Theory for the effects of impurities on the Raman spectra of superconductors

Thomas Peter Devereaux
Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-7000 Stuttgart 80, Federal Republic of Germany
(Received 6 December 1991)

A gauge-invariant theory for the effects of impurities on the electronic Raman scattering by pairs of quasiparticles in superconductors is presented. Special attention is given to the role of symmetry. The Raman spectrum for a superconductor is calculated using the exact-eigenstate formalism to account for impurity scattering in each symmetry channel L. We find that the Raman spectrum is strongly affected by impurity scattering in channels $L \neq 0$, and symmetry-dependent line shapes can be obtained for anisotropic impurity scattering. For strong impurity scattering the peak of the Raman spectra does not coincide with the energy gap. Also, the bound states and phonon spectral function in the $L \neq 0$ channel are investigated. Implications for the Raman spectra of the cuprates are discussed.

I. INTRODUCTION

The interest in Raman scattering in superconductors has grown tremendously with the results obtained from the experiments on both A-15's and high-T_c compounds.[1,2] While for many years various theoretical predictions for Raman scattering in superconductors predated the first experimental results, the experimental situation has now changed. Yet while there now exists a wealth of experimental data on more than a few systems, the theoretical work has been confined to the case of clean superconductors. Quite a detailed theory now exists for Raman scattering in clean BCS superconductors, where the effects of Coulomb interactions,[3] energy gap anisotropy,[4] and final-state residual phonon-mediated electron-electron interactions[5] have all been accounted for. These theories all point out the importance of large anisotropy of the effective Raman tensor as a requirement for large Raman cross sections. In the case of either a purely isotropic normal metal or a superconductor, at low temperatures one would only expect to see an electronic Raman signal for energy transfers near the plasmon frequency. The reason is that the isotropic density fluctuations produced by the incident light are coupled to long-range Coulomb forces and are thus screened by the collective plasma excitations. Therefore for low frequencies the Raman signal vanishes in the limit where the momentum transferred to the material approaches zero. This has been known for both normal metals[6–10] and superconductors[3–5] for some time. However, incident light with energy large enough to break a Cooper pair will in general produce excitations with different quantum numbers L, which can correspond to angular momenta in spherically symmetric metals or crystal harmonics in the more general case,[11] in materials with nonparabolic conduction energy bands. While the excitations with $L = 0$ correspond to the screened isotropic density fluctuations, excitations with quantum numbers $L \neq 0$ produce fluctuations in different regions of the Fermi surface that are antisymmetric in charge and thus produce no net charge fluctuations on a long-distance scale. Consequently, these excitations are not screened by Coulomb forces and can lead to a nonvanishing Raman signal in the zero-q limit.[3–10]

The effects of nonmagnetic impurities on Raman scattering in normal metals for $L \neq 0$ has also been well investigated. It has been shown that the interaction of electrons with impurities or phonons can further lead to an enhanced Raman signal. In the case of semiconductors, fluctuations from valley to valley mediated by either impurities or phonon-scattering processes do not couple to Coulomb forces and play a dominant role in Raman scattering.[7] The Raman spectra was found to have a Lorentzian form with widths that grow with increasing impurity concentrations. For metals, Ipatova *et al.*[8] showed that large Raman cross sections can be obtained for materials with nonparabolic conduction bands, while Zawadowski and Cardona[9] demonstrated how impurities strongly affect the width of the Lorentzian Raman cross section. Further, if the impurity scattering is anisotropic different line shapes can be obtained for different polarization orientations.

It is reasonable to believe that similar effects might be found in superconductors with significant concentrations of impurities. Since most of the A-15's and the high-T_c compounds which can be examined by light scattering have high normal state resistivities, it is plausible to believe that disorder may play a relevant role in these materials. Therefore it would be advantageous to have a theory of Raman scattering that incorporates the effects of disorder in a systematic fashion.

Light scattering experiments offer a unique opportunity to obtain symmetry-dependent material information. By altering the polarizations of both the incident and scattered light, one can selectively examine certain quantities that are dependent on a particular symmetry of the point group of the crystal. The experimental data have shown a rich and somewhat puzzling polarization dependence. Namely, the scattering intensity and line width both vary as the polarization directions are

changed. Therefore, a theory for Raman scattering in superconductors must show how the polarization orientations affect the scattering cross section.

This paper describes features of the Raman spectra due to the effects of impurities and polarization orientations in superconductors. It will be assumed that the superconductors are of the BCS s-wave-pairing-type. The plan of the paper is as follows; Sec. II will be devoted to formalism, Sec. III will concern the theoretical results for the Raman response functions in a disordered superconductor and the corresponding bound states as a function of impurity concentration and present comparisons of the theory with experimental data, Sec. IV will present a calculation for the optical phonon spectral function, and lastly Sec. V will discuss implications for the Raman spectra of the cuprate superconductors.

II. RAMAN RESPONSE FUNCTIONS FOR DISORDERED SUPERCONDUCTORS

A. "Effective" density correlation function

The Hamiltonian for electrons interacting with an external electromagnetic field is given by

$$H_{e-m} = \frac{e}{\hbar c}\sum_{\mathbf{k}} \mathbf{j}(\mathbf{k})\cdot\mathbf{A}(-\mathbf{k}) + \frac{e^2}{2\hbar^2 c^2}\sum_{\mathbf{k},\mathbf{k}'} A_\alpha(-\mathbf{k})\,\tau_{\alpha\beta}(\mathbf{k}+\mathbf{k}')\,A_\beta(-\mathbf{k}') \quad (1)$$

to second order in the vector potential $\mathbf{A}$ of the optical field. The current operator $j_\alpha(\mathbf{q})$ is given in terms of creation and annihilation operators $c^\dagger_\sigma(\mathbf{k}), c_\sigma(\mathbf{k})$ for an electron with spin σ as

$$j_\alpha(\mathbf{q}) = \sum_{\mathbf{k}} v_\alpha(\mathbf{k})\, c^\dagger_\sigma(\mathbf{k}+\mathbf{q}/2)\, c_\sigma(\mathbf{k}-\mathbf{q}/2), \quad (2)$$

where $v_\alpha(\mathbf{k}) = \frac{\partial\epsilon(\mathbf{k})}{\partial k_\alpha}$. The inverse effective mass tensor $\tau_{\alpha,\beta}$ is related to the curvature of the band energy $\epsilon(\mathbf{k})$ via

$$\tau_{\alpha,\beta}(\mathbf{q}) = \sum_{\mathbf{k}} \frac{\partial^2\epsilon(\mathbf{k})}{\partial k_\alpha \partial k_\beta} c^\dagger_\sigma(\mathbf{k}+\mathbf{q}/2), c_\sigma(\mathbf{k}-\mathbf{q}/2). \quad (3)$$

Applying Fermi's Golden Rule yields a differential Raman cross section given in terms of an "effective" density correlation function $\tilde{S}$:

$$\frac{d^2R}{d\omega\, d\Omega} = r_0^2 \tilde{S}(\mathbf{q},\omega), \quad (4)$$

where $r_0 = \frac{e^2}{mc^2}$ is the Thompson radius, and $\mathbf{q}$ and ω denote the momentum and energy transferred from the light to the material. From here on we set $\hbar = 1$. $\tilde{S}$ is related to the susceptibility by the fluctuation-dissipation theorem, $\tilde{S}(\mathbf{q},\omega) = -\frac{1}{\pi}[1+n(\omega)]\,\tilde{\chi}''(\mathbf{q},\omega)$ where $n(\omega)$ is the Bose distribution. The susceptibility

$$\tilde{\chi}(\mathbf{q},\omega) = <[\tilde{\rho}(\mathbf{q}),\tilde{\rho}(-\mathbf{q})]>_{(\omega)} \quad (5)$$

is formed with an "effective" density operator given by

$$\tilde{\rho}(\mathbf{q}) = \sum_{\mathbf{k}\sigma} \gamma(\mathbf{k}) c^\dagger_\sigma(\mathbf{k}+\mathbf{q}) c_\sigma(\mathbf{k}). \quad (6)$$

