Two-band model of Raman scattering on electron-doped high-$T_c$ superconductor

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We have analyzed the $B_{1g}$ and $B_{2g}$ Raman spectra of electron-doped cuprate superconductors Nd$_{2-x}$Ce$_x$CuO$_4$ and Pr$_{2-x}$Ce$_x$CuO$_4$ using a weakly coupled two-band model. One of these two bands is centered around $(\pm \pi/2, \pm \pi/2)$ and couples more strongly with the $B_{2g}$ mode, while the other is centered around $(\pm \pi, 0)$ and $(0, \pm \pi)$ and couples more strongly with the $B_{1g}$ mode. This model explains in a natural way why the $B_{2g}$ Raman peak occurs at a higher frequency than the $B_{1g}$ one at optimal doping, and how these two peaks change with doping in agreement with experiments. The result thus supports that there are two kinds of quasiparticles in electron-doped cuprates and $d_{x^2-y^2}$-wave superconductivity is driven by the holelike band and a proximity effect on the electronlike band.

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I. INTRODUCTION

Pairing symmetry of electron-doped high-$T_c$ cuprate superconductors such as Nd$_{2-x}$Ce$_x$CuO$_4$ and Pr$_{2-x}$Ce$_x$CuO$_4$ is a long standing problem. Although no consensus has been reached yet, more and more recent experimental results have suggested that the order parameter of electron-doped cuprates is likely to have $d_{x^2-y^2}$-wave pairing symmetry, in close resemblance to that of hole-doped materials. Among various experiments, the Raman scattering seems to suggest a different story.

For hole-doped superconductors, it was known that typical $B_{1g}$, $B_{2g}$, and $A_{1g}$ pair-breaking peaks appear, respectively, at the frequencies of 2, 1.6, and 1.2 times of the gap amplitude. However, in electron-doped materials the relative position of the $B_{1g}$ and $B_{2g}$ peaks changes with doping. The $B_{2g}$ peak appears first at a higher frequency than the $B_{1g}$ one in the underdoped regime. It then moves down and finally appears at a frequency lower than that of the $B_{1g}$ peak in the heavily overdoped regime.

The Raman scattering has the potential to probe different regions of the Fermi surface (FS), thus a thorough understanding of the experimental data can provide a better understanding on the momentum dependence of superconducting (SC) pairing gap. The observation of $B_{2g}$ Raman peak at higher frequency than that of $B_{1g}$ would imply a non-monotonic $d_{x^2-y^2}$-wave order parameter in a single-band system. This nonmonotonic order parameter seems to be also consistent with the observation of angle resolved photoemission spectroscopy (ARPES). However, this one-band picture may not be adequate to describe the nature of two kinds of charge carriers in electron-doped cuprate superconductors, as revealed by magneto-transport measurements.

A key clue towards the understanding of Raman data in Nd$_{2-x}$Ce$_x$CuO$_4$ comes from the doping evolution of the FS revealed by ARPES. At low doping, four small FS pockets first appear around $(\pm \pi, 0)$ and $(0, \pm \pi)$.

II. MODEL AND FORMALISM

We start by considering the two-dimensional $t$-$t'$-$t''$-$J$ model

$$H = -t \sum_{\langle ij \rangle \sigma} c_{i \sigma}^\dagger c_{j \sigma} - t' \sum_{\langle ij \rangle \sigma} c_{i \sigma}^\dagger c_{j \sigma} - t'' \sum_{\langle ij \rangle \sigma} c_{i \sigma}^\dagger c_{j \sigma} + J \sum_{\langle ij \rangle} \left( \vec{S}_i \cdot \vec{S}_j - \frac{1}{4} n_i n_j \right),$$

where $\langle ij \rangle_1$, $\langle ij \rangle_2$, and $\langle ij \rangle_3$ denote the nearest, second-nearest, and third-nearest neighbors between $i$ and $j$. No
double occupied sites are allowed in Eq. (1). All notations used in (11) are standard. Applying the slave-boson and MF decoupling (22), Hamiltonian (11) can be written in terms of two (diagonalized) bands in momentum space

\[ H = \sum_{k\sigma} \varepsilon_k \alpha_k \alpha_k + \xi_{k\sigma} \beta_k \beta_k, \]

where the prime denotes that the momentum summation is over the MBZ only \((-\pi < k_x \pm k_y \leq \pi)\) and

