean superconducting density response for large wave vectors q.[7] Equations (12) and (13) predict that the bound state position is never more than 1% less than the gap edge and that the strength of the bound state quickly decreases with disorder from 10^{-3} for small disorder to below 10^{-6} for large disorder. Thus disorder acts adversely to the collective mode, in agreement with Maki and Tsuneto.[3]

In order to obtain the disorder dependence of collective modes lying further from the gap edge, we numerically evaluate the real parts of the response functions via a Kramers-Kronig transform from the analytically obtained imaginary parts. The results obtained for the position and residue of the mode for different values of $V_L/V_{L=0}$ as a function of disorder are displayed in Table I. Here we have chosen $N_F V_{L=0} = 0.2$. We see that the collective mode is strongly affected by disorder and moves closer to the gap edge for increasing disorder. This effect has been previously presented in Refs. 3 and 4. Additionally, we see that the pole strength is greatly reduced with increasing disorder. Except for strong FSI ($V_L/V_{L=0} > 0.9$) the bound state has very little effect on the spectrum below 2Δ even for only moderately large impurity rates, i.e., $1/2\Delta\tilde{\tau}_L > 0.5$, and its influence diminishes rapidly for larger disorder. We estimate our numerical error for the position of the bound state to be better than 0.1%, while the error for the residue to be better than 1.0%.[11]

B. $\omega > 2\Delta$: Continuum

We now present analytic results for the response for frequencies just greater than the gap edge. We can already obtain the behavior of the response at the gap edge from Eqs. (28)–(30) of Ref. 6. Since the spectral functions all are constant at the gap edge, the corresponding real parts must diverge logarithmically. We now demonstrate this behavior analytically.

It is shown in the Appendix that asymptotically close to the gap edge such that $\sqrt{(\omega/2\Delta)^2 - 1} \ll \frac{1}{2\Delta\tilde{\tau}_L}$, the real parts of A and $B_\pm$ can be written as

TABLE I. Position ω_0 and residue Z_L of the pole in $\tilde{\chi}_L''(\mathbf{q},\omega)$. The asterisk represents less than 0.01% less than gap edge.

$\frac{1}{2\Delta\tilde{\tau}_L}$	$V_L/V_{L=0} = 0.4$		$V_L/V_{L=0} = 0.5$		$V_L/V_{L=0} = 0.6$		$V_L/V_{L=0} = 0.7$		$V_L/V_{L=0} = 0.8$		$V_L/V_{L=0} = 0.9$	
	$\omega_0/2\Delta$	Z_L	$\omega_0/2\Delta$	Z_L	$\omega_0 2/\Delta$	Z_L	$\omega_0/2\Delta$	Z_L	$\omega_0/2\Delta$	Z_L	$\omega_0/2\Delta$	Z_L
0.0	0.998	0.000125	0.990	0.000338	0.974	0.00153	0.937	0.00611	0.857	0.0191	0.684	0.0605
0.1	0.999	$<10^{-4}$	0.996	0.000191	0.987	0.000967	0.961	0.00375	0.900	0.0137	0.765	0.0458
0.2	1.0*	$<10^{-4}$	0.999	$<10^{-4}$	0.995	0.000421	0.979	0.00245	0.935	0.0100	0.830	0.0350
0.3	1.0*	$<10^{-4}$	1.0*	$<10^{-4}$	0.998	0.000248	0.989	0.00152	0.959	0.00713	0.879	0.0268
0.4	1.0*	$<10^{-4}$	1.0*	$<10^{-4}$	0.999	0.000132	0.995	0.000762	0.975	0.00486	0.915	0.0205
0.5	1.0*	$<10^{-4}$	1.0*	$<10^{-4}$	1.0*	$<10^{-4}$	0.998	0.000421	0.986	0.00277	0.943	0.0150

$$\frac{B'_{-,L}(\mathbf{q},\omega)}{\gamma_L^2} = 1/V_{L=0} - Y(2\Delta\tilde{\tau}_L) - N_F 2\Delta\tilde{\tau}_L(\omega/2\Delta)[E(2\Delta/\omega) - (2\Delta/\omega)^2 F(2\Delta/\omega)], \tag{16}$$

$$\frac{B'_{+,L}(\mathbf{q},\omega)}{\gamma_L^2} = N_F - N_F 2\Delta\tilde{\tau}_L(\omega/2\Delta)[E(2\Delta/\omega) - (2\Delta/\omega)^2 F(2\Delta/\omega)], \tag{17}$$

$$\frac{A'_L(\mathbf{q},\omega)}{\gamma_L^2} = 2\Delta\tilde{\tau}_L N_F F(2\Delta/\omega). \tag{18}$$

