Superconducting pairing and the pseudogap in the nematic dynamical stripe phase of La\(_{2-x}\)Sr\(_x\)CuO\(_4\)

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Abstract

Fully absorption coefficient corrected Raman spectra were obtained in La\(_{2-x}\)Sr\(_x\)CuO\(_4\). The \(B_{1g}\) spectra have a Fleury–Loudon type two-magnon peak (resonant term) whose energy decreases from 3180 cm\(^{-1}\) (394 meV) to 440 cm\(^{-1}\) (55 meV) on increasing the carrier density from \(x = 0\) to 0.25, while the \(B_{2g}\) spectra have a 1000–3500 cm\(^{-1}\) (124–434 meV) hump (hill) whose lower-edge energy increases from \(x = 0\) to 0.115 and then stays constant to \(x = 0.25\). The \(B_{2g}\) hump is assigned to the electronic scattering (non-resonant term) of the spectral function with magnetic self-energy. The completely different carrier density dependence arises from anisotropic magnetic excitations of spin–charge stripes. The \(B_{1g}\) spectra were assigned to the sum of \(k \parallel\) and \(k \perp\) stripe excitations and the \(B_{2g}\) spectra to \(k \perp\) stripe excitations according to the calculation by Seibold and Lorenzana (2006 Phys. Rev. B 73 144515). The \(k \parallel\) and \(k \perp\) stripe excitations in fluctuating spin–charge stripes were separately detected for the first time. The appearance of only \(k \perp\) stripe excitations in the electronic scattering arises from the charge hopping perpendicular to the stripe. This is the same direction as the Burgers vector of the edge dislocation in metal. The successive charge hopping in the Burgers vector direction across the charge stripes may cause Cooper pairs as predicted by Zaanen \textit{et al} (2004 Ann. Phys. 310 181). Indeed, this is supported by the experimental fact that the superconducting coherent length coincides with the inter-charge stripe distance in the wide carrier density range. The one-directional charge hopping perpendicular to the stripe causes the flat Fermi surface and the pseudogap near \((\pi, 0)\) and \((0, \pi)\), but the states around \((\pi/2, \pi/2)\) cannot be produced. The low-energy Raman scattering disclosed that the electronic states at the Fermi arc around \((\pi/2, \pi/2)\) are coupled to the \(A_{1g}\) soft phonon of the tetragonal–orthorhombic phase transition. This suggests that the Fermi arc is produced by the electron–phonon interaction. All the present Raman data suggest that Cooper pairs are formed at moving edge dislocations of dynamical charge stripes.

(Some figures may appear in colour only in the online journal)

1. Introduction

Soon after the discovery of the high temperature superconductor, an incommensurate spin modulation was found by neutron scattering \cite{1, 2}. Stabilities of the
superconducting states in spin stripes and spin plaquettes were investigated [3–10]. The inhomogeneous structures were expected to solve the question of why the two-dimensional Hubbard model may not stabilize the superconducting state [11–13]. A periodic lattice modulation was found in La$_2$−$x$Sr$_x$CuO$_4$ (LSCO) by EXAFS [14, 15] and the atomic pair distribution function analysis of neutron diffraction [16]. Tranquada et al found the spin–charge stripe structure in superconductivity suppressed La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$ (LNSCO) by neutron scattering [17]. Neutron scattering could not detect the charge density, so that the charge modulation was supposed from the lattice modulation. The charge modulation was certified by resonant soft x-ray scattering (RSXS) [18]. Yamada’s group disclosed that the stripe structure in LSCO was ubiquitous in the doped insulating and superconducting phase. It is active in the B$_{1g}$ spectra, if only nearest neighbor exchange interactions are taken into account [71]. The quantum spin fluctuation and far-site spin interactions make the A$_{1g}$ and B$_{2g}$ channels Raman active [67, 68, 72, 73]. The A$_{2g}$ symmetry becomes Raman active only if the time reversal symmetry is broken by the chiral spin interaction [67, 68, 73, 74]. The experimentally observed A$_{2g}$ intensity is 9% and the B$_{2g}$ is 16% of the B$_{1g}$ intensity in Gd$_2$CuO$_4$ [69]. Therefore the A$_{2g}$ and B$_{2g}$ terms can be ignored in the first approximation.

The most important discovery of this paper is the development of the hump structure from 1000 to 3500 cm$^{-1}$ (124–434 meV) in the B$_{1g}$ and B$_{2g}$ spectra, as temperature decreases from 300 to 5 K in the doped phase. The energy and intensity systematically change with the carrier density. The hump structure cannot be interpreted in the non-magnetic electronic excitations [67, 68, 75–84]. The carrier density dependent energy can be explained by the separated dispersion segments of the k $\perp$ stripe spin wave dispersion calculated by Seibold and Lorenzana [54, 56]. Therefore the Raman process is not the transitions in the electronic energy states, but the successive magnon excitations by hole hopping in the stripe state. This may be treated in the string model [85–92], but it has not been explicitly applied to the electronic Raman scattering. It should be noticed that the mean free path of a hole is very short $k_F l = 0.1$ [93], so that the most probable hopping steps are two sites emitting two magnons. Thus Fleury–Loudon type two-magnon scattering is active in the B$_{1g}$ channel and the successive magnon excitation by hole hopping is active in both B$_{1g}$ and B$_{2g}$ channels. The former process gives k $\parallel$ and k $\perp$ stripe excitations, while the latter process gives only k $\perp$ stripe excitations. Utilizing the polarization selection rule we succeeded in observing the individual k $\parallel$ and k $\perp$ stripe magnetic excitations in fluctuating stripes.

Many experimental results of Raman scattering have been reported with respect to the high temperature superconductivity. The superconducting gaps in hole-doped superconductors were investigated by low-energy Raman scattering [74, 83, 94–118]. Two-magnon scattering and high-energy electronic scattering were investigated by wide-energy Raman scattering [71–74, 82, 83, 95, 97, 98, 100, 105, 111, 119–124]. None of them distinguished the Fleury–Loudon type two-magnon scattering and the magnon excitations by hole hopping in electronic scattering. The carrier density dependence of the B$_{2g}$ spectra is scarcely reported, because the Fleury–Loudon type two-magnon scattering is active dominantly in the B$_{1g}$ spectra. Our observed hump structure from 1000 to 3500 cm$^{-1}$ (124–434 meV) has not been reported so far. The reason is the sensitivity of the
crystal surface. The mechanically polished surface loses this structure. Instead the defect-induced peak at 4500 cm\(^{-1}\) (558 meV) and high-energy luminescence appears. The justification of our spectra is proved by the proportionality relation of the generalized sum rule between the imaginary part of the electronic Raman susceptibility \(\chi''(\omega)\) and the optical conductivity \([67, 77, 125, 126]\). The Fleury–Loudon type two-magnon scattering is not correlated. It is presented separately [127].

The magnetic excitations created by the hole hopping have only \(k \perp\) stripe excitations. This means that the charge transfer is restricted only to the \(k \perp\) stripe direction. This surprising result is reminiscent of the Burgers vector of an edge dislocation in metal [128]. The edge dislocation and the screw dislocation easily slide and cause ductility in metal. In a two-dimensional layer only the edge dislocation is available. The edge dislocation slides in the Burgers vector direction which is perpendicular to the inserted stripe. Charges at the edge of a stripe move together with the edge dislocation and other charges are localized, because \(k \parallel\) stripe excitation is not observed in the \(B_{2g}\) electronic scattering. A looped (bridged) edge dislocation connecting two charge stripes has lower energy than the single edge dislocation [6], because the spin alignments on both sides of the charge stripe have opposite phase [17]. Zaanen [6, 9] proposed a superconducting model generated by bosonized charges at edge dislocations. The spin–charge separation is not observed experimentally. Therefore it is supposed that Cooper pairs are formed at the moving edge dislocations. This model is supported by the experimental fact that the superconducting coherence length [129, 130] is close to the inter-charge stripe distance \(d\) [19] at \(x < 0.2\). The coherence length is only twice the inter-charge distance on the assumption that charges are uniformly distributed. The superconducting state is in the crossover regime between BCS (Bardeen–Cooper–Schrieffer) and BEC (Bose–Einstein condensation) [131, 132].

The one-dimensional sliding motion of the charged edge dislocation in the charge stripe parallel to \(b\) makes the flat Fermi surface perpendicular to the \(a\) axis near \((0, \pi)\) and the pseudogap near \((\pi, 0)\) in the underdoped phase. In the same way the charge stripe parallel to \(a\) makes the flat Fermi surface perpendicular to the \(b\) axis near \((\pi, 0)\) and the pseudogap near \((0, \pi)\). The reduction of the pseudogap with increasing carrier density is naturally explained by the one-dimensional charge transfer. However, the Fermi surface near \((\pi/2, \pi/2)\) cannot be formed. We found that the \(B_{2g}\) spectra below 180 cm\(^{-1}\) (22 meV) are enhanced as temperature decreases. The enhanced part is composed of a Fano-like step structure of the orthorhombic \(A_g\) phonons folded from \((\pi, \pi)\). It reveals that the electronic states near \((\pi/2, \pi/2)\) are coupled states with \((\pi, \pi)\) phonons. The spin density wave/charge density wave (SDW/CDW) gap and the superconducting gap appear in these states. The \(B_{2g}\) superconducting gap energy is fixed by the phonon energy and not dependent on the \(T_c\).