\[ \xi_{k,l} = \frac{\varepsilon_k + \varepsilon_k + Q_2}{2} = \sqrt{4f^2m^2} + 4J^2m^2 \]

with \(Q \equiv (\pi, \pi)\) the AF wave vector and \(m \equiv (-1)^i(S_i^z)\) the AF order. Here

\[ \varepsilon_k = (2|t|\delta - J\chi)(\cos k_x + \cos k_y) - 4t\delta \cos k_x \cos k_y - 2t\delta(\cos 2k_x + \cos 2k_y) \quad (2) \]

with \(\chi = f_{i\sigma} f_{j\sigma}\) the uniform bond order and \(\delta\) the doping concentration \((f_{i\sigma}\) is the fermionic spinon operator).

In the SC state, we add a BCS coupling term to each band and assume the system to be described by the following Hamiltonian (21)

\[ H = \sum_{k\sigma} \varepsilon_k \alpha_k \alpha_k + \sum_{l\sigma} \Delta_{k,l} (\alpha_k \alpha_k + \beta_{k,l} \beta_{k,l}), \]

where \(l \equiv \alpha, \beta\) and \(\Delta_{k,l} = (\Delta_l/2) [\cos k_x - \cos k_y]\) are the \(d_{x^2-y^2}\)-wave gap functions.

The Raman scattering intensity is proportional to the imaginary part of the effective density-density correlation function \(\chi(q, \tau) = \langle T_\tau \hat{\rho}(q, \tau) \hat{\rho}(-q, 0) \rangle\) in the limit \(q \to 0\). Here \(\hat{\rho}(q, \tau) = \sum_{k,\sigma} \gamma_k \hat{c}_{k+q,\sigma}(\tau) \hat{c}_{k,\sigma}(\tau)\) is the effective density operator and \(\gamma_k\) is the Raman vertex. When the energy of incident light is smaller than the optical band gap, the contribution from the resonance channel is negligible. The Raman vertex can then be obtained in terms of the curvature of the band dispersion under the inverse effective mass approximation.

In the current two-band model, the effective density operator is decomposed as

\[ \hat{\rho}(q, \tau) = \sum_{k,\sigma} \gamma_k \delta_{k+q,\sigma}(\tau) \hat{c}_{k,\sigma}(\tau) + \gamma_{k+Q} \delta_{k+Q,\sigma}(\tau) \hat{c}_{k+Q,\sigma}(\tau), \quad (3) \]

where \(\gamma_k\) and \(\gamma_{k+Q}\) are transformed into \(\gamma_k\) and \(\beta_{k+Q}\), the Raman response function for each symmetric channel (S) is then given by (22)

\[ \chi^S(q \to 0, \tau) = -\sum_{k,l,\ell} \langle \gamma^S_{kl,\ell} \rangle [G_{l}(k, \tau) G_{l'}(k, -\tau) - \delta_{l,\ell} F_{l}(k, \tau) F_{l'}(k, -\tau)], \quad (4) \]

where \(G\) and \(F\) are the normal and anomalous Green functions for a superconductor, \(\epsilon_{l\ell} = 1\) if \(l = l'\) or \(-1\) if \(l \neq l'\). The intra- and interband vertex functions are

\[ \gamma_{k,\alpha\alpha}^S = \cos^2 \theta_k \gamma_{k,\alpha\alpha}^S + \sin^2 \theta_k \gamma_{k+Q,\alpha\alpha}^S, \]

\[ \gamma_{k,\beta\beta}^S = \sin^2 \theta_k \gamma_{k,\beta\beta}^S + \cos^2 \theta_k \gamma_{k+Q,\beta\beta}^S, \]

\[ \gamma_{k,\alpha\beta}^S = \sin \theta_k \gamma_{k,\alpha\beta}^S \quad \gamma_{k,\beta\alpha}^S = \sin \theta_k \gamma_{k,\beta\alpha}^S \]

(5)

where \(\cos 2\theta_k = (\varepsilon_k+Q - \varepsilon_k)/\sqrt{4f^2m^2 + 4J^2m^2}\). For the \(B_{1g}\) and \(B_{2g}\) channels, \(\gamma_{B_{1g}} = \gamma_{B_{1g}} - \gamma_{B_{2g}}\) and \(\gamma_{B_{2g}} = 2x_{1g}\). Here \(\gamma_{ij} = \partial^2 \varepsilon_k/\partial k_k \partial k_j\) (inverse effective mass approximation). Eq. (3) and (4) reduce to the famous ones in a one-band system when \(m = 0\) (18).

