Electronic Raman Scattering as a Probe of Anisotropic Electron Pairing
T. P. Devereaux

Received 5 January 1995

A theory for the electronic contribution to Raman scattering in anisotropic superconductors is presented. It is shown that Raman scattering can provide a wealth of polarization- (symmetry-) dependent information which probes the detailed angular dependence of the energy gap. Using a model band structure, the symmetry-dependent Raman spectra are calculated for \( d_{x^2-y^2} \) pairing and compared to the data taken on \( \text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8 \). Favorable agreement with the symmetry-dependent electronic spectra is shown. Further, the impurity dependence of theory is calculated, which provides an unique test of \( d_{x^2-y^2} \) pairing.

KEY WORDS: Raman scattering, polarization symmetry, \( d- \) wave, impurity scattering.

1. INTRODUCTION

The electronic Raman spectra of the cuprates in the superconducting phase shows a remarkable dependence on the incident and scattered photon polarization vectors[1]-[6]. This polarization dependence, which is consistently seen in all the cuprates regardless of the presence of chains and the number of \( \text{CuO}_2 \) planes, has been recently interpreted as evidence for an order parameter of predominantly \( d_{x^2-y^2} \) symmetry[7]. The key to the polarization dependence lies in the coupling of the screened Raman vertex \( \delta \gamma(k) = \gamma(k) - \langle \gamma(k) \rangle \), and the energy gap \( \Delta(k) \). The topology of the nodes of each of the functions leads to a polarization dependence which not only affects the peak positions for each polarization orientation, but also the behavior of the spectra for small frequency shifts. Defining the imaginary part of the Tsubota function as

\[
\chi''(k, \omega) = \Theta(\omega^2 - 4 \left| \Delta(k) \right|^2 \tanh(\omega/4T)) \times \frac{\left| \Delta(k) \right|^2}{\sqrt{\omega^2 - 4 \left| \Delta(k) \right|^2}},
\]

the screened Raman spectra can then be written as

\[
\chi''(\omega) \sim \left( \left| \delta \gamma(k) \right|^2 \lambda(k, \omega) \right)'' - \frac{\left( \left( \delta \gamma(k) \lambda(k, \omega) \right)^2 \right)''}{\lambda(k, \omega)}.
\]

where \( \omega \) denotes taking the imaginary part. The real parts of the Tsubota function can be obtained from the imaginary part via a Kramers Kronig transformation. The second term of Eq. (2) takes into account the complete screening of intercell charge fluctuations so that only anisotropic charge fluctuations (intracell) remain. This is the expression for the gauge invariant Raman response which has Coulomb screening and the Anderson–Bogoliubov gauge mode taken into account. It does not take into account any vertex corrections resulting from the pairing interaction in other channels other than the pairing channel. The derivation and consequences of the above expression have been put forth in Ref. [7].

2. RAMAN VERTEX

The polarization dependence enters through the coupling of \( \delta \gamma(k) \) and the energy gap \( \Delta(k) \) under the Fermi surface averaging, and leads to subsequently different frequency dependent Raman spectra for each polarization channel. For non-resonant scattering, the Raman vertex can be expressed in terms of the curvature of the energy band dispersion \( \epsilon(k) \),

\[
\gamma(k) = m \sum_{\alpha, \beta} \epsilon_{\alpha} \frac{\partial^2 \epsilon(k)}{\partial k_{\alpha} \partial k_{\beta}} \epsilon_{\beta}^*,
\]

where \( \epsilon_{\alpha}^S \) denote the scattered and incident polarization light vectors, respectively, which select elements of the Raman tensor. Thus under the average, the contribution to Raman scattering is due to both
(1) a large density of states, and (2) large curvature of the band near the Fermi level. We parameterize our band structure via the standard $t - t'$ model with lattice constant $a$,

$$\epsilon(\mathbf{k}) = -2t [\cos(k_x a) + \cos(k_y a)] + 4t' \cos(k_x a) \cos(k_y a).$$  

(4)

Here $t$ and $t'$ are the nearest and next nearest neighbor hopping parameters, respectively. This is the anti-bonding band derived from a reduction of a three band model which gives the largest contribution to the density of states at the Fermi level for the cuprate systems and adequately reproduces the observed photoemission data[8].