The process to reach the superconducting mechanism from Raman scattering is the following. (1) The discovery of the hump structure from 1000 to 3500 cm\(^{-1}\) (124–434 meV) in the \(B_{2g}\) spectra. (2) The assignment of the hump to the separated dispersion segment in the \(k \perp\) stripe direction. (3) Introduction of a new Raman scattering mechanism for the magnon excitations by the hole hopping. (4) The similarity of the hole hopping perpendicular to the stripe and the sliding motion of the edge dislocation in the Vergers vector direction. (5) The coincidence of the inter-charge stripe distance to the superconducting coherence length. (6) The one-dimensional hole hopping can explain the pseudogap and the deformation of the Fermi surface. Therefore it is concluded that the holes in the looped (bridged) edge dislocation form a Cooper pair. The outline is listed in table 1.

The difference between electronic Raman scattering with non-magnetic and magnetic excitations and Fleury–Loudon type two-magnon Raman scattering are summarized in section 2. The wide-energy Raman scattering, the analysis with respect to the anisotropy and isotropy in \(k\) space, \(k \parallel\) and \(k \perp\) stripe excitations, and the low-energy Raman scattering are presented in section 3. The pairing at the looped edge dislocations is proposed in section 4. The one-dimensional sliding motion is applied to the pseudogap in section 5. Discussions are given in section 6. The conclusion is presented in section 7.

2. Non-magnetic and magnetic Raman scattering mechanisms

2.1. Electronic Raman scattering

Electronic Raman scattering in simple metal is caused by the first order perturbation of the \(A^2\) term and the second order of the \(p \cdot A\) term in the electron–radiation interaction term \((\mathbf{P} - \frac{e}{c}\mathbf{A})^2\). The matrix element is given by \([133, 134]\)

\[
M = \varepsilon_i^\alpha \varepsilon_j^\beta \frac{1}{m} \left[ \delta_{\alpha\beta} + \frac{1}{m} \right] \times \left( \sum_b \frac{|\langle a, k_b P^\dagger | b, k_i + q|^\alpha| b, k_i + q|^\beta|^P^\alpha |a, k_b\rangle|}{\varepsilon_{a, k_b} - \varepsilon_{b, k_i + q} + \hbar \omega_{\alpha\beta}} + X \right),
\]

(1)

where \(X\) is the term with the different time order, \(m\) is the free electron mass, \(\varepsilon_i^\alpha\) and \(\varepsilon_j^\beta\) are polarization vectors of incident and scattered light, \(\alpha\) and \(\beta\) are the Cartesian coordinates, \(\omega_{\alpha\beta}\) is the incident photon energy and wavevector, \(a\) and \(b\) the initial and intermediate electronic states, and \(k_l\) and \(k_0\) the initial and final wavevectors of the electron. In the low energy and long wavelength approximation of the incident light, equation (1) has the same form as the \(\mathbf{k} \cdot \mathbf{p}\) perturbation. Hence equation (1) becomes

\[
M \approx \varepsilon_i^\alpha \varepsilon_j^\beta \frac{\partial^2 \epsilon(\mathbf{k})}{\partial k_\alpha \partial k_\beta} \approx \varepsilon_i^\alpha \varepsilon_j^\beta \left( \frac{1}{m^*} \right)_{\alpha\beta},
\]

(2)

where \((1/m^*)\) is the effective inverse mass tensor. The energy range of the Raman spectra is limited to less than a few tens cm\(^{-1}\) due to the momentum conservation with light. The scattering intensity goes to zero as the momentum shift \(q\) goes to zero.
Table 1. Outline.

| Symmetry | Wide-energy Raman scattering | Low-energy Raman scattering |
|----------|-----------------------------|-----------------------------|
| B₁g      | Fleury–Loudon type two-magnon + electronic scattering | Scattering of electronic states with the magnetic self-energy k \perp stripe |
| B₂g      | Scattering of electronic states with the magnetic self-energy k \perp stripe | Soft phonon of the tetra–ortho phase transition |
| Experiment | Charge transfer \perp stripe | Correlation between the soft mode and electronic states |
| Electronic states | \perp The same direction as the Burgers vector of the edge dislocation | Electron–phonon coupled states |
| Superconducting mechanism | Stripe-induced SC | |
| Coherence length = inter-charge stripe distance | (B₁g: pseudogap around (\pi, 0) and (0, \pi)) | Fermi arc around (\pi/2, \pi/2) |

The Raman intensity is proportional to [135]

\[
\left| \left\langle \mathbf{e}_1 \cdot \left( \frac{1}{m} \right) \cdot \mathbf{e}_2 \right\rangle \right|^2_{F} - \left| \left\langle \mathbf{e}_1 \cdot \left( \frac{1}{m} \right) \cdot \mathbf{e}_3 \right\rangle \right|^2_{F},
\]

where \( \langle \cdot \rangle_{F} \) represents an average over the Fermi surface. The second term represents the screening of the A₁g spectra by plasma excitations. The A₁g intensity is fully screened, if the mass tensor is the same along the Fermi surface. The B₁g and B₂g spectra are not screened, because the second term is zero. In the strongly correlated electron system, the incoherent part loses the k dependence and the symmetry dependence. As a result the B₁g and B₂g spectra become identical and the A₁g spectra are screened.

In the strongly correlated electron system, the upper and lower Hubbard bands of the Cu 3dₓ²−ᵧ² level are taken into account. In the Hubbard model coupled with light, the creation and annihilation operators of an electron develop as [67, 68]

\[
e^{-\frac{i}{\hbar c} \int \mathbf{A} \cdot d\mathbf{l}}.
\]

The interaction Hamiltonian between the Hubbard electron and an electromagnetic wave is expanded to the second order in \( A \) [67, 68]:

\[
H_{\text{int}} = -\left( \frac{e}{\hbar c} \right) \sum_{\mathbf{k}} \mathbf{j}(\mathbf{k}) \cdot \mathbf{A}(-\mathbf{k}) + \frac{e^2}{2\hbar c^2} \sum_{\mathbf{k}, \mathbf{k}'} \mathbf{A}(-\mathbf{k}) \tau_{\alpha, \beta}(\mathbf{k} + \mathbf{k}') \mathbf{A}(-\mathbf{k}'),
\]

where the current operator is

\[
\mathbf{j}_\alpha(\mathbf{q}) = \sum_{\mathbf{k}} \frac{\partial \mathbf{\epsilon}(\mathbf{k})}{\partial k_\alpha} c_\alpha^\dagger(\mathbf{k} + \mathbf{q}/2)c_\sigma(\mathbf{k} - \mathbf{q}/2),
\]

and the stress tensor is

\[
\tau_{\alpha, \beta}(\mathbf{q}) = \sum_{\mathbf{k}} \frac{\partial^2 \mathbf{\epsilon}(\mathbf{k})}{\partial k_\alpha \partial k_\beta} c_\alpha^\dagger(\mathbf{k} + \mathbf{q}/2)c_\sigma(\mathbf{k} - \mathbf{q}/2).
\]

The Raman matrix element of the nonresonant term is given by the first order perturbation of \( \tau \),

\[
\langle f|M_{\alpha, \beta}^R(\mathbf{q})|i\rangle = \langle f|\tau_{\alpha, \beta}(\mathbf{q})|i\rangle,
\]

and the resonant term is given by the second order perturbation of \( j \),

\[
\langle f|M_{\alpha, \beta}^R(\mathbf{q})|i\rangle = \sum_{\nu} \left( \frac{\langle f|\mathbf{j}(\mathbf{k})\rangle|v|\mathbf{j}(\mathbf{\nu} - \mathbf{k})\rangle|i\rangle}{\epsilon_v - \epsilon_i - \hbar \Omega_\alpha} + \frac{\langle f|\mathbf{j}(\mathbf{\nu} - \mathbf{k})\rangle|v|\mathbf{j}(\mathbf{k})\rangle|i\rangle}{\epsilon_v - \epsilon_i + \hbar \Omega_\ell} \right).
\]

An electron transferred to a neighboring site is excited to the upper Hubbard band in the intermediate state. The charge transfer excitation energy with double occupancy is close to the incident photon energy, so that the scattering is resonantly enhanced.

In the single-site dynamical mean field theory the \( k \) dependence is ignored [136]. The imaginary part of the Raman susceptibility of the nonresonant term is given by [77, 78, 80]

\[
\chi''(\omega) = \int \text{d}\epsilon V(\epsilon) \int \text{d}\omega' A(\epsilon, \omega')
\times A(\epsilon, \omega' + \omega)[f(\omega') - f(\omega' + \omega)],
\]

where

\[
\langle f|M_{\alpha, \beta}^R(\mathbf{q})|i\rangle = \sum_{\nu} \left( \frac{\langle f|\mathbf{j}(\mathbf{k})\rangle|v|\mathbf{j}(\mathbf{\nu} - \mathbf{k})\rangle|i\rangle}{\epsilon_v - \epsilon_i - \hbar \Omega_\alpha} + \frac{\langle f|\mathbf{j}(\mathbf{\nu} - \mathbf{k})\rangle|v|\mathbf{j}(\mathbf{k})\rangle|i\rangle}{\epsilon_v - \epsilon_i + \hbar \Omega_\ell} \right).
\]
angle-resolved photoemission spectroscopy (ARPES) \[137\].

\[\epsilon = \text{tight binding model:} \]

\[t \underset{\text{goes to zero, while the incoherent parts keep the intensity.}}{\text{where}}\]

scattering intensity from the coherent peak goes to zero as \(q\) peak (quasi-particle peak) and two incoherent parts. The other particles.