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**FIG. 1:** Schematic plot of the Fermi surfaces for the \(\alpha\) and \(\beta\) bands and the momentum dependence of the \(B_{1g}\) and \(B_{2g}\) Raman vertices. The \(B_{1g}\) and \(B_{2g}\) modes couple more strongly with the \(\alpha\) and \(\beta\) bands, respectively.

Shown in Fig. 1 are the \(B_{2g}\) and \(B_{1g}\) Raman vertices and how they are coupled to the SC quasiparticle excitations in \(k\) space. Since the \(B_{2g}\) vertex has \(d_{x^2-y^2}\) symmetry, the \(B_{2g}\) channel is dominated by the excitations of the \(\beta\) band. In contrast, the \(B_{1g}\) vertex has \(d_{x^2-y^2}\) symmetry, thus the \(B_{1g}\) channel is contributed mainly from the excitations of the \(\alpha\) band. Since both \(B_{2g}\) and \(B_{1g}\) vertices are odd-parity, their Raman intensities are not affected by the Coulomb screening. For the fully symmetric \(A_{1g}\) channel, in contrast, all regions of momentum space contribute and the Raman intensity is partially screened. Since the \(A_{1g}\) channel is more sensitive to the actual vertex as well as the screening effect, we will leave out \(A_{1g}\) and focus on the \(B_{1g}\) and \(B_{1g}\) channels only.

In electron doped cuprates, the SC state appears only when the \(\beta\) band emerges above the Fermi energy. This would suggest that we assume that it is the \(\beta\) band that drives the system to superconduct, while the \(\alpha\) band becomes superconducting mainly via the proximity effect. A simple picture for the understanding of the unusual Raman spectra in electron doped cuprates can then be
sketched as follows. In the under doped or optimally doped regime, the $\beta$ band couples more strongly with the AF fluctuations than the $\alpha$ band. This results in a relatively larger SC gap in the $\beta$ band ($\Delta_\beta$) than in the $\alpha$ band ($\Delta_\alpha$). The $B_{1g}$ channel probes mainly the quasiparticle (QP) excitations of the $\alpha$ band, thus the $B_{1g}$ Raman peak is mainly determined by $\Delta_\alpha$. Similarly, the $B_{2g}$ channel probes mainly the QP excitations of the $\beta$ band, its Raman peak is mainly determined by $\Delta_\beta$. If $\Delta_\alpha$ is much smaller than $\Delta_\beta$, one would then expect that the $B_{1g}$ peak appears at a frequency lower than that of the $B_{2g}$ peak, unlike in the hole doped case. In the heavily overdoped regime, the AF correlation becomes very weak and the band splitting vanishes. In this case the two-band model reduces essentially to a one-band model and the band splitting vanishes. This then suppresses the ratio between the $m$ band, its Raman peak is mainly determined by $\Delta_\beta$. If $\Delta_\alpha$ is about 1 particle (QP) excitations of the $\alpha$ band (∆α) and the $\alpha$ band (∆β) peak will be red shifted (blue shifted) compared with the result of hole-doped cuprate superconductors.

III. RESULTS AND DISCUSSIONS

A. On Nd$_{2-x}$Ce$_x$CuO$_4$

Pertaining to Nd$_{2-x}$Ce$_x$CuO$_4$, we have adopted $t'/t = 0.32$, $t'' = 0.3t$, $t'' = -0.2t$, and $j = 3t$ to simulate the band structure [22]. We take $\chi = -0.15$ and $m = 0.178$ for the optimally doped ($x = 0.15$) sample, $\chi = -0.15$ and $m = 0.15$ for the overdoped ($x = 0.16$) sample. These parameters are close to those obtained in self-consistent calculations for the normal states [22]. The chemical potentials are determined by the filling factor for each band to give the true doping concentration through $x = n_e - n_h$.

Theoretical fitting procedures are implemented as follows. First, the vertex functions are evaluated using [19]. Then the SC gaps $\Delta_\alpha$ and $\Delta_\beta$ and the smearing Lorentz width $\Gamma_\alpha$ and $\Gamma_\beta$ are adjusted to fit the peak positions and the overall spectral line shape (up to a constant multiplying factor).