While in general the resulting expression for $\tilde{\chi}$ is quite complicated, we can simplify the expression by only focusing on the strong disorder limit, in which Eqs. (16)–(18) are good approximations. Also, as seen in Ref. 6, the spectral functions in the vicinity of the gap edge become flat for large disorder and thus it is also an acceptable approximation to replace the spectral functions in Eq. (6) by the value at the threshold:

$$A''_L(2\Delta) = B''_{-,L}(2\Delta) = B''_{+,L}(2\Delta) = \gamma_L^2 N_F \pi \Delta\tilde{\tau}_L.$$

These approximations lead to the following expression for the response for frequencies just above the gap edge for large disorder $2\Delta\tilde{\tau}_L \ll 1$:

$$\tilde{\chi}''_L(\mathbf{q},\omega\sim 2\Delta) = 2\gamma_L^2 N_F \pi \Delta\tilde{\tau}_L (1 - g_L/g_0^*)^2 \left(\frac{1 - g_L/g_0^* - g_L B'}{g_L^3 B'^2/g_0^* + (1 - g_L/g_0^*)^2(1 - g_L B')}\right)^2, \tag{19}$$

where $B' = 2\Delta\tilde{\tau}_L F(2\Delta/\omega)$ diverges logarithmically for $\omega \to 2\Delta$. For $g_L \to 0$ the "pair approximation" $B''_{+,L}$ is recovered. The effect of the renormalization is that the spectrum now turns on continuously at the gap edge and then rises as $1/\ln^2(\omega - 2\Delta)$, in agreement with the behavior for the clean case[12] and both the clean and dirty density responses.[7,8] The peak of the spectrum is shifted away from the gap edge for increasing g_L, while for increasing disorder the spectral weight in the frequency region near the gap decreases. Further, the region of the $1/\ln^2$ rise becomes smaller for larger amounts of disorder. For $2\Delta\tilde{\tau}_L \sim \frac{1}{10}$ the turn on of the renormalized spectrum looks very much like a jump rather than a continuous rise. At higher frequencies, Eq. (19) shows little differences from $B''_{+,L}$, Eq. (28) in Ref. 6 for strong disorder. Thus the differences between the renormalized and the unrenormalized spectrum is confined to a narrow range of frequencies close to the gap edge which shrinks for larger disorder.

Using the analytic expressions for the imaginary parts of the response functions and the numerically determined real parts we show the effects of FSI on the Raman response for a larger frequency region above 2Δ in Fig. 1 and for general values of $2\Delta\tilde{\tau}_L$. While FSI effects below the gap quickly disappear for increasing disorder as seen in the previous subsection, its effect can still be seen above the gap. The numerically derived response shows a continuous logarithmic rise from the gap edge rather than a discontinuous jump for all values of disorder and FSI. However, the effects of FSI only are evident for frequencies close to the gap edge. FSI have little qualitative effects on the spectrum for large frequencies. For large disorder the spectrum is dominated by the impurity peak at a frequency of roughly $1/\tilde{\tau}_L$. For small to moderate disorder, FSI sharpen the shape of the Raman spectra in the immediate vicinity of the gap edge, but as the value of disorder is increased, the spectral weight is shifted from the region near 2Δ to higher frequencies rather quickly and the effect of g_L diminishes. This effect persists long after the pole below 2Δ is no longer visible, but eventually disappears altogether for larger disorder. In fact for $1/2\Delta\tilde{\tau}_L > 5.0$, except for extremely strong FSI, i.e., $V_L/V_{L=0} > 0.99$, FSI effects can be ignored for all frequencies and the "pair approximation" becomes quite adequate. This is similar to the effects of FSI on both the clean[7] and dirty[8] density responses, which showed that collective effects quickly lose importance for large q.