6 6 \[\text{where} \]

t 6 neighbor hopping integrals between Cu sites. The parameters

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\[\text{Figure 1. (a) The electron energy dispersion in La}_{1.875}\text{Sr}_0.125\text{CuO}_4 using the parameters reported by ARPES [137]. The contour maps of}\]

\[\left|\frac{\partial^2 \epsilon(k)}{\partial k_a \partial k_b}\right| \text{ for (b) } A_{1g}, \text{(c) } B_{1g}, \text{ and (d) } B_{2g}. \text{ The solid line shows the Fermi arc and the dashed line shows the depleted Fermi surface (pseudogap).}\]

where the form factor \(V(\epsilon)\) is

\[V(\epsilon) = \sum_k \left[ \frac{\partial^2 \epsilon(k)}{\partial k_a \partial k_b} \right]^2 \delta[\epsilon - \epsilon(k)], \quad (11)\]

where \(ij = xy\) for \(B_{1g}\) and \(ij = ab\) for \(B_{2g}\). \(f(\omega)\) is the Fermi–Dirac distribution function. The one-particle spectral function \(A\) is the imaginary part of the Green function:

\[A(\epsilon, \omega) = -\frac{1}{\pi} \text{Im} [\frac{1}{\omega + \mu - \Sigma(\omega) - \epsilon}], \quad (12)\]

where \(\Sigma\) is the self-energy representing the interactions with other particles. \(\Sigma\) is independent of \(k\) in the dynamical mean field theory. The spectral function is composed of a coherent peak (quasi-particle peak) and two incoherent parts. The scattering intensity from the coherent peak goes to zero as \(q\) goes to zero, while the incoherent parts keep the intensity.

Figure 1(a) shows the electron energy dispersion of the tight binding model:

\[\epsilon(k_a, k_b) = \epsilon_0 - 2t'(\cos k_a + \cos k_b) - 4t'' \cos k_a \cos k_b - 2t''t'\cos k_a \cos k_b, \quad (13)\]

where \(t, t'\), and \(t''\) are the first-, second-, and third-nearest neighbor hopping integrals between Cu sites. The parameters are \(t = 0.25 \text{ eV}, t' = -0.17t (-0.15t, -0.12t), t'' = -t'/2\), and \(\epsilon_0 = 0.55t \(0.81t, 0.99t\) for \(x = 0.07 \(0.15, 0.3\) by angle-resolved photoemission spectroscopy (ARPES) [137].

The light blue plane in figure 1(a) shows the chemical potential \(\mu = 0\). Figures 1(b)-(d) show \[|\frac{\partial^2 \epsilon(k)}{\partial k_a \partial k_b}|^2 \] for \(A_{1g}\), \(B_{1g}\), and \(B_{2g}\) at \(x = 0.07\). The \(B_{1g}\) intensity is given by \(|t'(\cos k_a - \cos k_b) + 4t''(\cos 2k_a - \cos 2k_b)|^2\) and the \(B_{2g}\) intensity by \(4t'(\sin k_a \sin k_b)^2\). The \(B_{1g}\) spectra are observed near \((\pi, 0)\) and the \(B_{2g}\) spectra are observed near \((\pi/2, \pi/2)\) [75, 76]. The intensity near \((\pi, 0)\) and \((0, \pi)\) in \(B_{1g}\) is much larger than that near \((\pi/2, \pi/2)\) in \(B_{2g}\), because \(|t|\) is much larger than \(|t'|\). The Fermi surface at \(x = 0.07\) is shown by the thick line and the dashed line. The dashed line indicates the pseudogap formed in the underdoped phase [137].

In the dynamical mean field theory the difference between the \(B_{1g}\) and \(B_{2g}\) spectra comes from the \(V(\epsilon)\) in equation (11). Figure 2 shows the \(V(\epsilon)\) for (a) the \(A_{1g}\), (b) \(B_{1g}\) and (c) \(B_{2g}\) symmetries, and (d) the density of states. The chemical potential is of energy zero. In order to obtain the Raman susceptibility of equation (10), the self-energy has to be calculated. This is far beyond the scope of the present work. What we can say is that the \(A_{1g}\) intensity increases with increasing the energy shift, while the \(B_{1g}\) and \(B_{2g}\) spectra decreases. The present experiment, however, revealed that the intensity of the \(A_{1g}\) spectra decreases more rapidly than the \(B_{1g}\) and \(B_{2g}\) spectra at high energies, indicating that the screening effect increases at high energies. The peak positions
in $A_{1g}$ and $B_{1g}$ shift from $\epsilon < 0$ to $\epsilon > 0$ in figure 2, because the zone boundary point of the Fermi surface changes from the $(0, \pi) - (\pi, \pi)$ to the $(0, 0) - (0, \pi)$ side in a quarter of the Brillouin zone.

The height of the $B_{2g}$ peak is about $1/40$ of the $B_{1g}$ peak. The $B_{2g}$ intensity may come from the resonant term, but the intensity is much smaller than the $A_{1g}$ and $B_{1g}$ channels. However, the present experiment revealed that the $B_{2g}$ intensity is the same order as the $B_{1g}$ intensity in the underdoped phase.

### 2.2. Magnetic excitations in electronic Raman scattering

The electronic Raman scattering process (nonresonant term) does not directly include magnetic excitations [67, 68, 75–83]. However, magnetic excitations emerge if hole hopping overturns spins. This is the key mechanism to understand the present Raman spectra of the hump structure from 1000 to 3500 cm$^{-1}$ (124–434 meV). The hole hopping from site A to the nearest neighbor site B is the same as a back hopping of an electron from B to A in figure 3(a). The electron spin is opposite to the stable spin direction at site A. Thus hole hopping causes the overturned spin trace shown in the lower panel. The red wavy lines show the increased energy bonds.

The overturned spin excitation propagates as a magnon at each hopping from A to B and from B to C. This process is treated by the string model [85–92]. If the mean free path is long, a hole sees a linearly rising potential as the migration distance increases in the limit of $t \gg J$. The energy levels become discrete:

$$E_n/t = -b + a_n(J/t)^{2/3},$$

where $a_n$ are the eigen values of a dimensionless Airy equation and $b = 3.28$ at $(\pi/2, \pi/2)$. The discrete peak height decreases with increasing $n$ and connects to a continuum at high energies. However, in hole-doped superconductors the mean free path $l$ is very short, $k_F l \approx 0.1$ [93]. Then the near-site hopping is dominant. From the present experiment two-site hopping gives the most probable magnetic excitation spectra. The magnetic excitations are presented in electronic Raman scattering through the self-energy $\Sigma$ in equation (12). The non-magnetic excitations are dominant in the low-energy part, but the magnetic excitations become dominant above 1000–2000 cm$^{-1}$ (124–248 meV).

### 2.3. Fleury–Loudon type two-magnon scattering

Fleury–Loudon type two-magnon scattering in the insulating phase is caused by the resonant term of equation (9). An electron at A hops to the neighboring site B by absorbing light and the original electron at B hops to A by emitting light in figure 3(b) [67, 68]. This process gives the same interaction Hamiltonian as the Fleury–Loudon type [65, 66]:

$$H_{\text{two-mag}} = \sum_{kl} A(e_i \cdot r_{kl})(e_i \cdot r_{kl})(S_k \cdot S_l),$$

where $r_{kl}$ is the unit vector connecting the $k$ and $l$ sites. In the case of classical spin interaction between nearest neighbors, the two-magnon scattering is active in $(aa)$ and $(xy)$ and inactive in $(ab)$. In $(xx)$ the two-magnon scattering Hamiltonian is the same as the system Hamiltonian $H = \sum_{kl} S_k \cdot S_l$ except for the proportionality constant. Then
two-magnon scattering is inactive, because the two-magnon Hamiltonian commutes to the system Hamiltonian. Thus the two-magnon scattering is active in the $B_{1g}$ channel.