Figure 2 compares the experimental data of Raman spectra for Nd$_{2-x}$Ce$_x$CuO$_4$ with the theoretical calculations. For all the cases considered in Fig. 2 our fitting curves are in good agreement with the experimental results. For the optimally doped sample ($x=0.15$) reported in Ref. [14] at $T = 8 K$ (first column), the $B_{1g}$ and $B_{2g}$ Raman peaks appear at 50 cm$^{-1}$ and 55 cm$^{-1}$, respectively. The corresponding gap and smearing parameters obtained by fitting are ($\Delta_\alpha, \Delta_\beta$) = (21 cm$^{-1}$, 48 cm$^{-1}$) and ($\Gamma_\alpha, \Gamma_\beta$) = (6 cm$^{-1}$, 8 cm$^{-1}$). The ratio between the $B_{1g}$ Raman peak frequency and $\Delta_\alpha$ is about 2.4, while the ratio between the $B_{2g}$ Raman peak frequency and $\Delta_\beta$ is about 1.2. The corresponding ratios in a hole doped d$_{x^2-y^2}$-wave superconductor are about 2 and 1.6, respectively. This difference between hole and electron doped cuprates is not difficult to be understood. In electron doped materials, the AF correlation splits the continuous FS into two separate sheets. This then suppresses the high (low) energy region to which the $B_{2g}$ ($B_{1g}$) probes. It is thus expected that the $B_{2g}$ ($B_{1g}$) peak will be red shifted (blue shifted) compared with the result of hole-doped cuprate superconductors.

![Figure 2](image-url)

**TABLE I**: Summary of fitting parameters for Nd$_{2-x}$Ce$_x$CuO$_4$ (refer to Fig. 2).

|        | Column 1 | Column 2 | Column 3 |
|--------|----------|----------|----------|
| $T_c$ (K) | 22       | 22       | 13       |
| $m$      | 0.178    | 0.178    | 0.15     |
| $\Gamma_\alpha$(cm$^{-1}$) | 6        | 4        | 8        |
| $\Gamma_\beta$(cm$^{-1}$) | 8        | 6        | 8        |
| $\Delta_\alpha$(cm$^{-1}$) | 21       | 27       | 21       |
| $\Delta_\beta$(cm$^{-1}$) | 48       | 57       | 37       |
| $\Delta_\beta/\Delta_\alpha$ | 2.29     | 2.11     | 1.76     |
| $\Delta_\beta/T_c$ | 2.18     | 2.59     | 2.84     |

For another set of data of the optimally doped sample reported in Ref. [14] at $T = 4 K$ (second column), the fitting parameters are ($\Delta_\alpha, \Delta_\beta$) = (27 cm$^{-1}$, 57 cm$^{-1}$) and ($\Gamma_\alpha, \Gamma_\beta$) = (4 cm$^{-1}$, 6 cm$^{-1}$). The corresponding parameters for the overdoped sample at $T = 4 K$ (third column) are ($\Delta_\alpha, \Delta_\beta$) = (21 cm$^{-1}$, 37 cm$^{-1}$) and $\Gamma_\alpha = \Gamma_\beta = 8$ cm$^{-1}$. The gap parameters obtained are consistent with the general expectation. Both $\Delta_\alpha$ and $\Delta_\beta$ decrease with increasing temperature at the same doping level and with increasing doping at the same temperature. The gap ratio, $r = \Delta_\beta/\Delta_\alpha$, is reduced from 2 at optimal doping to 1.7 at slightly overdoping, consistent with the scenario of AF-like fluctuation induced superconductivity. All fitting parameters for Nd$_{2-x}$Ce$_x$CuO$_4$ are summarized in Table I.
TABLE II: Summary of fitting parameters for Pr$_{2-x}$Ce$_x$CuO$_4$ (refer to Fig. 3).

|       | Column 1 | Column 2 | Column 3 |
|-------|----------|----------|----------|
| $x$   | 0.15     | 0.165    | 0.18     |
| $T_c$ (K) | 23.5    | 15       | 10       |
| $m$   | 0.15     | 0.12     | 0        |
| $\Gamma_\alpha$ (cm$^{-1}$) | 6        | 8        | 15       |
| $\Gamma_\beta$ (cm$^{-1}$) | 6        | 8        | 15       |
| $\Delta_\alpha$ (cm$^{-1}$) | 31       | 16       | 15       |
| $\Delta_\beta$ (cm$^{-1}$) | 68       | 30       | 15       |
| $\Delta_\beta/\Delta_\alpha$ | 2.19     | 1.88     | 1        |
| $\Delta_\beta/T_c$ | 2.89     | 2        | 1.5      |