In real systems, broadening due to gap anisotropy, inelastic quasiparticle collisions, or experimental resolution will cause the bound state to be indistinguishable from the Cooper continuum for small binding energies. For vanishing disorder, the bound state would be seen by additional spectral weight below the gap and an effective overall sharpening of the peak height and reduction of

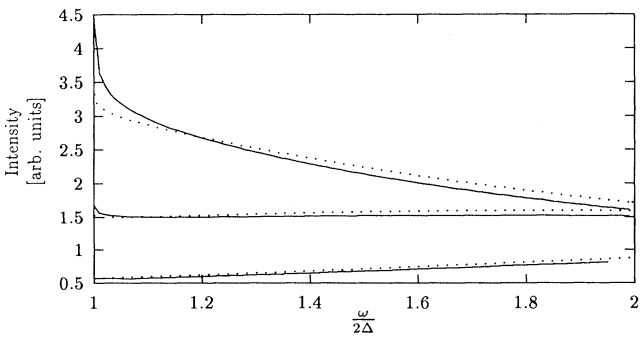

FIG. 1. Imaginary part of $\tilde{\chi}_L(\mathbf{q},\omega)$ above the gap for $V_L/V_{L=0} = 0.5$ (solid lines) and 0.3 (dotted lines). Upper (middle, lower) two curves are for $1/2\Delta\tilde{\tau}_L = 1.0$ (2.0, 5.0), respectively.

spectral weight at large frequencies.[2] Therefore, for different L channels, different line shapes can be obtained. Also, it has been shown in Ref. 6 that anisotropic impurity scattering, i.e., $\tilde{\tau}_L$ different for different L, can also cause polarization dependent line shapes. We see from Fig. 1 that disorder reduces the effect of g_L and thus only anisotropic impurity scattering can cause channel dependent spectra in disordered superconductors. Since the existing experimental data do not indicate a large bound state, one can conclude that for strong disorder, FSI can be ignored and its omission in Ref. 6 is justified in this case. In the following section we will consider the temperature dependence of the Raman spectra in the strong disorder limit where FSI can be neglected.

III. TEMPERATURE DEPENDENCE OF RAMAN SPECTRUM FOR LARGE DISORDER

We have seen in the previous section that the role of FSI diminishes rapidly for increasing disorder. Neglecting FSI, it was shown in Ref. 6 that in the limit of $1/2\Delta\tilde{\tau}_L \geq 1$ the peak of the Raman spectrum does not coincide with the gap edge and moves to higher frequencies with increasing disorder. Thus one would overestimate the gap energy if one identified the peak of the Raman spectrum with the gap, as it is in the clean case. Consequently, one would observe a non-BCS temperature dependence of the "gap" and it would appear that there were states below the "gap" even at $T = 0$. In this section we shall present a calculation of the Raman spectrum in a disordered superconductor for $T \neq 0$ neglecting FSI and examine the temperature dependence of the peak of the spectrum. As pointed out above, these calculations should be most appropriate for strongly disordered superconductors.

Neglecting FSI, the Raman spectrum is given by the imaginary part of $B_{+,L}$, which we take from Ref. 6 as

$$B_{+,L}(\mathbf{q}, i\omega) = \frac{1}{\pi}\int d\epsilon\, d\epsilon' \Phi''_L(\mathbf{q}, \epsilon - \epsilon') b_{\epsilon,\epsilon'}(i\omega), \tag{20}$$

with the spectral function

$$b_{\epsilon,\epsilon'}(i\omega) = \frac{EE' - \Delta^2 + \epsilon\epsilon'}{4EE'}\frac{f(E') - f(E)}{i\omega + E - E'} - \frac{EE' + \Delta^2 - \epsilon\epsilon'}{4EE'}\frac{1 - f(E') - f(E)}{i\omega - E - E'} + (i\omega \rightarrow -i\omega). \tag{21}$$

Here, $E^2 = \epsilon^2 + \Delta^2$, f denotes the Fermi function, and $(i\omega \rightarrow -i\omega)$ denotes the addition of terms which differ from the ones written only by the sign of $i\omega$. Φ''_L is the effective Raman density Kubo function for noninteracting electrons formed with the Raman vertex:

$$\Phi''_L(\mathbf{q}, \omega) = \sum_{\mathbf{kp}} \gamma_L \Phi''_{\mathbf{kp}}(\mathbf{q}, \omega)\gamma_L. \tag{22}$$

Within BCS theory, Φ''_L is the only source of disorder. For free electrons, the phase-space Kubo function $\Phi''_{\mathbf{kp}}$ can be expressed in terms of the Lindhard function. The Raman density Kubo function in the normal state has been well investigated, since it connects to the Raman susceptibility via the relation $\Phi''_L = \chi''_L/\omega$, which has been calculated in Refs. 6 and 13:

$$\chi''_{L,\mathrm{N.S.}}(\mathbf{q}, \omega) = \gamma_L^2 N_F \frac{\omega\tilde{\tau}_L^{-1}}{\omega^2 + \tilde{\tau}_L^{-2}}. \tag{23}$$