Quantum spin fluctuation and far-site exchange interactions give the $A_{1g}$ and $B_{2g}$ Raman activity as far as time reversal symmetry is satisfied as in two-site exchange interactions [67, 68]. If time reversal symmetry is broken by the chiral spin interaction $\mathbf{S}_i \cdot (\mathbf{S}_j \times \mathbf{S}_k)$, the $A_{2g}$ symmetry becomes Raman active [67, 68]. The $A_{2g}$ spectra are observed in the $(a, b)$, $(x, y)$, and $(R, R)$ polarization configurations, where $R$ is right-handed circularly polarized light. The spectral intensity for each symmetry in $La_2CuO_4$ was reported [74]. The integrated intensities of the $A_{1g}$, $A_{2g}$, and $B_{2g}$ spectra below 8000 cm$^{-1}$ (992 meV) relative to $B_{1g}$ are 100, 25, and 35% at 21 K, respectively. The $A_{1g}$ spectra include higher-order resonant phonon scattering. The estimated ratio for $A_{2g}$ is too high, because the 4500 cm$^{-1}$ (558 meV) peak is the defect peak induced by mechanical polishing and the high-energy spectra seem to include luminescence. The high-energy intensity is also reduced to about 1/2 times by the absorption correction as discussed in section 3.1. Then the $A_{2g}$ intensity is less than 10% of the $B_{1g}$ intensity. The $A_{1g}$, $A_{2g}$, and $B_{2g}$ intensities in Gd$_2$CuO$_4$ are 60, 9, and 16% of the $B_{1g}$ intensity, respectively [69]. The $A_{2g}$ intensity is small. The chiral spin interaction was searched in neutron scattering, because it plays an important role in some high temperature superconducting mechanisms [138–141]. However, strong evidence for the chiral spin interactions was not obtained. Therefore we analyze the Raman spectra within the time reversal symmetry.

Two magnons are simultaneously excited, so that the two magnons interfere and the total energy is reduced from the independently excited two magnons by the magnon–magnon interaction energy which is close to the exchange interaction energy $J$ [65, 66, 70]. The magnon–magnon interaction does not arise in the electronic scattering process in figure 3(a), if a magnon is excited at each hopping process. The symmetry dependence of the magnetic Raman scattering mechanism is summarized in table 2.

### 3. Raman scattering experiments

#### 3.1. Experimental procedure

A triple-grating spectrometer with the same focusing lengths of 600 mm was used. In order to obtain the wide-energy spectra, we carefully adjusted the spectrometer every 3–4 months. The focusing of the laser spot on the crystal surface and on the spectrometer slit was monitored by a camera mounted in the spectrometer. The sample position was restored using the camera at each temperature, because it moved when the temperature changed.

Single crystals were synthesized by a traveling-solvent floating-zone method. The solvent was melted by the radiation from four halogen lamps with elliptic mirrors. The excess oxygen in $La_2CuO_4$ crystals was reduced, but some excess oxygen remained. The oxygen is deficient in as-grown crystals of $x = 0.2$ and 0.25. They were annealed in one atm oxygen gas at 600 ℃ for 7 days. Raman spectra sensitively change by the surface treatment. Mechanical polishing causes a defect-induced peak at $4J \approx 4500$ cm$^{-1}$ (558 meV) and strong luminescence at high energies. The hump structure from 1000 to 3500 cm$^{-1}$ which develops in the $B_{1g}$ and $B_{2g}$ spectra at low temperatures is suppressed instead. The probability to obtain a good surface by cleavage was small, so we prepared many rectangular parallelepiped samples which were cut in the crystallographic axes. The sample was set in a cryostat within three minutes, if a good surface was obtained by the cleavage. The spectra changed if the sample was exposed in air for three hours after the cleavage. The sample was cooled down to 5 K in one hour and took the data from low temperature. Raman scattering was measured in a quasi-back scattering configuration using 514.5 nm laser light. The incident angle from the normal direction of the sample surface was 30°. The incident polarization direction was fixed to the horizontal direction (p-wave). The vertical or horizontal polarization of scattered light was selected. The $A_{1g}$, $B_{1g}$, and $B_{2g}$ symmetry excitations are separately obtained by the linearly polarized light combinations in the approximation that the interactions which break the time reversal symmetry are weak as discussed in section 2.3. The $B_{1g}$ and $B_{2g}$ spectra were obtained in the $(x, y)$ and $(a, b)$ polarizations, respectively. The $A_{1g}$ spectra were obtained from the calculation of the spectra $(x, x) + (a, a) - (x, y) - (a, b)/2$. The $B_{1g}$ and $B_{2g}$ spectra were obtained by rotating the sample keeping other optical geometries in the same positions.

The wavenumber and polarization dependences of the optical system were carefully corrected using reflected light from an optical standard white plate. The light source is an incandescent lamp with a known black body radiation temperature. The optical path for the measurement of the spectral efficiency was carefully adjusted to coincide with the Raman scattering experiment. The Raman cross section per unit volume is [142]

$$\frac{\partial^2 \sigma}{\partial \Omega \partial \omega} \propto \frac{\omega_i}{\omega} \left[ n(\omega, T) + 1 \right] \chi''(\omega, T),$$

where $\Omega$ is a solid angle, $\omega = \omega_i - \omega_s$, and $n(\omega, T)$ is the Bose–Einstein statistical factor. The $\omega_s/\omega_i$ term comes from the conversion between the scattered photon energy and the number. The back scattering intensity from the opaque material is

$$I_{\text{obs}} \propto \frac{\partial^2 \sigma}{\partial \Omega \partial \omega} \int_0^\infty e^{-\sigma(\omega_i)z} \sigma(\omega) dz \propto \frac{\omega_i}{\omega} \frac{[n(\omega, T) + 1]}{[\sigma(\omega_i) + \sigma(\omega)]} \chi''(\omega, T),$$
where $\alpha$ is the absorption coefficient. The observed intensity is inversely proportional to the sum of absorption coefficients for incident and scattered light. The reflection at the surface is ignored, because it is 11–19% in the used energy range. All the Raman spectra in this paper are plotted in $\chi''(\omega, T)$.

In order to obtain the absorption coefficient, near-normal incident in-plane ($e \perp c$) reflectivity spectra were measured using a vacuum-type Fourier-transform spectrometer (0.004–1.6 eV) and a grating spectrometer (0.8–6.6 eV). The reflection spectra were measured on the mechanically polished surface at 295 K. The spectra were connected to the high-energy data above 6 eV [143]. The optical conductivity was obtained by the Kramers–Kronig transformation assuming the Hagen–Rubens formula for the low-energy extrapolation to the DC conductivity. The details of the infrared spectroscopy were presented in our previous paper [144]. Figure 4 shows the optical conductivity at 295 K. The 12 000 cm$^{-1}$ (149 meV) peak which increases at high carrier density is larger than the reported one [143]. Figure 5 shows the absorption coefficient. The lower scale is the energy difference from the laser wavenumber. The upper scale is the absolute wavenumber. In the underdoped regime the absorption coefficient is corrected or not. The polished surface has no hump structure from 1000 to 3500 cm$^{-1}$ (124–434 meV) which develops as temperature decreases from 300 to 5 K. It is clearly observed in the $B_{2g}$ spectra, because the 300 K spectra are almost featureless. The hump structure is indicated by the downward triangles. The lowest energy 2 increases as the carrier density increases from $x = 0.035$ to 0.115 and then keeps constant above $x = 0.115$.

The high-energy spectra are quite different from other groups [74, 82, 83]. The difference comes from whether the crystal surface is cleaved or polished and whether the absorption coefficient is corrected or not. The polished surface has no hump structure from 1000 to 3500 cm$^{-1}$. Instead it has a defect-induced peak at 4500 cm$^{-1}$ (558 meV) and luminescence at high energies. The luminescence is stronger for the incident wavelength of 457.9 nm than for 514.5 nm. That our spectra are intrinsic is verified by comparing the Raman spectra to the optical conductivity. Shastry and Shraiman [67] pointed out that the imaginary part of the wide-energy Raman susceptibility $\chi''(\omega)$ and the optical conductivity $\sigma(\omega)$ is connected by $\chi''(\omega) \propto \omega \sigma(\omega)$, because it is the incoherent part of the spectral function. Freericks and Devereaux et al [77, 125] showed the sum rule $\int_0^\infty \omega \chi''(\omega) d\omega \equiv \int_0^\infty \omega^2 \sigma(\omega) d\omega$ in the dynamical mean field theory. The present $B_{1g}$ and $B_{2g}$ spectra have the relation

$$\int_0^\Omega \omega \chi''(\omega) d\omega \propto \int_0^\Omega \omega^p \sigma(\omega) d\omega \tag{18}$$

as a function of the cutoff frequency $\Omega$ if the Fleury–Loudon type two-magnon component in the $B_{1g}$ spectra is removed. The exponent $p$ is 1.3 for the $B_{1g}$ spectra and 1.5 for the $B_{2g}$ spectra at $x = 0.15$. It is presented separately.

Two-magnon excitations were also observed in O K edge RIXS [145]. The energy is around 450 meV independently of the carrier density from $x = 0$ to 0.22. The energy is higher than the $B_{2g}$ hump energy.

3.2. Wide-energy spectra: $k \parallel$ and $k \perp$ stripe excitations

Figure 6 shows the wide-energy Raman spectra. All the spectra are plotted in the same intensity scale. The sharp peaks from 700 to 1400 cm$^{-1}$ (87–174 meV) at $x = 0$ are two-phonon peaks. Four- and six-phonon peaks are observed in the $A_{1g}$ and $B_{2g}$ spectra. The multi-phonon spectra are 20 times stronger in the $A_{1g}$ spectra than in the $B_{1g}$ or $B_{2g}$ spectra at $x = 0$. The multi-phonon intensity rapidly decreases to 1/60 at $x = 0.035$ and almost completely disappears at $x \geq 0.08$ in the $B_{1g}$ and $B_{2g}$ spectra, while the small intensity remains in the whole carrier density range in the $A_{1g}$ spectra.