B. On Pr$_{2-x}$Ce$_x$CuO$_4$

The Raman scattering measurement has also been carried out in electron-doped Pr$_{2-x}$Ce$_x$CuO$_4$ at various doping levels [15]. The Raman spectra of Pr$_{2-x}$Ce$_x$CuO$_4$, as shown in Fig. 3, behave similarly as for Nd$_{2-x}$Ce$_x$CuO$_4$. At optimal doping, the $B_{2g}$ peak appears at a frequency higher than that of the $B_{1g}$ peak. With increasing doping, the frequency of the $B_{1g}$ peak approaches to and finally surpasses the $B_{2g}$ peak in the overdoped regime.

FIG. 3: Comparison between the theoretical results (solid curves) and the Raman measurement data from Ref. 20 (circles) for Pr$_{2-x}$Ce$_x$CuO$_4$ at $T = 4K$. $\omega_L = 1.9eV$.

Figure 3 compares the measurement data of Pr$_{2-x}$Ce$_x$CuO$_4$ with our theoretical calculations. For the optimally doped sample ($x = 0.15$), the Raman peak appears at 80 cm$^{-1}$ for the $B_{2g}$ mode and at 62 cm$^{-1}$ for the $B_{1g}$ mode. The parameters obtained by fitting are $(\Delta_\alpha, \Delta_\beta) = (31 \text{ cm}^{-1}, 68 \text{ cm}^{-1})$ and $\Gamma_\alpha = \Gamma_\beta = 6 \text{ cm}^{-1}$, with $\chi = -0.15$ and $m = 0.15$. For the slightly overdoped sample ($x = 0.165$), the $B_{2g}$ and $B_{1g}$ peaks appear at the same frequency at 37 cm$^{-1}$ and the parameters we obtained are $(\Delta_\alpha, \Delta_\beta) = (16 \text{ cm}^{-1}, 30 \text{ cm}^{-1})$ and $\Gamma_\alpha = \Gamma_\beta = 8 \text{ cm}^{-1}$, with $\chi = -0.15$ and $m = 0.12$. For the heavily over-doped sample ($x = 0.18$), the Raman peaks appear at 25 cm$^{-1}$ and 30 cm$^{-1}$ for the $B_{2g}$ and $B_{1g}$ modes, respectively. The relative peak positions of these two modes are similar as in a one-band $d_{x^2-y^2}$-wave superconductor. This is not unexpected since at such a high doping level, the two-band model reduces essentially to a one-band model. In this case, the parameters we obtained are $\Delta_\alpha = \Delta_\beta = 15 \text{ cm}^{-1}$ and $\Gamma_\alpha = \Gamma_\beta = 15 \text{ cm}^{-1}$, with $\chi = -0.15$ and $m = 0$. For the above three samples, the ratio $\Delta_\beta/\Delta_\alpha$ changes from 2.3, to 1.9, and finally to 1 with increasing doping. With increasing doping, the AF order is depressed and the gap amplitudes is decreased. The results are consistent with neutron scattering measurement [30]. All fitting parameters for Pr$_{2-x}$Ce$_x$CuO$_4$ are summarized in TABLE II.

In the top-left panel of Fig. 3 for the $B_{2g}$ mode, the experimental data are taken under the strong resonant regime as emphasized in Refs. 14, 15. In this case, the contribution from the resonance channel becomes important and the inverse effective mass approximation is not valid. Our theoretical result including only the non-resonant contribution can give a good explanation to the low-frequency part of the spectrum, but the height of the peak is much lower than the experimental one. The resonance channel may also have some contribution to the $B_{2g}$ spectrum in the second column of Fig. 2 for Nd$_{2-x}$Ce$_x$CuO$_4$.

IV. SUMMARY

In summary, we have analyzed the Raman spectra of electron-doped cuprate superconductors based on a weakly coupled two-band model. Our result gives a unified explanation to the experimental data in the whole doping range. It suggests strongly that the SC pairing in electron-doped cuprate superconductors results from the same pairing mechanism as in hole doped ones and the inverse effective mass approximation is not valid. Our theoretical result including only the non-resonant contribution can give a good explanation to the low-frequency part of the spectrum, but the height of the peak is much lower than the experimental one. The resonance channel may also have some contribution to the $B_{2g}$ spectrum in the second column of Fig. 2 for Nd$_{2-x}$Ce$_x$CuO$_4$.

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