The strength of the scattering is given by the bare Raman vertex, $\gamma(\mathbf{k})$. For light with incident (scattered) polarization vectors $\mathbf{e}^i(\mathbf{e}^s)$, $\gamma(\mathbf{k})$ can be written in terms of the Raman tensor as[6,3]

$$\gamma(\mathbf{k}) = \sum_{\alpha,\beta} e^s_\alpha \gamma_{\alpha\beta}(\mathbf{k}) e^i_\beta, \quad (7)$$

with

$$\gamma_{\alpha\beta}(\mathbf{k}) = \delta_{\alpha,\beta} + \sum_\nu \left(\frac{(\mathbf{k}|v_\alpha|\mathbf{k}_\nu)(\mathbf{k}_\nu|v_\beta|\mathbf{k})}{\epsilon(\mathbf{k})-\epsilon(\mathbf{k}_\nu)+\omega_i} + \frac{(\mathbf{k}|v_\beta|\mathbf{k}_\nu)(\mathbf{k}_\nu|v_\alpha|\mathbf{k})}{\epsilon(\mathbf{k})-\epsilon(\mathbf{k}_\nu)-\omega_s} \right). \quad (8)$$

Here m_0 is the free-electron mass, ν denotes the band index of the electron excited out of the conduction band, and ω_i, ω_s denote incident, scattered light energies, respectively. $\gamma(\mathbf{k})$ does not depend on q since $q << k_F$. In the limit of small external frequencies, $\omega_i, \omega_s << |\epsilon(\mathbf{k})-\epsilon(\mathbf{k}_\nu)|$, Eq. (8) reduces to

$$\gamma_{\alpha\beta}(\mathbf{k}) = m_0 \frac{\partial^2\epsilon(\mathbf{k})}{\partial k_\alpha \partial k_\beta}. \quad (9)$$

In the case of a metal with free energy bands the Raman vertex is just the density vertex, $\gamma(\mathbf{k}) = 1$. Coulomb forces couple to the isotropic density fluctuations to screen them out completely for $\mathbf{q} = 0$ as a consequence of particle number conservation. However, the effective mass tensor is not a conserved quantity in general and for nonparabolic bands will have off-diagonal terms which do not couple to Coulomb forces.[10,12]

The remaining terms in the Hamiltonian, $H = H_{e-m} + H'$, are

$$H' = H_0 + H_{\rm int},$$

$$H_0 = \sum_{\mathbf{k},\sigma} \epsilon(\mathbf{k}) c^\dagger_\sigma(\mathbf{k}) c_\sigma(\mathbf{k}) + \sum_{\mathbf{k},\mathbf{k}',\sigma} V^{\rm imp}_{\mathbf{k},\mathbf{k}'} c^\dagger_\sigma(\mathbf{k}) c_\sigma(\mathbf{k}'),$$

$$H_{\rm int} = \sum_{\mathbf{k},\mathbf{k}'} V^{\rm int}_{\mathbf{k}',\mathbf{k}} c^\dagger_\uparrow(\mathbf{k}') c^\dagger_\downarrow(-\mathbf{k}') c_\downarrow(-\mathbf{k}) c_\uparrow(\mathbf{k}),$$

where $V^{\rm imp}$ and $V^{\rm int}$ 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. 5, 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 in Sec. III.

B. Raman response functions for disordered superconductors

We see from Eq. (4) that obtaining the Raman cross section in a superconductor amounts to calculating an effective density correlation function in a superconductor. In this section we will evaluate $\tilde{\chi}(\mathbf{q},\omega)$ in a gauge-invariant manner for dirty superconductors.

Reference 13 used the exact eigenstate formalism to construct a gauge-invariant theory for general correlation functions for BCS superconductors with arbitrary amounts of disorder, and explicitly showed the calculation for the density response and the longitudinal and transverse current responses. The idea is to first introduce new fictitious eigenfunctions $\Psi_n(\mathbf{x})$ and eigenenergies E_n, Anderson's "exact eigenstate" basis set,[14] that diagonalizes the noninteracting part of the Hamiltonian, H_0. The disorder renormalizations are then expressed in terms of correlation functions for noninteracting electrons. We will closely follow the formalism of Ref. 13 to calculate the "effective" density response of Eq. (5). Therefore we will be quite brief.

The graphical representation of the solutions for the susceptibility and the renormalized vertex are shown in Figs. 1 and 2 of Ref. 13. We first transform to the exact eigenstate basis set and perform an average over the random positions of the impurities according to the procedure outlined in detail in Ref. 13. The corresponding integral equations can be written as

$$\tilde{\chi}(\mathbf{q},i\Omega) = -T\sum_{i\omega}\int d\epsilon\, d\epsilon' \sum_{\mathbf{k},\mathbf{p}} F_{\mathbf{k},\mathbf{p}}(\mathbf{q};\epsilon,\epsilon')\mathrm{tr}\gamma(\mathbf{k},\mathbf{q})G(\epsilon,i\omega-i\Omega)\Gamma(\mathbf{p},-\mathbf{q};i\Omega)G(\epsilon',i\omega), \tag{10}$$

$$\Gamma(\mathbf{p},\mathbf{q};i\Omega) = \gamma(\mathbf{p},\mathbf{q}) - T\sum_{i\omega}\sum_{\mathbf{k}',\mathbf{p}'}\int d\epsilon\, d\epsilon' F_{\mathbf{k}',\mathbf{p}'}(\mathbf{q};\epsilon,\epsilon')\tau_3 G(\epsilon,i\omega-i\Omega)\Gamma(\mathbf{p}',\mathbf{q};i\Omega)\tau_3 V^{\mathrm{int}}(\mathbf{p},\mathbf{p}'), \tag{11}$$

where tr denotes the trace, and the Raman vertex $\gamma(\mathbf{p},\mathbf{q})$ is given by Eq. (7). The integral equations are identical to Eqs. (2.11)–(2.13) of Ref. 13 except a generalized interaction $V^{\mathrm{int}}(\mathbf{p},\mathbf{p}')$ has been used in place of the constant model potential, $-V$. Here, F is a correlation function for noninteracting electrons. It is defined in terms of the exact eigenstate wave functions as

$$F_{\mathbf{k},\mathbf{p}}(\mathbf{q};\epsilon,\epsilon') = \int d\mathbf{x}_1 d\mathbf{x}_2 d\mathbf{x}_3 d\mathbf{x}_4 e^{-i(\mathbf{k}-\mathbf{q}/2)\cdot\mathbf{x}_1 + i(\mathbf{k}+\mathbf{q}/2)\cdot\mathbf{x}_2 + i(\mathbf{p}-\mathbf{q}/2)\cdot\mathbf{x}_3 - i(\mathbf{p}+\mathbf{q}/2)\cdot\mathbf{x}_4} \times \sum_{m,n} < \Psi_n^*(\mathbf{x}_1)\Psi_n(\mathbf{x}_3)\Psi_m^*(\mathbf{x}_4)\Psi_m(\mathbf{x}_2)\delta(\epsilon-E_n)\delta(\epsilon'-E_m) >_{\mathrm{av}}, \tag{12}$$

where the brackets $< \cdots >_{\mathrm{av}}$ denote performing the impurity average. It has been discussed extensively in Refs. 13 and 15 that to a very good approximation $F_{\mathbf{k},\mathbf{p}}$ depends only on the difference $\epsilon-\epsilon'$ of its energy arguments and can be written in terms of the absorptive part of the phase-space density Kubo function[16] $\Phi''_{\mathbf{k},\mathbf{p}}(\mathbf{q};\epsilon-\epsilon')$:

$$F_{\mathbf{k},\mathbf{p}}(\mathbf{q};\epsilon,\epsilon') \approx (1/\pi)\Phi''_{\mathbf{k},\mathbf{p}}(\mathbf{q};\epsilon-\epsilon'). \tag{13}$$