Figure 4. In-plane optical conductivity in LSCO at 295 K.

Figure 5. Absorption coefficient in LSCO at 295 K. The lower scale is the energy shift from 19 435 cm$^{-1}$ (514.5 nm) laser light.
Figure 6. Wide-energy (a) A\textsubscript{1g}, (b) B\textsubscript{1g} and (c) B\textsubscript{2g} Raman spectra. The downward triangles correspond to the dispersion segments with the same number and color (blue, green and red) as in the \( k_\parallel \) stripe magnetic excitations in figure 7.

We analyze the B\textsubscript{1g} and B\textsubscript{2g} spectra, because the high-energy part of the A\textsubscript{1g} spectra is strongly screened. The smooth B\textsubscript{2g} spectra at 300 K in figure 6(c) may be interpreted by the electronic Raman scattering theory with strong correlation [77, 78, 80, 82, 146]. However, the hump which develops from 1000 to 3500 cm\(^{-1}\) as temperature decreases cannot be interpreted by the above models. The hump is isotropic and the energy depends on the carrier density. The enhancement of the hump on cooling is largest at \( x = 0.035 \) and smallest at \( x = 1/8 \) in figure 6(c). The ‘hour-glass’ like magnetic susceptibility observed in neutron scattering is mainly analyzed by the dynamical stripes with mixed directions [49–56] or the interacting fermion liquid [57–60]. We analyze the Raman spectra by individual magnetic excitations for the \( k_\parallel \) and \( k_\perp \) stripe directions calculated by Seibold and Lorenzana [54, 56].

Figures 7(a) and (b) show the \( \omega \chi''(\omega, q) \) for \( k_\parallel \) and \( k_\perp \) stripe in the metallic vertical bond-centered stripe (VBC) phase calculated by Seibold and Lorenzana [54], respectively. Here \( \chi''(\omega, q) \) is the imaginary part of the transverse magnetic susceptibility. The intensity representation is simplified from the original contour map [54]. The blue, green, and red curves represent the dispersions at \( x = 0.06 \) (\( d = 8 \)), 0.08 (\( d = 6 \)), and \( x = 0.125 \) (\( d = 4 \)), respectively, where \( d = \pi/\delta \) is the stripe width (inter-charge stripe distance) in the unit of Cu–Cu distance. In the \( k_\parallel \) stripe of figure 7(a) the dispersion energy rapidly decreases as well as the decrease of the high-energy intensity with increasing the carrier density. On the other hand in the \( k_\perp \) stripe of figure 7(b) the dispersion curve is separated into \( d \) segments because of the Brillouin zone folding. The highest energy at \( (0, \pi) \) decreases little with increasing the carrier density. The energy of each dispersion segment increases with increasing the carrier density from \( x = 0.06 \) to 0.125, because the number of segments decreases. The separated dispersion has a large energy gap between the first and second dispersion segments. The black line shows the uniform spin wave dispersion along the \( a \) or \( b \) axis at \( x = 0 \) with the nearest and the next nearest neighbor exchange interaction energies \( J = 840 \, \text{cm}^{-1} \) (104 meV) and \( J' = -145 \, \text{cm}^{-1} \) (−18 meV) [147].

The B\textsubscript{1g} Fleury–Loudon type two-magnon peak energy in figure 6(b) decreases with increasing the carrier density in the same way as the \( k_\parallel \) stripe magnetic excitations in figure 7(a).

The B\textsubscript{2g} hump in figure 6(c) indicated by the downward triangles shifts from 900 to 3500 cm\(^{-1}\) at \( x = 0.06 \) to 1600–3500 cm\(^{-1}\) at \( x = 0.25 \). The triangles are numbered
so that the energies are about twice the energy of dispersion segments in figure 7(b). The higher energy segments are included in the high-energy part of the hump. The first dispersion segment may be included in the low-energy peak below 400 cm\(^{-1}\) (50 meV), because the low-energy peak is strongly enhanced at low temperatures in the same way as the 900–3500 cm\(^{-1}\) hump at \(x = 0.035\) and 0.06. The strong enhancement is, however, observed only in the B\(_{2g}\) spectra. It is different from the hump which is observed in both the B\(_{1g}\) and the B\(_{2g}\) spectra. The hump has the following properties. (1) The energy of triangle 2 increases with increasing the carrier density from \(x = 0.06\) to 0.115 and then becomes constant above \(x = 0.115\). (2) The hump develops as temperature decreases from 300 K to 5 K. (3) The hump is small near \(x = 1/8\). (4) The hump is large near the insulator–metal transition. (5) The same hump is observed in the B\(_{1g}\) spectra. The hump structure seems to appear in the B\(_{1g}\) spectra of the report by Machtoub et al [124] at 2200 and 3100 cm\(^{-1}\) (273–384 meV) at low temperatures, although there is no description.

Figure 8 shows the comparison between a half the energy of edge 2 in the B\(_{2g}\) spectra and the energy of the second dispersion segment in the \(k \perp\) stripe excitations in figure 7(b) [54]. The vertical bar is the energy width of the segment. In the metallic phase the energy 2 increases in accordance with the calculated energy of the second dispersion segment from \(x = 0.035\) to 0.115. Above \(x = 0.115\) the energy 2 remains constant, while the calculated energy keeps increasing. The incommensurability \(\delta\) obtained from neutron scattering [19] is shown by the dashed line in figure 8.

The \(\delta\) has the similar carrier density dependence to the energy 2 of the present experiment. The saturation above \(x = 1/8\) might be related to the recent Compton scattering that the excess hole orbital populates in Cu d\(_{3z^2−r^2}\) besides O p in the overdoped phase [148].

Thus we conclude that the B\(_{1g}\) spectra have the \(k ||\) and \(k \perp\) stripe excitations and the B\(_{2g}\) spectra have \(k \perp\) stripe excitations. The electronic scattering (nonresonant term) has only the \(k \perp\) stripe component. The results are summarized in table 2.

### 3.3. Wide-energy spectra: anisotropic or isotropic electronic dispersion in \(k\) space

The one-particle spectral function in the strongly correlated electron system has the coherent part which has the properties of the original particle and the incoherent part which is isotropic in \(k\) space and no symmetry dependence. In order to know whether the hump structure is isotropic or not and where the crossover region of coherent and incoherent parts is, the screening effect of the A\(_{1g}\) spectra and the equivalence between the B\(_{1g}\) and B\(_{2g}\) spectra are analyzed.

Figure 9 shows the carrier density dependence of the A\(_{1g}\) spectra at 5 K. The intensity rapidly decreases as the carrier density increases from \(x = 0\) to 0.1 and then the spectra keep the same shape at \(x \geq 0.1\). The spectra have a broad peak at 500 cm\(^{-1}\) (62 meV) and a long tail to high energy at \(x > 0.1\).

Figure 10 shows the comparison of the A\(_{1g}\), B\(_{1g}\), and B\(_{2g}\) spectra at 5 K. The B\(_{1g}\) and B\(_{2g}\) spectra approach each other as the energy shift increases and become the same above 4000 cm\(^{-1}\) at \(x = 0.035\), 2000 cm\(^{-1}\) at \(x = 0.1\), 4000 cm\(^{-1}\) at \(x = 0.15\), and 5000 cm\(^{-1}\) at \(x = 0.25\). This indicates that the anisotropy of the electron energy dispersion in \(k\) space decreases as the energy moves away from the chemical potential, that is, the energy dispersion becomes isotropic at high-energy shift. The unscreened A\(_{1g}\) intensity is expected to increase, because the form factor \(V(\epsilon)\) increases with increasing energy in figure 2. However, the A\(_{1g}\) spectra
Figure 9. Carrier density dependent wide-energy $A_{1g}$ spectra at 5 K.

Figure 10. Comparison of the $A_{1g}$, $B_{1g}$ and $B_{2g}$ spectra at 5 K.

Figure 11. Differential spectra between the $B_{1g}$ and $B_{2g}$ symmetries.

The intensity decreases much faster than the $B_{1g}$ and $B_{2g}$ spectra. This indicates that the scattering becomes isotropic at high energies and the screening increases.

Figure 11 shows the differential spectra between the $B_{1g}$ and $B_{2g}$ symmetries. The Fleury–Loudon type two-magnon peak at $x = 0$ is rather sharp, because the multi-phonon and electronic scattering components are removed. The two-magnon scattering is basically inactive in the $B_{2g}$ channel. At $x = 0.1$ the intensity above 2000 cm$^{-1}$ is zero, that is, the $B_{1g}$ and $B_{2g}$ spectra are the same. The $B_{1g}$ intensity decreases below 2000 cm$^{-1}$ due to the formation
of the pseudogap around $(\pi, 0)$ [137, 149–151]. A similar structure is observed from $x = 0.035$ to 0.115 if the Fleury–Loudon type two-magnon peak is removed. At $x = 0.115$ the differential spectra are the same as $x = 0.1$ from 300 to 100 K. At 5 K a weak hump at $2010 \text{ cm}^{-1}$ (249 meV) and a long high-energy tail emerges. The hump enlarges and the peak energy softens as the carrier density increases. In the overdoped phase the pseudogap closes and the B$_{1g}$ scattering intensity becomes increasingly large, as the carrier density increases. The intensity of the B$_{1g}$ spectra at $x = 0.25$ is 4.1 times the B$_{2g}$ spectra at $150 \text{ cm}^{-1}$ (19 meV) and 1.8 times for the integrated intensity from 16 to 6000 $\text{ cm}^{-1}$. Thus the electronic states are approaching the band model.