We rewrite Eq. (20) as the sum of two contributions:

$$B_{+,L}(\mathbf{q}, \omega) = B_{+,L}(\mathbf{q}, \omega, T = 0) + B_{+,L}(\mathbf{q}, \omega, T \neq 0). \tag{24}$$

The imaginary part of Eq. (20) for $T = 0$ has been calculated in Ref. 6, and we simply write down the result

$$B''_{+,L}(\mathbf{q}, \omega, T = 0) = \gamma_L^2 \frac{4\Delta\omega}{\omega + 2\Delta}\Theta(\omega - 2\Delta)\frac{N_F\tilde{\tau}_L^{-1}}{\omega^2 + \tilde{\tau}_L^{-2}} \times \left(\frac{(\omega - 2\Delta)^2}{4\Delta\omega}E(\beta) + \frac{\tilde{\tau}_L^{-2} + \omega^2 + 4\Delta\omega}{\tilde{\tau}_L^{-2} + \omega^2 + 2\Delta\omega}F(\beta) + \frac{8\Delta^2\omega^2}{(\omega^2 + \tilde{\tau}_L^{-2})^2 - 4\Delta^2\omega^2}\Pi(N, \beta)\right), \tag{25}$$

where

$$N = \frac{(\omega - 2\Delta)^2 + \beta^2\tilde{\tau}_L^{-2}(1 - \frac{4\Delta^2}{\omega^2\tilde{\tau}_L^{-2}})}{(\omega - 2\Delta)^2 + \tilde{\tau}_L^{-2}(1 - \frac{4\Delta^2}{\omega^2\tilde{\tau}_L^{-2}})}, \quad \beta = \frac{\omega - 2\Delta}{\omega + 2\Delta}.$$

Here Θ is the theta function and F, E, and Π are complete elliptical integrals of the first, second, and third kinds, respectively. For all values of $\tilde{\tau}_L^{-1}$, $B''_{+,L}(T = 0)$ is discontinuous at the threshold 2Δ. For $\omega \gg 2\Delta$, $B''_{+,L}(T = 0)$ approaches the normal state susceptibility, Eq. (23).[13] For large disorder, it was shown in Ref. 6 that the peak of the spectrum moves to frequencies above the gap, similar to the behavior seen in the large q limit for both clean and dirty $L = 0$ channels.[8,14] The impurity scattering rate replaces the role of momentum in $L \neq 0$ channels. The spectrum jumps at 2Δ to a value $\gamma_L^2 N_F \pi\Delta\tilde{\tau}_L$ and then rises linearly with ω for large disorder.

We could not find a way to express $B''_{+,L}(T \neq 0)$ in closed form by tabulated functions. In the limits of small and large frequencies, we find

$$B''_{+,L}(\mathbf{q}, \omega, T \neq 0) = \gamma_L^2 N_F \tilde{\tau}_L \omega \frac{1}{1 + e^{\Delta/T}} \quad \text{for } \omega \ll T, \tag{26}$$

$$B''_{+,L}(\mathbf{q},\omega,T\neq 0)=\gamma_L^2 N_F\tilde{\tau}_L\Delta K_1(\Delta/T)(1+\Delta/T),$$

$$\text{for } \omega\geq 2\Delta, \quad (27)$$

where K_1 is a Bessel function. For arbitrary frequencies at arbitrary temperatures, Eq. (20) must be evaluated numerically.

We plot $B''_{+,L}$ for several temperatures in Fig. 2. We normalized the response to the normal metal expression, Eq. (23). We see that the scattering intensity for $\omega < 2\Delta$ grows for increasing temperature due to the scattering from thermally excited quasiparticles, while there always exists a discontinuous jump at $2\Delta(T)$ due to scattering via pair creation. The results are qualitatively similar to the results for the clean case for large q.[15]