As a correlation function for noninteracting electrons, Φ'' can be calculated by a variety of techniques. It contains all the disorder information in the system. Equations (10) and (11) are completely general ones that form a closed system of integral equations for the correlation function $\tilde{\chi}$. As discussed in Ref. 13, the above system of equations ensures gauge invariance if the matrix Green's function G is calculated in the generalized Hartree-Fock scheme which constitutes the standard theory of superconductivity. In this paper we choose to use the BCS model with an isotropic energy gap.[17]

Proceeding to solve to the integral equation we first expand the effective electron-electron interaction $V^{\mathrm{int}}(\mathbf{p},\mathbf{p}')$ and the bare Raman vertex $\gamma(\mathbf{k},\mathbf{q})$ in terms of a complete set of orthonormal functions defined on the Fermi surface:

$$V^{\mathrm{int}}(\mathbf{p},\mathbf{p}') = \sum_{L,L'} V_{L,L'}\phi_L^*(\mathbf{p})\phi_{L'}(\mathbf{p}'), \tag{14}$$

$$\gamma(\mathbf{k},\mathbf{q}) = \sum_L \gamma_L\phi_L(\mathbf{k}), \tag{15}$$

where we have used the fact that the Raman vertex is not strongly dependent on wave vector $\mathbf{q}$. The terms in the expansion take into account the anisotropy of V^{int} and γ by a classification in terms of quantum numbers L. In the general case, the functions $\phi_L(\mathbf{k})$ can be chosen to be Fermi surface harmonics,[11] and each index L can be identified with an irreducible representation of the point group of the crystal, but several L's belong to the same irreducible representation. In general, V^{int} can have off-diagonal as well as diagonal terms in the L basis, and thus the integral equations in general couple different channels L and L' which transform to the same irreducible representation. For simplicity we will assume that the

interaction is rotationally invariant. Thus we can use spherical harmonics for the functions $\phi_L(\mathbf{k})$, where L is a shorthand notation for $L = \{L, m\}$ in the three-dimensional case or azimuthal quantum numbers m for the two-dimensional case. Then $V_{L,L'} = \delta_{L,L'} V_L$ is diagonal in the spherical harmonics basis. The general case was discussed for clean superconductors in Ref. 4. Further, since we will be working in the BCS model, the first term in the expansion, $V_{L=0}$, will be the largest negative term in the series. The remaining terms can be of either sign.

The effect of a nonzero $V_{L\neq 0}$ has been considered regarding the existence of collective modes in both clean[18,5] and disordered[19] superconductors. It was found that for an attractive coupling, $V_L < 0$, an electron pair bound state with angular momentum L is formed which is orthogonal to the Cooper pairs, while for a repulsive interaction, an electron-hole exciton is formed. The resulting bound state has a lower energy than 2Δ and thus would show up as a precursor excitation before the Cooper edge continuum. It has also been shown that the bound states are not damped by disorder, but their binding energies are strongly affected and the bound state is pushed closer to the gap with increasing disorder. There so far has been no conclusive experimental evidence for a strongly bound state,[20] but even a bound state with small binding energy can dramatically alter the shape of the Raman spectrum, making it difficult to distinguish between gap anisotropy and collective mode effects.[5]

In order to proceed further, we make the assumption that the renormalized vertex is independent of $\mathbf{q}$ and thus $\Gamma_{L,L'} = \delta_{L,L'}\Gamma_L$ does not mix quasiparticles in different channels. The integral equation then decouples into separate integral equations for each channel. The above integral equations can be solved by expanding the vertex Γ_L in a quaternion basis and comparing coefficients.[13,21] The result is that the susceptibility in channel L can be expressed in terms of three response functions in a superconductor:

$$\tilde{\chi}_L(\mathbf{q}, i\Omega) = 2C_L(\mathbf{q}, i\Omega)/[1 + V_L C_L(\mathbf{q}, i\Omega)], \tag{16}$$

$$C_L(\mathbf{q}, i\Omega) = B_{+,L} + V_L A_L^2/(1 - V_L B_{-,L}). \tag{17}$$

Here the response functions in channel L are

$$A_L(\mathbf{q}, i\Omega) = i\Omega\frac{\Delta}{\pi}\int d\epsilon\, d\epsilon' \Phi''_{L,L}(\mathbf{q}, \epsilon - \epsilon') a_{\epsilon,\epsilon'}(i\Omega), \tag{18}$$

$$B_{\pm,L}(\mathbf{q}, i\Omega) = \frac{1}{\pi}\int d\epsilon\, d\epsilon' \Phi''_{L,L}(\mathbf{q}, \epsilon - \epsilon') b_{\epsilon,\pm\epsilon'}(i\Omega), \tag{19}$$

with the spectral functions

$$b_{\epsilon,\epsilon'}(i\Omega) = \frac{EE' - \Delta^2 + \epsilon\epsilon'}{4EE'}\frac{f(E') - f(E)}{i\Omega + E - E'} - \frac{EE' + \Delta^2 - \epsilon\epsilon'}{4EE'}\frac{1 - f(E') - f(E)}{i\Omega - E - E'} + (i\Omega \to -i\Omega), \tag{20}$$

$$a_{\epsilon\epsilon'}(i\Omega) = \frac{-1}{4EE'}\frac{f(E') - f(E)}{i\Omega + E - E'} - \frac{1}{4EE'}\frac{1 - f(E') - f(E)}{i\Omega - E - E'} + (i\Omega \to -i\Omega). \tag{21}$$

Here, $E^2 = \epsilon^2 + \Delta^2$, f denotes the Fermi function, and $(i\Omega \to -i\Omega)$ denotes the addition of terms which differ from the ones written only by the sign of $i\Omega$. $\Phi''_{L,L}$ is the "effective" Raman density Kubo function for noninteracting electrons formed with the Raman vertex:

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

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 $\omega\Phi'' = \chi''$.[16] The effect of impurities has also been well studied. Fal'kovsky[22] investigated the normal state Raman susceptibility in the presence of impurities, but only the $L = 0$ channel was investigated in detail. Zawadowski and Cardona[9] investigated the L-channel-dependent susceptibility in the zero-q limit with the assumption that the impurity potential is only weakly momentum dependent. We generalize their result to finite q by evaluating the Kubo function diagrammatically in the ladder approximation as in Ref. 9. We obtain

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

where N_F is the density of states per spin at the Fermi level. $\tilde{\tau}_L^{-1}$ can be written as

$$\tilde{\tau}_L^{-1} = \tau_{L=0}^{-1} - \tau_L^{-1} + Dq^2, \tag{24}$$

where D is the diffusion constant related to the resistivity ρ by an Einstein relation, $D^{-1} = 2e^2 N_F \rho$. The impurity scattering rate is related to the expansion of the impurity potential

$$|V^{\text{imp}}_{\mathbf{k},\mathbf{k}'}|^2 = \sum_L \phi_L^*(\mathbf{k})\Gamma_L\phi_L(\mathbf{k}')$$

via $1/\tau_L = 2\pi N_F \Gamma_L$, where we assume that the impurity potential is rotationally invariant. Additionally we assumed only a weakly $\mathbf{k}$ dependent impurity potential so that $\tau_{L=0}^{-1}$ will be larger than the scattering rate in any other channel. Light scattering in the L channel is given by particle-hole excitations. Thus the width of excitations in the L channel, $\tilde{\tau}_L^{-1}$, is due to scattering processes which take the excitations out of the L channel. Therefore vertex corrections of elastic scattering within the L channel reduce the scattering to another channel and subsequently reduce the total scattering rate.