The Fleury–Loudon type two-magnon peak decreases in intensity and energy as $x$ increases from $x = 0$ to 0.08. The two-magnon peak energy at $x \leq 0.08$ continues to the hump energy at $x \geq 0.115$. The characteristic hump at 1000–3500 $\text{ cm}^{-1}$ does not appear in the differential spectra of figure 11, representing that the B$_{1g}$ spectra have the same hump as the B$_{2g}$ spectra at all temperatures.

The isotropic and anisotropic regions in the $k$ space obtained from the B$_{1g}$ and B$_{2g}$ spectra are shown in figure 12 on the assumption that the electronic properties are symmetric at the chemical potential. It is noted that the boundaries are continuous. The decrease of the B$_{1g}$ intensity below 2000 $\text{ cm}^{-1}$ in the underdoped phase is due to the opening of the pseudogap near $(\pi, 0)$ [137, 149–151]. In the overdoped phase the pseudogap closes and the B$_{1g}$ intensity becomes larger than the B$_{2g}$ intensity. The boundary roughly divides the coherent and incoherent parts of the spectral function.

It is noticed that the hump structure from 1000 to 3500 $\text{ cm}^{-1}$ which is assigned to the successive magnon excitations by the hole hopping (section 2.2) is isotropic, even if the energy is lower than the above anisotropy–isotropy boundary. It is quite different from the Fleury–Loudon type two-magnon peak which is observed only in the B$_{1g}$ spectra dominantly.

A similar phase diagram can be obtained from the A$_{1g}$ scattering. The isotropy increases as the energy goes away from the chemical potential similarly to figure 12. The A$_{1g}$ spectra have almost the same structure at $x \geq 0.1$ in figure 9, so that the boundary at $x = 0.15$ is missing. The isotropic momentum dependence is also observed in YBa$_2$Cu$_3$O$_{6.5}$ above 100 meV in neutron scattering [48].

Figure 13 shows the carrier density dependent (a) B$_{1g}$ and (b) B$_{2g}$ average scattering intensity from 16 to 7000 $\text{ cm}^{-1}$ (solid lines). The B$_{1g}$ intensity decreases from $x = 0$ to 0.1, because the Fleury–Loudon type two-magnon scattering intensity decreases. The electronic scattering intensity increases as the carrier density increases. The B$_{2g}$ scattering intensity increases from $x = 0$ to 0.06 and then gradually decreases with increasing the carrier density. The rather large average intensity at $x = 0$ is due to the natural hole doping of our sample. An example of small B$_{2g}$ intensity at $x = 0$ was reported [72]. The B$_{2g}$ average intensity has a small dip at $x = 1/8$ in figure 13(b). It is caused by less enhancement of the hump from 1000 to 3500 $\text{ cm}^{-1}$ at 1/8 as temperature decreases from 300 to 5 K. The dashed lines in figures 13(a) and (b) show the intensity at 150 $\text{ cm}^{-1}$ in the B$_{1g}$ spectra and 100 $\text{ cm}^{-1}$ in the B$_{2g}$ spectra, respectively. The average intensity of the B$_{2g}$ wide-energy spectra has similar carrier

![Phase diagram of electronic states with the isotropic or anisotropic $k$ dependence](image)

**Figure 12.** Phase diagram of electronic states with the isotropic or anisotropic $k$ dependence on the assumption that the state is symmetric at the chemical potential ($\epsilon = 0$). The anisotropy decreases as the energy moves away from the chemical potential and the state is smoothly connected to the isotropic state. Note that the boundary is smooth.

![Intensity vs. carrier density](image)

**Figure 13.** (a) The B$_{1g}$ low-energy intensity at 150 $\text{ cm}^{-1}$ and the average intensity from 16 to 7000 $\text{ cm}^{-1}$. (b) The B$_{2g}$ intensity at 100 $\text{ cm}^{-1}$ and the average intensity from 16 to 7000 $\text{ cm}^{-1}$. The differential spectra between 40 and 300 K at 2500 $\text{ cm}^{-1}$ are also shown.
density dependence to the low-energy intensity. The carrier density dependences of the low-energy $B_{1g}$ and $B_{2g}$ intensities are consistent with the ARPES intensities near $(\pi, 0)$ and $(\pi/2, \pi/2)$, respectively [137]. The fine structure is, however, different, as discussed in section 3.4.

Figure 14 shows the central energy of the $B_{1g}$ (solid line) and $B_{2g}$ (dashed line) spectra below 7000 cm$^{-1}$. The $B_{1g}$ central energy gradually decreases as the carrier density increases. The decrease is more moderate than that of the peak energy. The $B_{2g}$ central energy decreases from $x = 0$ to 0.125 and then increases.

3.4. Low-energy spectra: polaron and SDW/CDW gap

The low-energy spectra are different depending on the symmetry. The $B_{1g}$ spectra observe the antinodal gap near $(\pi, 0)$ and the $B_{2g}$ spectra observe the nodal gap near $(\pi/2, \pi/2)$ in accordance with the tight binding band model of equation (13) [75, 76]. The absorption coefficient corrected low-energy spectra are shown in figure 15. The insets show the contour maps of $|\partial^2 \epsilon(k)/\partial k_\alpha \partial \beta|^2$ in figure 1. The absorption uncorrected spectra were presented in the previous paper [113]. The $B_{1g}$ and $B_{2g}$ spectra are similar to other groups [74], but our spectra have finer structure because all the spectra were obtained on fresh cleaved surfaces.

The structural transition temperature from the tetragonal $I4/mmm$ to orthorhombic $Abma$ ($Cmca$ by changing the axes $(a, b, c) \rightarrow (c, a, b)$) phase decreases from 525 K at $x = 0$ to 10 K at $x = 0.21$ [152, 153]. The orthorhombic crystallographic axes $a$ and $b$ rotate by 45$^\circ$ from the tetragonal axes $x$ and $y$ and the unit cell volume doubles. The $(\pi, \pi)$ points in the tetragonal structure are folded into the $\Gamma$ point in the orthorhombic structure. The optical phonon modes are $2A_{1g} + 2E_g + 3A_{2u} + 2B_{2u} + 4E_u$ in the tetragonal structure and $5A_{g} + 3B_{1g} + 6B_{2g} + 4B_{3g} + 4A_{u} + 6B_{1u} + 4B_{2u} + 7B_{3u}$ in the orthorhombic structure. The $2A_{g}$ modes are the $2A_{1g}$ modes at $(0, 0)$ and the $3A_{g}$ modes come from the $3\Sigma$ modes.

Figure 15. (a) $A_{1g}$, (b) $B_{1g}$ and (c) $B_{2g}$ low-energy Raman spectra. The insets show the contour maps of $(1/m^*)^2$ for $A_{1g}$, $B_{1g}$ and $B_{2g}$.

![Figure 14](image-url) Center of the spectral weight below 7000 cm$^{-1}$.

![Figure 15](image-url) (a) $A_{1g}$, (b) $B_{1g}$ and (c) $B_{2g}$ low-energy Raman spectra. The insets show the contour maps of $(1/m^*)^2$ for $A_{1g}$, $B_{1g}$ and $B_{2g}$.
at $(\pi, \pi)$ in the tetragonal phase. The selection rule viewed from the tetragonal axes is listed in table 3.

The $A_{1g}$ and $B_{2g}$ modes are rapidly enhanced as carriers are doped, while the $B_{1g}$ spectra are weak. The $A_{1g}$ and $B_{2g}$ spectra below 180 cm$^{-1}$ (22 meV) are strongly enhanced as temperature decreases in the underdoped phase. The $A_{1g}$ and $B_{2g}$ intensities decrease in the overdoped phase and the $B_{1g}$ spectra become strong instead.

The $A_{1g}$ (in the tetragonal symmetry representation) spectra in figure 15 have many phonon peaks. Some of them are clearly assigned from the $(c, c)$ spectra, because the $A_g$ phonon peaks are sharp and strong in $(c, c)$ at $x = 0$. Two $A_g$ phonons at 229 and 426 cm$^{-1}$ (28 and 53 meV) are the tetragonal $A_{1g}$ modes at $(0, 0)$ [154–156]. The intensities in the in-plane polarization spectra are weak in figure 15(a). The other $A_g$ peaks in the $(c, c)$ spectra are 126, 156, and 273 cm$^{-1}$ (16, 19, and 34 meV) at 5 K. These modes are folded into $(0, 0)$ from $(\pi, \pi)$ in the tetragonal reciprocal lattice. The 126 cm$^{-1}$ peak energy decreases toward the orthorhombic–tetragonal transition temperature [154, 156]. Therefore this mode is the soft mode causing the transition.

The peaks which come from $(\pi, \pi)$ disappear at $x \geq 0.22$ [155, 156], because the orthorhombic structure ends at $x = 0.21$ and 10 K [153].