Recently it has been suggested that the presence of another band, due to, e.g., another CuO$_2$ plane as in Y 1:2:3, can lead to substantially different results than the case for a single band[9]. This is in conflict with experiments, since the details of the number of planes and chains do not seem to affect the observed spectra[1] - [6]. This has been shown theoretically in Ref. [10], which considers multiband scattering of two planes, the response can be constructed from the real space wavefunctions in each plane, labelled by $| 1 \rangle$ and $| 2 \rangle$, for planes 1 and 2, respectively. Diagonalizing the Hamiltonian, we arrived at the ± bands, defined as $| + \rangle = \sqrt{2} (| 1 \rangle + | 2 \rangle)$ and $| - \rangle = \sqrt{2} (| 1 \rangle - | 2 \rangle)$. For the case of no wavefunction overlap between plane 1 and 2, the combined response can be written as

$$\chi_{11} + \chi_{22} = \frac{1}{2} [\chi_{+,+} - \chi_{-,+} - \chi_{+,+} + \chi_{-,+}]$$

$$\chi_{12} + \chi_{21} = \frac{1}{2} [\chi_{+,+} + \chi_{-,+} + \chi_{+,+} - \chi_{-,+}]$$

$$= \chi_{-,+} + \chi_{+,+}. \quad (5)$$

and the response is simply additive. If an overlap exists between planes 1 and 2, then $\chi_{+,+}$ must be kept. Nevertheless, it’s prefactor is smaller than the single layer terms by the amplitude for creating an electron-hole pair on different planes, for instance via an intermediate state such as the bridging oxygens. This term depends on the difference of vertices for bands + and −, e.g.[10],

$$\chi_{+,+} \sim (\gamma_+ - \gamma_-)^2. \quad (6)$$

Since the bands $\{+,-\}$ are nearly degenerate for the double layer compounds, again this leads to a small mixing term. Thus the full response is well approximated by the addition of the intraband scattering in each band, in agreement with experiments.

3. CALCULATIONS

Using a gap, $\Delta(\mathbf{k}, T) = 0.5 \Delta_0(T) [\cos(k_x a) - \cos(k_y a)]$, of $d_{x^2-y^2}$ symmetry, we evaluated the averages in Eq. (2) numerically using a Fermi surface given by the band structure, Eq. (4). Our results for the channel dependent spectra are given in Fig. (1), using the parameters $2t'/t = 1.3$ and $\mu/2t = -0.5$ appropriate for dopings which produce the highest $T_c$ values[11]. We have also performed the calculations for a Fermi surface which is more square-like ($\mu/2t = -0.2, t' = 0.1$) and obtain qualitatively the same results shown in Fig. (1). We immediately see that the spectra is extremely polarization dependent, in contrast to the case of isotropic $s$-wave superconductors which is dominated by the square root divergence at the threshold in each channel. We see that the peak in the Raman spectra lies at different frequencies $\omega_{\text{peak}} \sim 2 \Delta_0(T), 1.5 \Delta_0(T)$ and $1.25 \Delta_0(T)$ for the $B_{1g}, B_{2g}$ and $A_{1g}$ channels, respectively. The symmetry dependence is also manifest in the low frequency behavior. The spectra rise slower in the $B_{1g}$ channel ($\sim \omega^3$) than the $A_{1g}$ or $B_{2g}$ channels ($\sim \omega$). The power-laws are insensitive to vertex corrections and arise solely due to topology arguments. The channel dependence of the exponents are unique to a $d_{x^2-y^2}$ pair state. The symmetry dependence of the spectra is a direct consequence of the angular averaging which couples the gap and Raman vertex, and leads to constructive (destructive) interference under averaging if the vertex and the gap have the same (different) symmetry. Thus it has been reasoned that the symmetry which shows the highest peak position gives an unique indication of the predominant symmetry of the gap [7]. These channel-dependent power laws and relative peak positions have been observed in the electronic contribution to Raman scattering in Bi 2:2:1:2 [7, 3], Y 1:2:3 [1], TI 2:2:n-1:a (for $n = 1, 2, 3$) [4, 6, 5], and Sr doped La 2:1:4[2], and are strong evidence for a $d$-wave gap of this symmetry as opposed to $d_{xy}, d_{xz}$ or $d_{yz}$ symmetry, which also have nodes on lines on the Fermi surface [7]. The overall height scales with the hopping parameter $t$ for the $B_{1g}$ and $A_{1g}$ channels, and with $t'$ for the $B_{2g}$ channel. Similar results earlier obtained via a truncated Fermi surface harmonic expansion[7] validate the as-