For $q = 0$, we recover the form of $\Phi''_{L,L}$ from Ref. 9, while for the $L = 0$ channel the impurity scattering rate drops out due to particle number conservation in that channel and we recover the form from Ref. 22. There is no conservation rule in the $L \neq 0$ channels, which leads

to the violation of f-sum rules in that channel,[12] and thus the Kubo function does not go to zero for zero wave numbers. This is why the Raman spectrum in a normal metal does not vanish for $q \to 0$ in the $L \neq 0$ channel while it does for the case of isotropic density fluctuations (the $L = 0$ channel) where the excitations are screened out by long-range Coulomb forces. Furthermore, the bare diffusion pole that one has in the $L = 0$ channel is shifted by the impurities and thus has a pole at $q = 0$ for frequencies equal to the total impurity scattering rate in channel L.

In the case of isotropic scattering, $\tau_L >> \tau_{L=0}$, the position of the diffusion pole will be the same in different channels while for anisotropic scattering the pole of the Kubo function is channel dependent. This is why in the normal state Raman cross section one would expect polarization dependent line shapes and intensities only for the case of anisotropic impurity scattering.[9] This will have interesting implications for the superconducting case.

Returning to the calculation for a superconductor, we note that for isotropic density fluctuations ($L = 0$ channel), Eq. (23) for the Kubo function reduces to the density-density correlation function in the presence of disorder. The response functions for $L = 0$ were calculated in Ref. 13. It was found that a collective mode exists, the Anderson-Bogoliubov mode, in disordered superconductors as it does it clean superconductors. It was found that impurities simply normalize the speed of the sound mode. Inclusion of Coulomb interactions act as the Higgs mechanism to push the mode up to the plasmon frequency. Further, no disorder generated collective modes appear in the electromagnetic response.

Since the low-energy density fluctuations are screened in the $L = 0$ channel, they will not contribute to the Raman intensity. For $L = 0$, one only expects a large Raman signal for frequencies near the plasmon frequencies in superconductors. Thus for low frequencies the contributions to the Raman spectra only occur due to nonparabolic conduction band dispersion. This was shown by Abrikosov and Genkin, where the Raman vertex in the $L = 0$ channel was replaced by fluctuations of the Raman vertex away from sphericity but no channel dependence entered into the response function equivalent to $B_{+,L}$, Eq. (19). This is appropriate for clean superconductors with an isotropic energy gap and no final-state interactions, i.e., $V_{L\neq 0} = 0$. It is also appropriate for the additional case of isotropic disordered superconductors with isotropic impurity scattering rates since $\tilde{\tau}_L^{-1}$ is the same in each channel. However, anisotropy of either the gap, the final-state interactions, and/or the impurity scattering will lift the Raman spectra channel degeneracy. Therefore in most cases one would in general expect channel dependent Raman cross sections, and one must use both L dependent Raman vertices and susceptibilities in calculating the Raman cross section. This will be shown explicitly in the next section.

III. CHANNEL DEPENDENT SUSCEPTIBILITIES

In Ref. 13 the response functions in Eqs. (18) and (19) were evaluated for $T = 0$ in the $L = 0$ channel analytically for small $\mathbf{q}, \omega$, and were evaluated numerically for larger values. We now generalize those results to all L channels, and find analytic solutions for all $\mathbf{q}, \omega$.

It is convenient to first calculate the absorptive parts of the functions $A''_L, B''_{\pm,L}$. Since the imaginary parts of the spectra have delta functions representing energy conservation, one of the energy integrals can be readily performed. Analytically continuing to the real axis ($i\Omega \to \Omega + i0$) and taking the imaginary part of Eqs. (18)–(21) and performing one energy integration, we obtain for $T = 0$,

$$A''_L(\mathbf{q},\Omega) = \Theta(\Omega - 2\Delta)\frac{\Omega\Delta}{2}\int_\Delta^{\Omega-\Delta} \frac{dE}{\sqrt{E^2-\Delta^2}}\frac{1}{\sqrt{(\Omega-E)^2-\Delta^2}}\Phi_{+,L}, \tag{25}$$

$$B''_{\pm,L}(\mathbf{q},\Omega) = \Theta(\Omega - 2\Delta)\frac{1}{2}\int_\Delta^{\Omega-\Delta} \frac{dE}{\sqrt{E^2-\Delta^2}}\frac{1}{\sqrt{(\Omega-E)^2-\Delta^2}} \times\{[E(\Omega-E)+\Delta^2]\Phi_{+,L} \mp \sqrt{E^2-\Delta^2}\sqrt{(\Omega-E)^2-\Delta^2}\Phi_{-,L}\}, \tag{26}$$

where

$$\Phi_{\pm,L} = \Phi''_{L,L}[\mathbf{q}, \sqrt{E^2-\Delta^2} - \sqrt{(\Omega-E)^2-\Delta^2}] \pm \Phi''_{L,L}[\mathbf{q}, \sqrt{E^2-\Delta^2} + \sqrt{(\Omega-E)^2-\Delta^2}]. \tag{27}$$

The procedure for obtaining the corresponding real parts via a Kramers-Kronig transform is explicitly laid out in Ref. 13. Using Eq. (23) for the Kubo function, it is shown in the Appendix that the integrals can be performed analytically and can be expressed in terms of complete elliptical integrals:

$$B''_{+,L}(\mathbf{q},\Omega) = \gamma_L^2\frac{4\Delta\Omega}{\Omega+2\Delta}\Theta(\Omega-2\Delta)\frac{N_F\tilde{\tau}_L^{-1}}{\Omega^2+\tilde{\tau}_L^{-2}} \times\left(\frac{(\Omega-2\Delta)^2}{4\Delta\Omega}E(\alpha) + \frac{\tilde{\tau}_L^{-2}+\Omega^2+4\Delta\Omega}{\tilde{\tau}_L^{-2}+\Omega^2+2\Delta\Omega}F(\alpha) + \frac{8\Delta^2\Omega^2}{(\Omega^2+\tilde{\tau}_L^{-2})^2-4\Delta^2\Omega^2}\Pi(N,\alpha)\right), \tag{28}$$

$$B''_{-,L}(\mathbf{q},\Omega) = \gamma_L^2 \frac{4\Delta\Omega}{\Omega+2\Delta}\Theta(\Omega-2\Delta)\frac{N_F\tilde{\tau}_L^{-1}}{\Omega^2+\tilde{\tau}_L^{-2}+2\Delta\Omega}\left(\frac{2(\tilde{\tau}_L^{-2}+\Omega^2)}{\Omega^2+\tilde{\tau}_L^{-2}-2\Delta\Omega}\Pi(N,\alpha)-F(\alpha)\right), \tag{29}$$

$$A''_L(\mathbf{q},\Omega) = \gamma_L^2 \frac{4\Delta\Omega}{\Omega+2\Delta}\Theta(\Omega-2\Delta)\frac{N_F\tilde{\tau}_L^{-1}}{\Omega^2+\tilde{\tau}_L^{-2}+2\Delta\Omega}\left(\frac{4\Delta\Omega}{\Omega^2+\tilde{\tau}_L^{-2}-2\Delta\Omega}\Pi(N,\alpha)+F(\alpha)\right), \tag{30}$$

where

$$N = \frac{(\Omega-2\Delta)^2+\alpha^2\tilde{\tau}_L^{-2}(1-\frac{4\Delta^2}{\Omega^2\tilde{\tau}_L^{-2}})}{(\Omega-2\Delta)^2+\tilde{\tau}_L^{-2}(1-\frac{4\Delta^2}{\Omega^2\tilde{\tau}_L^{-2}})},$$

$$\alpha = \frac{\Omega-2\Delta}{\Omega+2\Delta}.$$

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 $\tilde{\tau}_L^{-1}$, the three spectra are discontinuous at the threshold 2Δ, which is the required energy to break a Cooper pair. For large frequencies compared to the gap, $\Omega >> 2\Delta$, $B''_{+,L}$ approaches the normal state susceptibility,[9] while $B''_{-,L}$ and A''_L approach $\pi/2$ and 0, respectively.