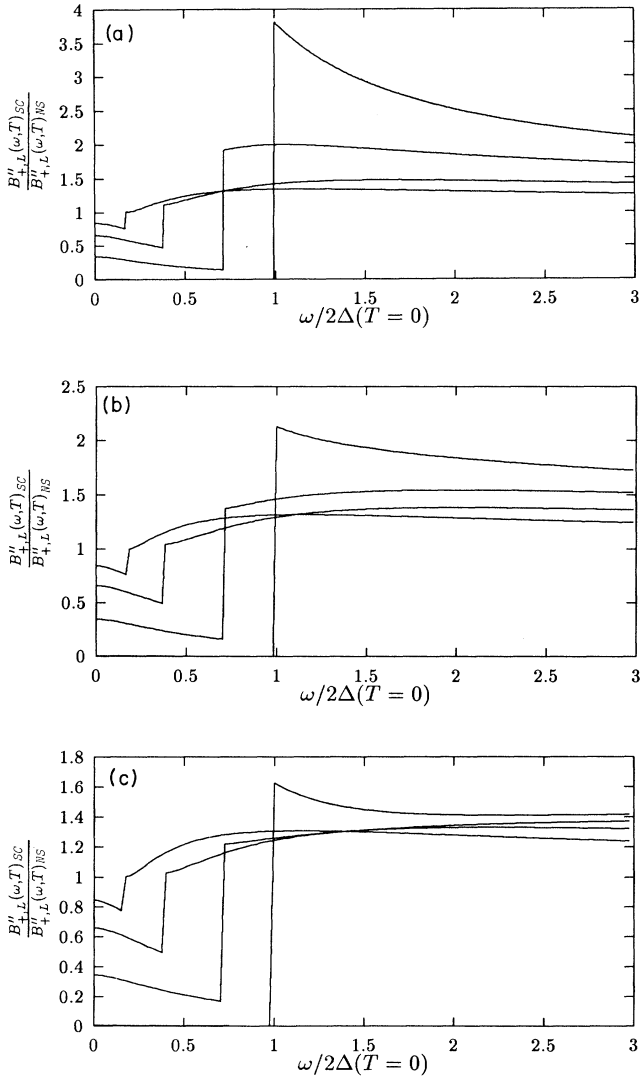

FIG. 2. Temperature dependence of the Raman spectrum normalized to the normal state value, neglecting FSI for $T/T_c = 0.3$ (highest curve), 0.8, 0.95, and 0.99 (lowest curve), respectively. Values of $2\Delta(T=0)\tilde{\tau}_L$ are (a) 1.2, (b) 0.6, and (c) 0.2, respectively.

In real systems there always exists some form of broadening of the Raman spectrum due to experimental resolution, gap anisotropy, or inelastic quasiparticle collisions that would smear the discontinuous onset of the spectrum at 2Δ. In Fig. 3 we plot the Raman spectrum for various temperatures for $2\Delta\tilde{\tau}_L = 1.2$ and 0.4 with a temperature independent broadening width $\Gamma = 0.3 \times 2\Delta$.[16] We see that (1) the spectrum inside the gap fills in for larger temperatures recovering its normal state Lorentzian behavior, Eq. (23), and (2) that the peak does not coincide with the gap for all temperatures and moves to higher frequencies for larger values of disorder. In fact for large scattering, the peak of the spectrum always occurs at the energy corresponding to the impurity scattering rate (as it does in the normal metal) which becomes the dominant energy scale in the system.

We thus see that the identification of the peak of the spectrum as the superconducting gap (as one would do in the case of clean superconductors) would overestimate the gap and lead to a temperature dependence which deviates substantially from BCS for large disorder. In Fig. 4 we plot the temperature dependence of the peak $\omega_p(T)$ of the Raman spectrum for various impurity lifetimes. It can be seen that for large impurity scattering rates the peak position saturates at one value — the impurity scattering rate — for increasing temperatures. For impurity concentrations such that $2\Delta\tilde{\tau}_L \simeq 1$, the peak position decreases as it does in the clean case but gradually levels off at a nonzero value for $T \to T_c$. For larger concentrations, the peak does not decrease at all and stays fixed at $\tilde{\tau}_L^{-1}$.