Figure 16 shows the $A_g$ spectra of $x = 0, 0.035, 0.06$ at 5 K and 0.1 from 5 to 300 K. The phonon energies of the $\Sigma$ branch at $(0, 0)$ and $(\pi, \pi)$ in neutron scattering and the $A_g$ phonon energies in the $(c, c)$ Raman spectra at $x = 0$ are shown. The upper black, red, orange, green triangles are $\Sigma_1 - \Sigma_4$ mode energies at room temperature, respectively [157–159]. The light blue triangles are $\Sigma_1$ modes at room temperature [160]. The blue triangles are $\Sigma$ modes [161, 162]. The lowest energy mode at $(\pi, \pi)$ softens toward the orthorhombic–tetragonal transition temperature. The purple rhombi show the $A_g$ mode in the $(c, c)$ Raman spectra at 5 K [154]. The open symbols are the $(0, 0)$ modes and the filled symbols are the $(\pi, \pi)$ modes in the tetragonal structure. The determination of the $A_g$ phonon modes from the $(c, c)$ Raman spectra is most reliable, because the scattering peak is sharp and the phonon symmetry is clearly separated.

The $A_1$ peak intensity increases from $x = 0.035$ to 0.06 and decreases from $x = 0.115$ to 0.15. The same kind of carrier density dependent intensity is observed in the low-energy $B_{1g}$ spectra below 150 cm$^{-1}$. The carrier density range is the increasing regime of the incommensurability $\delta$ [19]. It is supposed that the width of the spin–charge stripe is fluctuating and the charge density in the charge stripes is also fluctuating. The stripe width becomes constant above $x = 1/8$ and the fluctuation decreases. The $A_1$ peak and the low-energy $B_{1g}$ spectra are assigned to the excitations in fluctuating charge stripes [109].

### Table 3. Selection rule for phonon modes.

| Symmetry in $D_{4h}$ | $A_{1g}$ | $B_{1g}$ | $B_{2g}$ |
|----------------------|----------|----------|----------|
| Polarization in $D_{4h}$ | $(\sigma x + (aa) - (xy) - (ab))/2$ | $(xy)$ | $(ab)$ |
| Tetragonal ($D_{4h}$) | $2A_{1g}$ | 0 | 0 |
| Orthorhombic ($D_{2h}$) | $5A_{g}$ | $3B_{1g}$ | $5A_{g}$ |

The $A_2$ and $A_3$ peaks are the same as the $A_g$ peaks in the $(c, c)$ spectra. The $A_2$ peak is assigned to the zone boundary $(\pi, \pi)$ modes of the longitudinal acoustic mode ($\Sigma_1, 125$ cm$^{-1}$) and the transverse acoustic mode ($\Sigma_3, 136$ cm$^{-1}$). The $A_3$ peak is assigned to the $\Sigma_1$ mode of 148 cm$^{-1}$ at $(\pi, \pi)$ [160]. This branch is flat from 149 cm$^{-1}$ at $(0, 0)$ to 148 cm$^{-1}$ at $(\pi, \pi)$. The $A_4$ peak is not due to the $A_g$ mode. It becomes strong with doping. It is assigned to the $\Sigma$ branch at $(\pi, \pi)$ (177 cm$^{-1}$). The $A_2$ and $A_4$ peak intensities decrease faster than the $A_3$ peak, as temperature increases.

The $A_2$, $A_3$, and $A_4$ peaks are observed in infrared spectroscopy as $B_{3u}$ modes of the orthorhombic structure [163]. The orthorhombic crystal structure $Abma$ has inversion symmetry, so that the Raman and infrared activities are exclusive. The appearance in both spectra means the inversion symmetry breaking. The modes are not the simple phonons, but may be electron–phonon coupled modes. In fact the $A_g$ spectra are strongly enhanced at low temperatures when carriers are doped. The 126 ($A_2$) and 156 cm$^{-1}$ ($A_1$) peaks
in the \((c, c)\) spectra rapidly broaden as carriers are doped [155, 156]. The hump from 240 to 330 cm\(^{-1}\) is assigned to the second order of the A\(_2\), A\(_3\) and A\(_4\) peaks.

Zhou et al [164] observed multiple phonon spectra of about 140 cm\(^{-1}\) in the electron dispersion along \((0, 0) - (\pi, \pi)\) in ARPES of underdoped LSCO. The energy is just the same as the average energy of the peaks A\(_2\), A\(_3\), and A\(_4\). The energy resolution in ARPES is 12 and 20 meV, while that of Raman scattering is 0.7 meV. Therefore the Raman scattering presents the fine structure of the electron–phonon coupled modes. The difference is that the first order peaks are stronger than the second order peaks in Raman scattering, while the higher-order peaks are stronger than the first order peaks in ARPES [164]. The multiple phonon structure is included in the electron self-energy [165, 166].

The B\(_{2g}\) spectra at 300 K in figure 15(c) are strongly enhanced by the small carrier doping even in the insulating phase of \(x = 0.035\). The low-energy part below 180 cm\(^{-1}\) is further enhanced as temperature decreases at \(x \geq 0.035\). It makes the Fermi arc near \((\pi/2, \pi/2)\).

Figure 17 shows the comparison between the A\(_{1g}\) (black and blue), B\(_{1g}\) (dark green and green) and B\(_{2g}\) (red and orange) spectra at 5 and 40 K. The B\(_{2g}\) peak below 180 cm\(^{-1}\) has the steps B\(_2\), B\(_3\), and B\(_4\) as denoted in the spectra of \(x = 0.06\) in figure 17(a). The energies of the peaks A\(_2\), A\(_3\), and A\(_4\) are the same as the energies of steps B\(_2\), B\(_3\), and B\(_3\). This is more clearly observed by taking the derivative of the B\(_{2g}\) spectra with respect to the energy.

Figure 18 shows the A\(_{1g}\) (blue), B\(_{2g}\) (red), and the \(\frac{d(B_{2g})}{d\omega}\) (green). The A\(_K1\) and A\(_K2\) kinks and A\(_2\), A\(_3\), and A\(_4\) peaks correspond to the maxima and minima of
the $(\Delta B^{-2})/d\omega$ from $x = 0.035$ to 0.15. This proves that the step structure in the $B_2g$ spectra are produced by the Fano resonance between the continuum electronic scattering and the sharp phonon peaks. It is clear evidence that the $B_2g$ states below 180 cm$^{-1}$ are electron–phonon coupled polaronic states. The weak $B_2g$ hump from 180 to 380 cm$^{-1}$ is the second order of the $B_2g$–$B_1g$ peaks. The steps are also observed at $B_5$, $B_9$, and $B_{10}$ in figure 17(a) which have the same energies of the peaks $A_5$, $A_9$, and $A_{10}$, respectively.

ARPES observed a kink at 70 meV in the electronic dispersion in the nodal direction [167–169]. It is assigned to the half-breathing mode [170]. It is the $\Delta_1$ branch at $(\pi, 0)$ [171–177]. The small hump $A_{12}$ in figure 17 is produced by this mode. No structure is observed in the $B_2g$ spectra at 70 meV. The $A_{13}$ peak is produced by the $\Gamma$ point mode in the highest $\Delta_1$ and $\Sigma_1$ longitudinal phonon branch. The $A_{14}$ peak is the breathing mode which is the $(\pi, \pi)$ mode in the $\Sigma_1$ branch.

The $B_2g$ intensity at 100 cm$^{-1}$ is shown in figure 13(b) as a representative of the low-energy peak which is enhanced at low temperatures. The intensity rapidly increases from $x = 0$ to 0.06 and then gradually decreases with increasing the carrier density. It is consistent with the ARPES intensity near $(\pi, 0)$ [171–177]. The small hump $A_{12}$ is assigned to the half-breathing mode [170]. It is the $\Delta_1$ branch at $(\pi, 0)$ [171–177]. The small hump $A_{12}$ in figure 17 is produced by this mode. No structure is observed in the $B_2g$ spectra at 70 meV. The $A_{13}$ peak is produced by the $\Gamma$ point mode in the highest $\Delta_1$ and $\Sigma_1$ longitudinal phonon branch. The $A_{14}$ peak is the breathing mode which is the $(\pi, \pi)$ mode in the $\Sigma_1$ branch.

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Raman active mode does not interact with the long wavelength plasma, so that it is not affected by the carrier doping. On the other hand the infrared active mode interacts with the plasma. The energy of the longitudinal optical mode changes from $\omega_{LO}$ to $\omega_{TO} (\omega_{TO} < \omega_{LO})$, as the plasma energy $\omega_{pl}$ exceeds $\omega_{LO}$. If the crystal loses the inversion symmetry, some of the Raman active modes become infrared active. However, the higher energy shift of the satellite mode cannot be explained by the coupled mode, even if the $218 \text{ cm}^{-1}$ ($x = 0.06$) mode becomes infrared active. The coexistence of the original peak and the satellite peak suggests microscopic inhomogeneity in the crystal. This is discussed in section 4.1.