The existence of collective modes and the effects of disorder in $L \neq 0$ channels have been thoroughly investigated by Maki and Tsuneto, and Fulde and Strassler.[19] They found that the collective mode exists for arbitrary strength of the coupling V_L, and is undamped even in the presence of disorder. Similarly, interaction in the electron-hole channel also produces a bound state. However, disorder drastically affects the position of the collective mode, and pushes the mode closer to the gap edge for increasing disorder. We have verified these results by using the above functions in the expression for the renormalized polarizability, $C_L(\mathbf{q},\Omega)$, and obtain plots similar to Fig. 2 from the calculation by Maki and Tsuneto.[19] We find that except for sufficiently strong coupling, $V_L > 0.5V_{L=0}$, the collective mode has a small binding energy and is thus indistinguishable from the gap edge. This is in agreement with the lack of experimental verification of a collective mode.[20] However, we do note that even if the collective mode is very close to the gap edge, its presence strongly affects the line shapes of the susceptibility, making it difficult to distinguish between the effects of collective modes and possible gap anisotropies to produce a broadened gap edge. While at present one cannot exclude the possibility of collective modes in disordered superconductors, one can draw the conclusion that the mode has a very small binding energy. Beyond collective effects, it can be shown that the vertex corrections only modify the frequency dependence of the susceptibility in a narrow range of frequencies very close to the gap edge,[13] and thus the "pair approximation," i.e., the susceptibility without vertex corrections, is completely adequate for our purposes. Therefore, for the remainder of the paper we choose to ignore all vertex corrections and thus focus only on the response function $B_{+,L}$.

We have plotted $B''_{+,L}(\mathbf{q},\Omega)$ for different impurity scattering rates in Fig. 1. We see that the impurity scattering rate cuts off the square root singularity that one obtains in the clean BCS limit without final-state interactions. At the threshold, $\Omega = 2\Delta$, the response functions have the following form:

$$B''_{+,L}(\mathbf{q},\Omega=2\Delta) = B''_{-,L}(\mathbf{q},\Omega=2\Delta) = A''_L(\mathbf{q},\Omega=2\Delta) = \gamma_L^2 N_F \frac{\pi\Delta}{\tilde{\tau}_L^{-1}}.$$

The spectrum becomes flatter and the peak maximum moves out to higher frequencies for increasing impurity rates. Note that the spectra are qualitatively different from those of clean superconductors with isotropic energy gaps and no final-state interactions.[4,3] First, in the case for $\mathbf{q} = \mathbf{0}$, the peak of the spectrum always coincides with the gap edge for clean superconductors. However, impurities affect the height and position of the peak even for $\mathbf{q} = \mathbf{0}$ in the channels $L \neq 0$. Thus while for clean superconductors the identification of the peak maximum as the energy gap is appropriate, such an analysis would overestimate the value of the energy gap in superconductors with impurities. Second, in clean superconductors the channel dependence of the spectra enters only through the channel dependence of the Raman vertex. Since this is just an overall prefactor, only the magnitude of the spectrum would be channel dependent. However, in the dirty limit the channel dependence also enters through the impurity scattering rate, and thus the line shape of the spectrum is altered as well. These features are the same as in the normal metal case.[9] While these effects

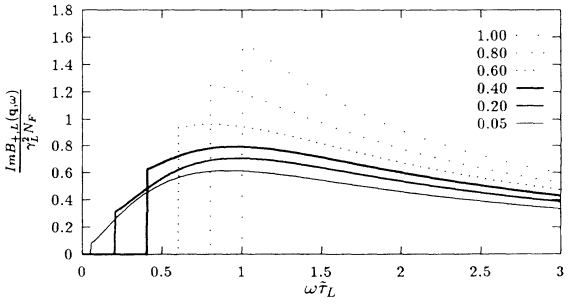

FIG. 1. Normalized response function $B''_{+,L}$ of Eq. (28) for different impurity scattering rates. The values of $2\Delta(T=0)/\tilde{\tau}_L^{-1}$ used are given in the upper right of the figure. The maximum of the peak position moves away from the gap edge for increasing scattering rates.

are small for weak impurity scattering, they become quite appreciable for $2\Delta\tilde{\tau}_L \sim 1$.

To demonstrate the effect of impurity-dependent line shapes, we have convoluted Eq. (28) with a Gaussian of fixed width to mimic the effects of broadening due to either gap anisotropy, inelastic scattering, or instrumental resolution. The overall magnitude of the scattering is governed by the Raman tensor, which is in principle obtainable from band-structure calculations but is yet unknown for even a simple metal such as aluminum. Using the energy gap and impurity scattering rate as fit parameters and a fixed Gaussian width $\Gamma = 0.10 \times 2\Delta$, we have fit the convoluted spectrum to the Raman data on V_3Si (Ref. 4) for different polarization geometries which select different contributions from A_{1g}, T_{2g}, and E_g symmetries. Since Ref. 4 did not normalize their data to the normal metal, the Raman vertex was simply used as a fit parameter. For the O_h point group, the channels with $L = 0, 4$ and higher angular momenta contribute to A_{1g} symmetry, while the channels with $L = 2$ contribute to both E_g and T_{2g} symmetries. The fits are shown in Figs. 2 and 3. We have fixed the gap at $2\Delta = 41$ cm^{-1} for each symmetry orientation. The fits show very good agreement using impurity scattering rates $\tilde{\tau}_L^{-1}/2\Delta = 0.1, (0.2)$ for $3/4E_g, (A_{1g} + E_g$, and $A_{1g} + 1/4E_g + T_{2g})$ symmetries, respectively.

The fits presented in Ref. 4 were obtained by assuming that the line shape was governed by a Gaussian distribution of energy gaps with widths ranging from $10 \sim 15\%$ of the gap. Additionally, the fit required an energy gap of E_g symmetry which was roughly 7% lower than the other symmetry gaps. Impurity scattering was not taken into account. Our fits do not require gap anisotropy. The different peak positions can be attributed to impurity scattering which is anisotropic and thus different for different L channels. Similarly, while a distribution of energy gaps could account for the magnitude of line broadening, it seems more likely that impurity enhanced inelastic quasiparticle scattering processes due to Coulomb and/or electron-phonon interactions is responsible for line broadening. A recent theory[23] predicts an inelastic scattering rate for the degree of disorder in this material that is comparable with the broadening parameter used in our fits. Thus our fits indicate that the gap for this material could be spherically symmetric or have a much smaller percentage of the gap that deviates from 41 cm^{-1}.

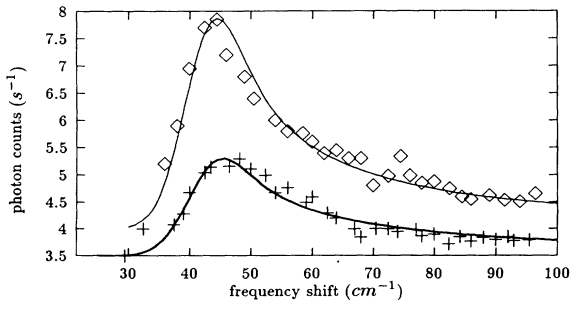

FIG. 2. Comparison of theory with experimental data on V_3Si from Ref. 4 for $\frac{3}{4}$ A_{1g} (upper line) and $A_{1g} + B_g$ symmetries (bottom line), respectively. Parameters used are defined in the text.