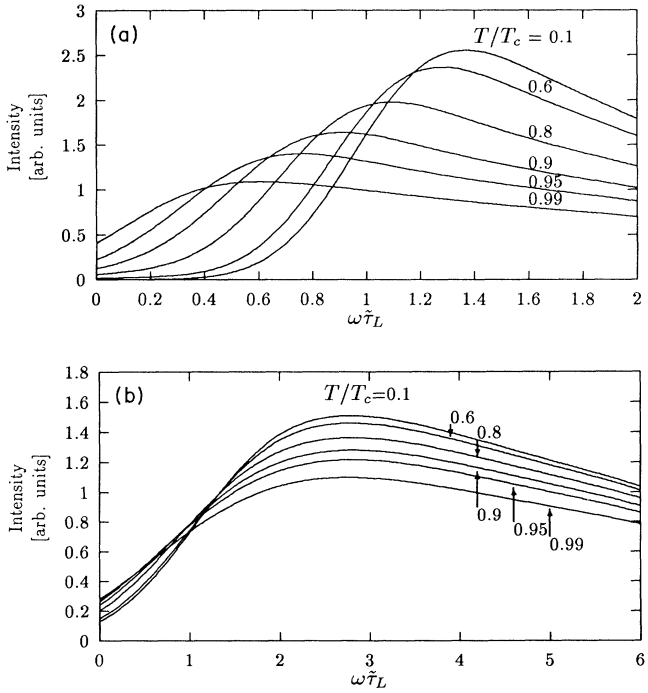

FIG. 3. Temperature dependence of the Raman spectrum neglecting FSI for (a) $2\Delta(T=0)\tilde{\tau}_L = 1.2$, and (b) $2\Delta(T=0)\tilde{\tau}_L = 0.4$, using a Gaussian width $\Gamma = 0.3 \times 2\Delta(T=0)$. Reduced temperatures ($T/T_c$) are indicated.

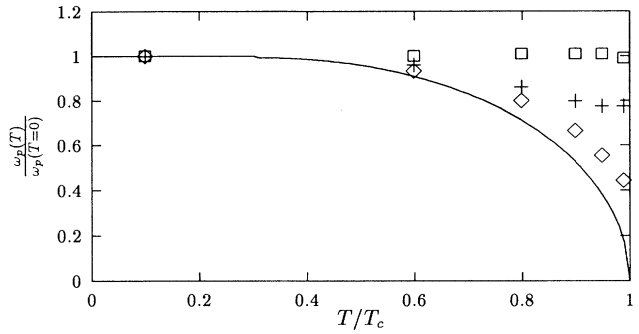

FIG. 4. Temperature dependence of the peak position $\omega_p(T)$ of the Raman spectrum. Solid line is the BCS result, while the diamonds (crosses, squares) correspond to values of $2\Delta(T=0)\tilde{\tau}_L = 1.2$ (0.8, 0.4), respectively.

An accurate and parameter free determination of the gap can be obtained from the static response. An important consequence of the static limit is that all vertex corrections vanish, as can be seen from Eqs. (6), (7), and (A6). Thus the response is given solely by $B''_{+,L}$.[6] Taking Eq. (26) and dividing it by its normal metal value ($\Delta = 0$) we obtain

$$\frac{B''_{+,L}(\mathbf{q},\omega=0,\Delta,T)}{B''_{+,L}(\mathbf{q},\omega=0,\Delta=0,T)} = \frac{2}{e^{\Delta(T)/T}+1}, \tag{28}$$

and the impurity scattering rate drops out. Thus the ratio of the superconducting and normal responses in the static are given by a Fermi function and the full temperature dependent gap can be extracted independently from disorder and FSI.[17]

IV. CONCLUSION AND RELEVANCE TO THE CUPRATE SUPERCONDUCTORS

We note that channel dependent, non-BCS behavior for the Raman spectrum has been seen in several cuprate superconductors.[18] Using values of $2\Delta\tilde{\tau}_L$ ranging from 0.9 to 1.2, both the temperature and polarization dependencies of the Raman spectrum on $YBa_2Cu_3O_7$ can be fit using the above theory. We note that the polarization orientations for which the temperature dependence of the obtained gap showed the greatest deviations from BCS also showed evidence for a finite density of states below the "gap." We note that this is consistent with the theory, since the "gap" identified from a pure BCS fit will be greater than the real gap and thus it would appear that there exists a finite density of states down to energies corresponding to the actual gap. While the magnitude of the impurity rate used is similar to the value recently needed to obtain fits to the changing phonon linewidths in these compounds,[19] we note that our fits require two polarization gaps with energies of 193 cm^{-1} and 310 cm^{-1}. Also, similar non-BCS temperature dependencies of the gap have also been seen in infrared conductivity measurements.[20] However, we note that the symmetry dependent deviations from BCS in the present theory are tied with the particular case of anisotropic mass fluctuations which couple directly to a Raman probe which selects many L channels, while an optical conductivity probe selects only the $L = 1$ channel. Therefore while a non-BCS "gap" can be obtained from the present theory applied to the case of the infrared conductivity, it seems unlikely that a similar explanation can be used to explain the channel dependence of the anomalous temperature dependence of the gap for the optical conductivity.[21]

We conclude that unlike the case for clean superconductors, careful attention must be paid in order to obtain information on the superconducting gap from Raman spectra at large disorder. Similarly, we have shown that FSI can be safely neglected in the limit of large disorder.