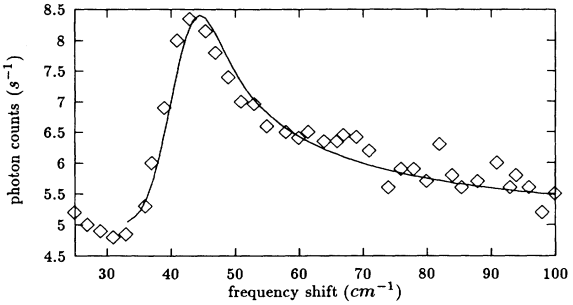

FIG. 3. Comparison of theory with experimental data on V_3Si from Ref. 4 for $A_{1g} + \frac{3}{4}$ $B_g + T_{2g}$ symmetry. Parameters used are defined in the text.

IV. PHONON SPECTRAL FUNCTION

The influence of superconductivity on phonon frequencies has received much interest. Zeyher and Zwicknagl[24] have considered the case of strongly coupled superconductors in the presence of nonmagnetic impurity scattering, and were able to account for the phonon frequency shifts and changing line widths in $YBa_2Cu_3O_7$ (Ref. 25) as a function of temperature. It was found that the presence of impurity scattering was essential to the theoretical agreement.

For completeness, we now consider a calculation of the optical phonon spectral function in disordered BCS superconductors. The spectral function for an optical phonon is given by

$$\mathrm{Im}D(\mathbf{q}=0,\omega) = \frac{4\omega_0^2\Sigma''(\omega)}{[\omega^2 - \omega_0^2 - 2\omega_0\Sigma'(\omega)]^2 + 4\omega_0^2\Sigma''^2}, \tag{31}$$

where ω_0 is the optical phonon frequency and Σ', Σ'' are the real and imaginary parts of the $\mathbf{q} = \mathbf{0}$ phonon self-energy, respectively. The interaction of optical phonons and electrons is assumed to be of the simple form:

$$H_{e\text{-ph}} = \sum_{\mathbf{k},\mathbf{q},\gamma} g_{\mathbf{k}}^{\gamma} c_{\mathbf{k}+\mathbf{q},\sigma}^{\dagger} c_{\mathbf{k},\sigma} (b_{\mathbf{q},\gamma}^{\dagger} + b_{-\mathbf{q},\gamma}), \tag{32}$$

where $g_{\mathbf{k}}^{\gamma}$ is the matrix element for scattering an electron from $\mathbf{k} \rightarrow \mathbf{k} + \mathbf{q}$, and $b_{\mathbf{q},\gamma}$, $b_{\mathbf{q},\gamma}^{\dagger}$, are the phonon field operators for branch γ. In this place the scattering occurs via a phonon instead of a photon, and one can thus proceed along the lines considered for photon scattering with the substitution of the Raman vertex $\gamma_{\mathbf{k}}$ by the electron-phonon coupling vertex $g_{\mathbf{k}}$. The full vertex renormalization and self-energy equations are thus described by Eqs. (18) and (19) with the replacement of $g_{\mathbf{k}}$ expanded in terms of spherical harmonics.

It was shown by Littlewood and Varma[26] that Coulomb interactions supress interaction effects in the

$L = 0$ channel in the same way as the Raman scattering is supressed in that channel. Therefore, the renormalization of the phonon spectral function only comes from anisotropic couplings in the $L \neq 0$ channels.

Ignoring vertex corrections due to final-state interactions we have plotted the $L \neq 0$ phonon spectral function in Fig. 4 for various impurity scattering rates using the response function $B''_{+,L}(\mathbf{q} = \mathbf{0}, \omega)$ of Eq. (28) for the self-energy $\Sigma''(\omega)$. The real parts were obtained numerically by a Kramers-Kronig transform. We find the same conclusions reached in Ref. 24, namely, (1) that for phonon frequencies below the gap, impurities smear out the satellite peak above 2Δ, and the spectral weight of the satellite decreases with increasing distance from the gap edge and (2) for phonon frequencies above the gap, impurities again smear out the precursor excitation which exists below the gap and an asymmetric phonon peak is obtained for phonon energies close to the gap. This can be seen in Figs. 4(a) and 4(b).

While the satellite peak that appears below 2Δ has been previously invoked to explain the Raman peaks below the gap in $NbSe_2$ (Ref. 26) and Nb_3Sn,[27] we find that the peak should merge into the phonon peak for increasing concentration of impurities. However a different explanation in terms of coupling of superconducting and modulating charge density energy gap modes has been presented.[26] The disappearance of the precursor peak with increasing impurity concentrations has been observed for $NbSe_2$.[28] In the high-T_c compounds, so far no anomalous spectral function features have been observed (except for the asymmetric Fano resonances[25]) even though there exists several phonons near the gap that could produce structure. Our results are in agreement with these experiments. It seems likely that impurity scattering is strong enough in these compounds to smear out any anomalous features in the spectral function.

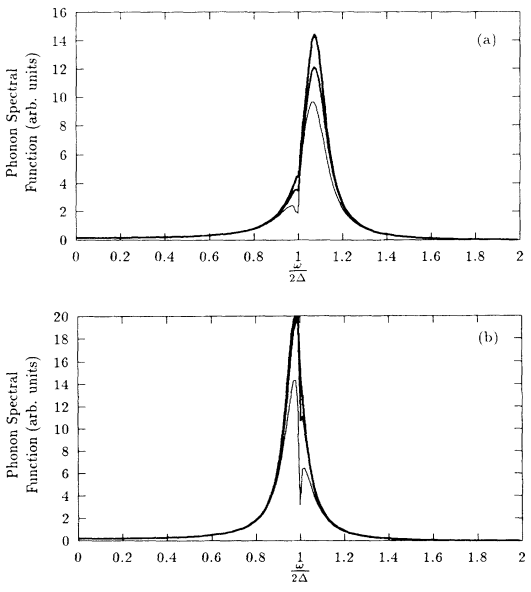

FIG. 4. Phonon spectral function for an optical phonon of energy (a) 1.05 $\times 2\Delta$, (b) 0.95 $\times 2\Delta$, respectively. Lower, middle, and upper lines are for $\tilde{\tau}_L^{-1}/2\Delta = 0.1$, 0.5, and 1.0, respectively, for both figures.

V. DISCUSSION AND IMPLICATIONS FOR THE CUPRATES

We have obtained expressions for the Raman response functions in superconductors including modifications due to impurities. We have demonstrated that impurities dramatically affect the shape of the Raman spectrum, and we have pointed out that the identification of the gap with the position of the maximum of the Raman spectra will overestimate the value of the energy gap. Good agreement is found with the experimental data on V_3Si and the general behavior of the Raman spectra of other A-15 compounds. We now turn the discussion to the implications for the Raman spectra of the cuprates.

The unusually flat Raman spectrum extending out to approximately 1 eV in the cuprate superconductors has signaled the development of various theories to explain this anomalous normal state behavior. The "marginal-Fermi-liquid" hypothesis was borne from the Raman spectra in the normal state, leading to a phenomenological form for the susceptibility $\tilde{\chi}''(\omega, T)$ which is linear in both frequency and temperature.[29] Recently it has been shown that a form for the susceptibility suggested in Ref. 29 can be derived from consideration of a nested or partially nested Fermi surface,[30] a feature that many of the high-T_c compounds and some actinides share. Good agreement was found with the normal state data on $BiSrCaCu_2O_8$.[31] The key to the theory was the presence of nonparabolic conduction bands, which allows for large Raman intensities, and the frequency dependence of the quasiparticle damping rate due to electron-electron collisions on parts of the nested Fermi surface, which was found to be linearly dependent on frequency. However, analysis was restricted to the normal state, since at low temperatures and frequencies the theory breaks down, and thus the superconducting state could not be examined.[32] It is estimated that the frequency and temperature scale for which normal Fermi-liquid behavior is recovered is at least on the order of the superconducting gap and T_c in the high-T_c compounds.[30] Therefore although the normal state might be "marginal," it may be plausible that there is still a Fermi liquid in the superconducting state and one may use Fermi-liquid-based approaches to examine Raman scattering.