ACKNOWLEDGMENTS

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APPENDIX: ANALYTIC EXPRESSIONS FOR THE REAL PARTS OF A_L AND $B_{\pm,L}$

Our starting point in this appendix are Eqs. (2.20) and (2.21) of Ref. 8 for the response functions. We first calculate the static response. From Eq. (2.21a) of Ref. 8 we see that $A_L(\mathbf{q},0) = 0$. For $B_{\pm,L}(\mathbf{q},0)$ we choose a contour path that avoids a pole in the lower complex plane as discussed in Ref. 8 and obtain

$$B_{\pm,L}(\mathbf{q},0) = \frac{-\gamma_L^2 N_F}{\pi}\mathrm{Im}\int d\epsilon \int_\Delta^\infty \frac{ix\,dx}{\sqrt{x^2-\Delta^2}}\frac{1}{\pm\epsilon - ix}\frac{1}{\epsilon - ix - i\tilde{\tau}_L^{-1}}. \tag{A1}$$

For B_- we can perform the ϵ integral first and then the x integral to obtain

$$B_{-,L}(\mathbf{q},0) = 1/V_{L=0} - Y(2\Delta\tilde{\tau}_L), \tag{A2}$$

with Y given by Eq. (11) and

$$1/V_{L=0} \equiv N_F \int_\Delta^\infty \frac{dx}{\sqrt{x^2-\Delta^2}}.$$

For $B_{+,L}$ performing the ϵ integration first produces a result of the form $0\cdot\infty$. To avoid this we perform the x integral first and obtain

$$B_{+,L}(\mathbf{q},0)=\frac{\gamma_L^2 N_F\tilde{\tau}_L\Delta}{\pi}\mathrm{Im}\int d\epsilon[\epsilon I(i\epsilon)+(i/\Delta\tilde{\tau}_L-\epsilon)I(1/\Delta\tilde{\tau}_L+i\epsilon)], \tag{A3}$$

where

$$I(y)=\frac{\Delta}{\sqrt{-y^2+\Delta^2}}[\pi/2+i\arcsin(iy)].$$

The remaining integral can be done and we find that

$$B_{+,L}(\mathbf{q},0)=\gamma_L^2 N_F \tag{A4}$$

for all values of disorder.[10] This is similar to the clean case calculation, which showed that the static response, which reduces to $B_{+,L}$ as seen from Eq. (7), has no terms in $v_F q/\Delta$ to any order.[7]

We now calculate the real parts of the dynamical response functions in the limit of large and small disorders. Instead of first summing over Matsubara frequencies and then integrating over energies to calculate the real parts, it is convenient to first perform both of the energy integrations by using contours which are alternatingly closed in the upper and lower planes, respectively, leaving a Matsubara sum which reduces at zero temperature to

$$B_{\pm,L}(\mathbf{q},\omega)=\frac{\gamma_L^2 N_F}{2}\int d\Omega\frac{\frac{\Omega_+\Omega_--\Delta^2}{\sqrt{\Omega_+^2-\Delta^2}\sqrt{\Omega_-^2-\Delta^2}}\mp 1}{\sqrt{\Omega_+^2-\Delta^2}+\sqrt{\Omega_-^2-\Delta^2}+i/\tilde{\tau}_L}, \tag{A5}$$

$$A_L(\mathbf{q},\omega)=\frac{\gamma_L^2 N_F\omega\Delta}{2}\int\frac{d\Omega}{\sqrt{\Omega_+^2-\Delta^2}\sqrt{\Omega_-^2-\Delta^2}}\frac{1}{\sqrt{\Omega_+^2-\Delta^2}+\sqrt{\Omega_-^2-\Delta^2}+i/\tilde{\tau}_L}, \tag{A6}$$

with $\Omega_\pm=\Omega\pm\omega/2$. To make a connection to previous results we note that $B_{+,L}$ corresponds to $-\gamma_L^2\cdot I_{22}^-$ and $B_{-,L}$ corresponds to $\gamma_L^2\cdot I_{11}^-$ of Ref. 3. We next change variables of integration by letting $\Omega=x(\omega-2\Delta)/2$ and then expand the disorder dependent terms and take the real part of the expression. After performing the integral, we find that for small disorder, $1/2\Delta\tilde{\tau}_L\ll 1$, below the gap the response functions are given as