However, the Fermi-liquid-based theories have yet to describe the Raman spectra in the superconducting state. The experimental work of Ref. 33 on $YBa_2Cu_3O_7$ modeled their data with a BCS form of the Raman spectrum, which led to polarization dependent gaps. Further, the temperature dependence of the gap obtained differed greatly from a BCS T dependence, and it appeared as though the gap saturated at $T = T_c$ for certain symmetries. Also the fits required gap anisotropies as large as 80% and distributions of gaps that range from 27 to 67% of the mean-gap values. In view of the fact that most of the polarization data are taken in-plane, it seems un-

likely that the superconducting gap could have such a large degree of anisotropy as the fits require if the pairing is isotropic, i.e., S wave.[34] Further, the high normal state resistivities in the cuprates in general suggest that the impurity scattering length is of the order of the coherence length. This is equivalent to the magnitude of the scattering rate required to obtain the fits of Ref. 24. Therefore it seems unlikely that any gap anisotropy would remain. However, the broadening which is observed is of the correct order of magnitude to be described by the disorder enhancement of quasiparticle inelastic scattering processes due to either Coulomb or electron-phonon interactions depending on the strength of the electron-phonon coupling. For $T/T_c = 0.3$, one obtains a quasiparticle scattering rate due to Coulomb interactions that is roughly 30% that of the energy gap.[23]

We have attempted to fit the data of Ref. 33. Convoluting Eq. (28) with a Gaussian of fixed width $\Gamma = 0.3 \times 2\Delta$, we obtain excellent agreement with Fig. 2 of Ref. 33 for the four polarization orientations selected. However, the fits require polarization dependent gaps. While the gap was set to be 193 cm^{-1} for the 0° $\perp$, 0° $\|$, and 45° $\|$ polarization orientations, a value of 310 cm^{-1} was needed for the 45° $\perp$ polarization orientation. The notation is that used in Ref. 33. The impurity scattering rate, $\tilde{\tau}_L^{-1}/2\Delta$, was chosen to be roughly equal to 1 for each of the four orientations. While this is consistent with the magnitude of the impurity scattering rate used in Ref. 24, it does not seem to be in agreement with such a large degree of gap anisotropy. This might be indicative of anisotropic electron pairing.[34]

Also, while good agreement is found for low frequencies, at higher frequencies, the present theory deviates substantially from experiment since the theoretical spectrum dies off as $1/\omega$ while a flat spectrum is seen roughly up to 1 eV. However, this can be remedied phenomenologically by assuming a constant impurity scattering rate at low frequencies, and for frequencies much larger than the gap, an effective frequency dependent scattering rate is used such that the susceptibility has the form suggested in Refs. 30 and 29. This interpretation agrees with the theory of Ref. 30, if one assumes that the lower frequency cutoff occurs at roughly 4Δ.

Lastly, we close with a discussion of the temperature dependence of the energy gap obtained in Ref. 33. The large value for the impurity scattering rate required by our fits has interesting implications for the temperature dependence of the energy gap. One can see from Fig. 1 that the peak of the Raman spectra should be much higher than the energy gap for $\tilde{\tau}_L^{-1} \sim 2\Delta$. Similarly in the normal state, a peak would be obtained at frequencies near $\tilde{\tau}_L^{-1}$. Therefore, one can expect that the maximum of the Raman spectra would drop with decreasing temperature and then saturate at frequencies roughly equal to the impurity scattering rate as the line shape recovers the Lorentzian normal state form. If one equated the peak of the Raman spectra with the energy gap, then it would appear as if the energy gap never vanishes as one reaches T_c. Therefore one would interpret a temperature dependence of the energy gap which was substantially different from BCS. This has been seen in Ref. 33. It is important to note that strong coupling effects also produce a non-BCS temperature dependence of the gap. However, the gap must go to zero at T_c, and therefore strong coupling effects cannot explain the behavior seen in Ref. 33. This further points out the care that is needed in interpreting the Raman data in superconductors. One can conclude that more experimental data is needed to examine the impurity concentration dependence of the theory, and more theoretical work is needed to account for the magnitude of the parameters required for the fits presented.

ACKNOWLEDGMENTS

The author would like to thank A. Zawadowski for remarks, communicated by D. Belitz, which led to this investigation, and thank both for correspondence and early encouragement in this work. I would also like to thank M. Cardona, P. Fulde, C. Thomsen, and G. Zwicknagl for many useful discussions. I am particularly indebted to P. Fulde and D. Belitz for critically reading the manuscript. Lastly, I would like to thank P. Fulde and the Max-Planck-Institut for financial support.

APPENDIX: EXACT SOLUTION OF SPECTRAL RESPONSE FUNCTIONS

In this appendix we show the exact solutions of Eqs. (25)–(27) for the spectrum of the response functions $A''_L, B''_{\pm,L}$. We begin by first changing the integration variable $E = \frac{1}{2}(x + \Omega - 2\Delta)$ and integrating over x. The products under the square root can be factorized:

$$\sqrt{E^2 - \Delta^2}\sqrt{(\Omega - E)^2 - \Delta^2}$$

$$= \tfrac{1}{4}(\Omega - 2\Delta)(\Omega + 2\Delta)\sqrt{(1 - \alpha^2 x^2)(1 - x^2)},$$

with α defined to be

$$\alpha = \frac{\Omega - 2\Delta}{\Omega + 2\Delta}.$$

Using Eq. (23) for the spectrum of the Raman Kubo function into the functions $\Phi_{\pm,L}$ of Eq. (27) and carrying out some algebra, we then can rewrite Eqs. (25) and (26) in the following form:

$$B''_{+,L}(\mathbf{q},\Omega) = \frac{\Theta(\Omega - 2\Delta)(\Omega + 2\Delta)N_F\tilde{\tau}_L^{-1}}{2(\Omega - 2\Delta)^2(\Omega^2 + \tilde{\tau}_L^{-2})}\int_0^1 dx \frac{b_0 + b_2 x^2 + b_4 x^4}{(x^2 + \omega_1)\sqrt{(1 - \alpha^2 x^2)(1 - x^2)}}, \tag{A1}$$

$$B''_{-,L}(\mathbf{q},\Omega) = \frac{\Theta(\Omega-2\Delta)(\Omega+2\Delta)N_F\tilde{\tau}_L^{-1}}{2(\Omega-2\Delta)^2(\Omega^2+\tilde{\tau}_L^{-2})}\int_0^1 dx \frac{b'_0+b'_2x^2}{(x^2+\omega_1)\sqrt{(1-\alpha^2x^2)(1-x^2)}}, \tag{A2}$$

$$A''_L(\mathbf{q},\Omega) = \frac{\Theta(\Omega-2\Delta)(2\Omega\Delta)N_F\tilde{\tau}_L^{-1}}{(\Omega+2\Delta)(\Omega-2\Delta)^2(\Omega^2+\tilde{\tau}_L^{-2})}\int_0^1 dx \frac{a_0+a_2x^2}{(x^2+\omega_1)\sqrt{(1-\alpha^2x^2)(1-x^2)}}, \tag{A3}$$

with

$$\omega_1 = \frac{\tilde{\tau}_L^{-2}(\tilde{\tau}_L^{-2}+\Omega^2-4\Delta^2)}{(\Omega-2\Delta)^2(\Omega^2+\tilde{\tau}_L^{-2})}.$$

The coefficients are defined as

$$b_0 = 8\Delta^2\alpha + 2\tilde{\tau}_L^{-2}\frac{\Omega^2+4\Delta^2}{(\Omega+2\Delta)^2},$$

$$b_2 = -2\alpha^2(\tilde{\tau}_L^{-2}-\Omega^2-8\Delta^2),$$

$$b_4 = -2\alpha^2(\Omega-2\Delta)^2,$$

$$b'_0 = 2\alpha\Omega^2 + 2\tilde{\tau}_L^{-2}\frac{\Omega^2+4\Delta^2}{(\Omega+2\Delta)^2},$$

$$b'_2 = -2\alpha^2(\tilde{\tau}_L^{-2}+\Omega^2),$$

$$a_0 = 2\tilde{\tau}_L^{-2}+\Omega^2-4\Delta^2, \quad a_2 = (\Omega-2\Delta)^2.$$

The above integrals of the form

$$I_n = \int_0^1 dx \frac{x^n}{(x^2+\omega_1)\sqrt{(1-\alpha^2x^2)(1-x^2)}}$$

can be expressed recursively in terms of complete elliptical integrals of the third kind, Π:

$$I_0 = \frac{1}{\omega_1}\Pi\left(-\frac{1}{\omega_1},\alpha\right),$$

$$I_2 = F(\alpha) - \frac{\omega_1}{\alpha}I_0,$$

$$I_4 = F(\alpha) - E(\alpha) - \frac{\omega_1}{\alpha}I_2,$$

where F and E are complete elliptical integrals of the first and second kinds, respectively. To transform the third elliptical integral in the circular case to positive arguments we utilize the tranformation

$$\Pi\left(-\frac{1}{\omega_1},\alpha\right) = \frac{\omega_1(1-\alpha^2)}{(1+\omega_1)(1+\omega_1\alpha^2)}\Pi(N,\alpha) + \frac{\omega_1\alpha^2}{1+\omega_1\alpha^2}F(\alpha), \tag{A4}$$

with $N = \frac{1+\omega_1\alpha^2}{1+\omega_1}$. After some straightforward but tedious algebra, we arrive at the form of the solutions for the spectra given by Eqs. (28)–(30). This completes the purpose of the Appendix.

[1] S.L. Cooper and M.V. Klein, Comments Condens. Mat. Phys. **15**, 99 (1990), and references therein.

[2] M. V. Klein, in *Superconductivity in d- and f-Band Metals*, edited by W. Buckel and W. Weber (Kernforschungszentrum Karlsruhe, Karlsruhe, 1982).

[3] A.A. Abrikosov and V.M. Genkin, Zh. Eksp. Teor. Fiz. **65**, 842 (1973) [Sov. Phys. JETP **38**, 417 (1974)].

[4] M. V. Klein and S. B. Dierker, Phys. Rev. B **29**, 4976 (1984).

[5] H. Monien and A. Zawadowski, Phys. Rev. B **41**, 8798 (1990).

[6] P. A. Wolff, Phys. Rev. **171**, 436 (1968).

[7] G. Contreras, A. K. Sood, and M. Cardona, Phys. Rev. B **32**, 924 (1985); I. P. Ipatova, A. V. Subashiev, and V. A. Voitenko, Solid State Commun. **37**, 893 (1981).

[8] I. P. Ipatova, M. I. Kaganov, and A. V. Subashiev, Zh. Eksp. Teor. Fiz. **84**, 1830 (1983) [Sov. Phys. JETP **57**, 1066 (1983)].

[9] A. Zawadowski and M. Cardona, Phys. Rev. B **42**, 10 732, (1990).

[10] B. S. Shastry and B. I. Shraiman, Phys. Rev. Lett. **65**, 1068 (1990); Int. J. Mod. Phys. B **5**, 365 (1991).

[11] P. Allen, Phys. Rev. B **13**, 1416 (1976).

[12] J. Kosztin and A. Zawadowski, Solid State Commun. **78**, 1029 (1991).

[13] D. Belitz, S. De Souza-Machado, T. P. Devereaux, and D. W. Hoard, Phys. Rev. B **39**, 2072 (1989).

[14] P. W. Anderson, J. Phys. Chem. Solids **11**, 26 (1959).

[15] B. Keck and A. Schmid, J. Low Temp. Phys **24**, 611 (1976); see also E. Abrahams, P.W. Anderson, P. A. Lee, and T. V. Ramakrishnan, Phys. Rev. B **24**, 6783 (1981).

[16] See, e.g., D. Forster, *Hydrodynamic Fluctuations, Broken Symmetry, and Correlation Functions* (Benjamin, New York, 1975).

[17] However, in order to handle the disorder averaging correctly to preserve gauge invariance, the choice of BCS-Gorkov Greens' functions (L. P. Gorkov, Zh. Eksp. Teor. Fiz. **37**, 1407 (1959) [Sov. Phys. JETP **37**, 998 (1960)]) forces us to make a "noncrossing" approximation, which is equivalent to the factorization $< G\Gamma G >_{\rm av} \approx < G > \Gamma < G >_{\rm av}$. Diagrammatically, this means that we do not allow crossings of interaction and impurity lines. If in the future it

should become possible to avoid the noncrossing approximation in calculating G, it will be straightforward to adopt the present theory accordingly.

[18] T. Tsuneto, Phys. Rev. **118**, 1029 (1960); A. Bardasis and J. R. Schrieffer, *ibid.* **121**, 1050 (1961).

[19] K. Maki and T. Tsuneto, Prog. Theor. Phys. (Kyoto) **28**, 163 (1962); P. Fulde and S. Strassler, Phys. Rev. **140**, A519 (1965).

[20] For a review, see P. Martin, in *Superconductivity*, edited by R. Parks (Marcel Dekker, London, 1969).

[21] R.E. Prange, Phys. Rev. **129**, 2495 (1963).

[22] L. A. Fal'kovsky, Zh. Eksp. Teor. Fiz. **95**, 1146 (1989) [Sov. Phys. JETP **68**, 661 (1989)].

[23] T. P. Devereaux and D. Belitz, Phys. Rev. B **43**, 3736 (1991); **44**, 4587 (1991).

[24] R. Zeyher and G. Zwicknagl, Z. Phys. B **78**, 175 (1990).

[25] C. Thomsen and M. Cardona, in *Physical Properties of High T_c Superconductors*, edited by D. M. Ginsberg (World Scientific, Singapore, 1989); C. Thomsen, in *Festkörperprobleme/Advances in Solid State Physics*, edited by U. Rössler (Vieweg, Braunschweig, 1991), Vol. 31.

[26] P. B. Littlewood and C. M. Varma, Phys. Rev. Lett. **47**, 811 (1981); Phys. Rev. B **26**, 4883 (1982).

[27] R. Hackl, R. Kaiser, and W. Glaser, Physica **162-164**, 431 (1989); X. L. Lei, C. S. Ting, and J. L. Birman, Physica **135B**, 486 (1985).

[28] R. Sooryakumar, M. V. Klein, and R. F. Frindt, Phys. Rev. B **23**, 3222 (1981).

[29] C. M. Varma, P. B. Littlewood, S. Schmitt-Rink, and E. Abrahams, Phys. Rev. Lett. **63**, 1996 (1989).

[30] A. Virosztek and J. Ruvalds, Phys. Rev. Lett. **67**, 1657 (1991).

[31] T. Staufer, R. Hackl, and P. Müller, Solid State Commun. **75**, 975 (1990).

[32] The reason is that at low temperatures and/or frequencies, the quasiparticle will be so narrowly confined to the orbit that the trajectory has no nested overlap and thus the exemptions to the phase-space limitations that give rise to the large scattering rate are gone, and the response reverts back to a conventional Fermi liquid.

[33] R. Hackl, P. Müller, D. Einzel, and W. Glaser, Physica C **162-164**, 1241 (1989).

[34] Of course anisotropic pairings and in general higher order L pairings of electrons would lead to anisotropic Raman data. However, impurities are expected to strongly modify the order parameter in this case. This is beyond the scope of this paper.