$$\frac{B_{-,L}(\mathbf{q},\omega)}{\gamma_L^2}=1/V_{L=0}+N_F\alpha\frac{\arcsin(\alpha)}{\sqrt{1-\alpha^2}}+\frac{N_F}{2\Delta\tilde{\tau}_L}\frac{E(\alpha)}{1-\alpha^2}, \tag{A7}$$

$$\frac{B_{+,L}(\mathbf{q},\omega)}{\gamma_L^2}=\frac{N_F}{\alpha}\frac{\arcsin(\alpha)}{\sqrt{1-\alpha^2}}-\frac{N_F}{(2\Delta\tilde{\tau}_L)\alpha^2}\left(\frac{2-\alpha^2}{1-\alpha^2}E(\alpha)-2F(\alpha)\right), \tag{A8}$$

$$\frac{A_L(\mathbf{q},\omega)}{\gamma_L^2}=\frac{N_F\arcsin(\alpha)}{\sqrt{1-\alpha^2}}-\frac{N_F}{(2\Delta\tilde{\tau}_L)\alpha}\left(\frac{1}{1-\alpha^2}E(\alpha)-F(\alpha)\right), \tag{A9}$$

while for strong disorder, $1/2\Delta\tilde{\tau}_L\gg 1$, the real parts are given by Eqs. (8)–(11) of Sec. II.[10] Here $\alpha=\omega/2\Delta$ and F and E are complete elliptical integrals of the first and second kinds, respectively. We note that for frequencies close to the gap edge, Eqs. (A7)–(A9) are not valid since we have neglected disorder dependent terms which are more singular in $1-(\omega/2\Delta)^2$ than the ones retained. It was shown in Ref. 6 that the imaginary parts of $A, B_\pm$ are constant at the gap edge and equal

$$B''_{+,L}(\mathbf{q},2\Delta)=B''_{-,L}(\mathbf{q},2\Delta)=A''_L(\mathbf{q},2\Delta)=\gamma_L^2 N_F\pi\Delta\tilde{\tau}_L.$$

Therefore, the real parts are logarithmically divergent at the gap edge. However, we find that the expansion for the disorder dependent term in Eqs. (A5) and (A6) are also correct asymptotically close to the gap edge, $\frac{1}{2\Delta\tilde{\tau}_L}\gg\sqrt{1-\alpha^2}$, and thus the behavior for the response functions is given by Eqs. (8)–(11) in this limit. For $\omega/2\Delta>1$ we can make a similar expansion of Eqs. (A5) and (A6) for the real parts and we arrive at Eqs. (16)–(18) which hold in the limit $\frac{1}{2\Delta\tilde{\tau}_L}\gg\sqrt{\alpha^2-1}$. Thus the response diverges logarithmically at the gap edge from both directions. This completes the purpose of this appendix.

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[10]We note that the expressions for $B_{+,L}$ in both the limits $1/2\Delta\tilde{\tau}_L \gg 1$, Eq. (9), and $1/2\Delta\tilde{\tau}_L \ll 1$, Eq. (A8), are not in agreement with the corresponding expression given in Ref. 3, and also that the expressions for the disorder correction to $B_{-,L}$, Eq. (A7), for small disorder disagrees with $B_{-,L}$ obtained in Ref. 3. These differences concern disorder corrections to the static response functions, which can be calculated analytically as shown in the Appendix. Thus we believe that the results obtained in Ref. 3 are incorrect since they predict disorder corrections to $B_{+,L}(\omega = 0)$ and give the wrong sign for the disorder correction to $B_{-,L}(\omega = 0)$. This is further clarified in the Appendix.
[11]While no analytic expressions could be found for the position and residue of the bound state for arbitrary $g_L/g_{L=0}$, we note that

$$\omega_0 = \sqrt{1 - \left(\frac{\pi\rho_L}{4(1+1/2\Delta\tilde{\tau}_L)}\right)^2}$$

and

$$Z_L = \rho_L^3/2\omega_0,$$

where

$$\rho_L = g_L \frac{1/g_{L=0} - Y(1/2\Delta\tilde{\tau}_L)}{1/g_L - 1/g_{L=0} + Y(1/2\Delta\tilde{\tau}_L)},$$

with Y given by Eq. (7), gives a fairly adequate phenomenological description of the disorder dependence of the bound state, with the best agreement for $1/2\Delta\tilde{\tau}_L > 0.5$.
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[21]However, it may be the case that fluctuations of transverse current which come from $L \neq 0$ components of FSI might be important in this case. This remains to be explored in the future.