Figure 21 shows the differential spectra between 5 and 40 K. The superconducting pair-breaking peaks are shown by arrows. The gap energies (pair-breaking peak energies) are shown in figure 19. The $B_{1g}$ and $B_{2g}$ gap energies are consistent with the reported results [74, 105, 109, 113, 118]. The $B_{2g}$ gap energies are located between the $A_{1g}$ and $A_{2}$ peak energies at $x \leq 0.15$. It should be noted that the $A_{1g}$ and $B_{2g}$ gap energies are determined by the phonon energies and are independent of the $T_c$. The coupling between electrons and phonons has been observed in many experiments. For example, tunnel spectroscopy observed the coupling between the gap structure and phonons [178]. On the other hand the $B_{1g}$ gap energy decreases with decreasing $T_c$ at $x \geq 0.15$.

The $B_{1g}$ superconducting gap at $x = 0.15$ closes above $T_c$. This is different from ARPES stating that the pseudogap near $(\pi, 0)$ remains until $T^* = 150 \text{ K}$ [150, 151]. The $B_{2g}$ superconducting pair-breaking peak appears in the polaronic states. The SDW/CDW gap and the electron–phonon coupled peaks are the fine structure of the Fermi arc. ARPES did not detect the SDW/CDW gap. The different results may come from the higher resolution 0.7 meV and the longer penetration depth 0.1 $\mu$m of light in Raman scattering than the 15–20 meV and $\sim 5 \text{ Å}$ of the electron escape depth in ARPES [137, 150, 151]. The electron escape depth is shorter than the lattice constant along $c$, 13.1 Å.

4. Superconducting pairing model

4.1. Pairing at the edge dislocation of the stripe

Why does the electronic scattering show only $k \perp$ stripe excitations? In other words, why is the hole hopping restricted in the perpendicular direction to a stripe? It is reminiscent of the sliding of an edge dislocation in the Burgers vector direction [6, 9]. It is well-known that ductility of metal is induced by edge dislocations and screw dislocations [128]. In a two-dimensional layer only edge dislocations work. The edge dislocation easily slides in the direction perpendicular to the inserted stripe.

Figure 22(a) shows a single edge dislocation and (b) a looped (bridged) edge dislocation [6]. The hatched and white areas are oppositely spin ordered stripes. The boundaries are charge stripes. The open arrows are Burgers vectors. The Burgers vector is a vector that represents the direction and
Figure 22. Edge dislocation of (a) a single half charge stripe and (b) a looped charge stripe [6]. The hatched and white areas have opposite spin alignment: (b) is more stable than (a), because the both sides of the charge stripe have opposite spin arrangement. The dashed lines show the change of the charge stripes. The arrow shows the Burgers vector for the movement of the edge dislocation. (c) Movement of the edge dislocation from the blue to the red dashed lines. Spins of blue open circles and crosses change into red ones. Circle: up spin; cross: down spin; line: charge stripe.

Figure 23. The edge dislocation A in (a) moves to B in (b). Then the edge dislocation C moves to D in (c). The black and white stripes denote the two different antiferromagnetic spin alignments and the boundary is the charge stripe.
inter-charge stripe distance $d$ disturbs the stripe structure. The electronic states change into the dislocation density strongly increases and the movement intensity as discussed in section 2.1. In the overdoped phase the dislocation density strongly increases and the movement disturbs the stripe structure. The electronic states change into the normal metal at $x \approx 0.28$. At the same time the stripe component disappears in neutron scattering [24].

### 4.2. Coherence length

The superconducting coherence length $\xi$ is the size of superconducting pairs. It is known that the common coherence length $\xi = 1.5$ nm of many hole-doped high temperature superconductors is exceptionally short [129, 130, 183, 184]. It is in the crossover region of the BCS–BEC diagram [131, 132]. Figure 24 shows the carrier density dependence of the coherence length [129, 130] and the inter-charge stripe distance [19]. Both are surprisingly close at $x \leq 0.2$. This supports the model that two holes at the looped edge dislocation form a pair. The increase of the $\xi$ at $x > 0.2$ may be related to the increase of the edge dislocation density and the stripe structure changing into the normal metallic state. The coherence length is only twice the inter-charge distance, $\sqrt{a^2/\pi}$, where $a$ is the Cu–Cu distance on the assumption that all doped carriers form pairs. If we take into account the instantaneous picture that many carriers except for edges are localized, the overlap of pairs is much reduced. In the weak coupling BCS regime the Fermi surface is crucial for the stability of the superconducting state, but in the strong BEC region the Fermi surface is not important. As a result the high temperature superconducting state appears in spite of a pseudogap and a SDW/CDW gap.

### 5. Pseudogap

The pseudogap was first found in nuclear magnetic resonance (NMR) [185]. The pseudogap is observed in NMR [186], resistivity [181, 187], magnetic susceptibility [188], infrared spectroscopy [189, 190], polarized neutron diffraction [191–193], tunnel spectroscopy [194], polar Kerr-effect [195], Nernst effect [196], ARPES, and many other experiments [197, 198]. Many pseudogap models including the preformed superconducting pairs [179, 199–203], antiferromagnetic correlation [204, 205], and a density wave [206, 207] were proposed. ARPES reported that the pseudogap opens at the antinodal region near $(0, \pi)$ and $(\pi, 0)$ below $T^*$ on the $d$ wave superconducting gap curve (one-gap model) [149, 208, 209]. Recent ARPES, however, reported that the pseudogap energy is much higher than the extrapolated $d$ wave superconducting gap energy (two-gap model) in LSCO [151, 210], Bi$_{2-x}$Pb$_x$Sr$_2$CaCu$_2$O$_y$ (Bi2212) [211–214], and Bi$_2$Sr$_2$Ca$_{1-x}$Y$_x$Cu$_2$O$_4$ (Bi2212) [212, 215, 216]. The energy is about 80 meV (640 cm$^{-1}$) at the insulator–metal transition point in LSCO [137] and Bi2212 [215]. Hashimoto et al. [214] observed the particle–hole symmetry breaking in Bi2201, indicating that the pseudogap is distinct from the preformed superconducting gap.

We propose a new model based on our finding that the charge transfer is restricted only in the direction perpendicular to the stripe. Figure 25(a) shows the Fermi surface (thick solid line and the extending dashed line) and the group velocity (arrow) for the energy dispersion of equation (13) [137] at $x = 0.07$. The velocity is perpendicular to the Fermi surface. A quarter of the tetragonal Brillouin zone is shown. If the stripe is parallel to the $b$ axis, the allowed charge hopping direction is $a$. The one-dimensional conductor has a flat Fermi surface perpendicular to the conducting direction. The velocity of the Fermi surface near $(0, \pi)$ is parallel to the allowed charge transfer direction, but that near $(\pi, 0)$ is orthogonal to the allowed direction. Therefore the electronic transition across the Fermi surface near $(\pi, 0)$ is suppressed. It is observed as the pseudogap. The $B_{1g}$ electronic scattering spectra becomes the same as the $B_{2g}$ spectra above 2000 cm$^{-1}$ in the underdoped phase as discussed in section 3.3. It was understood that the isotropy in $k$ space for the electronic transition increases as the energy shift increases and the transition becomes completely isotropic above 2000 cm$^{-1}$ in the underdoped phase. The positions of $E = \pm 1000$ cm$^{-1}$ are shown by two thin solid curves in figure 25(a), although the isotropy in $k$ space indicates that the momentum is not a good quantum number. The curve on the $(0, 0)$ side is $E = -1000$ cm$^{-1}$ and that on the $(\pi, 0)$ side is $E = 1000$ cm$^{-1}$. The transition within these two curves is anisotropic and shows the pseudogap near $(\pi, 0)$. The pseudogap closes for the transition between the outer sides of the two curves. The stripe direction is fluctuating in the $a$ or $b$ direction. For the stripe parallel to $a$, the Fermi surface near $(0, \pi)$ has a pseudogap.

Figure 25(b) shows the Fermi surface at the optimum doping $x = 0.15$. The pseudogap is plotted so that the velocity on the Fermi surface has the same range of gradient as in the pseudogap at $x = 0.06$. The pseudogap decreases, because the position of the Fermi surface in $k$ space changes. The thin solid curves indicate the $E = \pm 1000$ cm$^{-1}$ positions.

Figure 25(c) shows the Fermi surface in the overdoped phase at $x = 0.22$. The velocity is not perpendicular to the $a$ axis on almost the whole Fermi surface except for the very small spot on the $(0, 0)$–$(0, \pi)$ line. Therefore the pseudogap does not appear. Thus the carrier density
Figure 25. The Fermi surface and the group velocity (arrow) for the dispersion of equation (13) at (a) $x = 0.07$, (b) $x = 0.15$, and (c) $x = 0.22$. The thick line is the Fermi arc of polaron states and the extending dashed line is the pseudogap. The red lines near $(0, \pi)$ and $(\pi, 0)$ are flat Fermi surfaces for the charge transfer in the $a$ and $b$ direction, respectively. The green line is the Fermi surface perpendicular to the $(\pi, \pi)$ phonons, because the electronic states near $(\pi/2, \pi/2)$ strongly interact with the $(\sigma, \pi)$ phonons. The two thin lines indicate the positions of $E = \pm 1000$ cm$^{-1}$ in (a) and (b) and $E = \pm 2500$ cm$^{-1}$ in (c). The dashed thin line shows the shadow Fermi surface. The line connecting $(\pi, 0)$ and $(0, \pi)$ in (a) and (b) is the Brillouin zone boundary of the orthorhombic structure and the antiferromagnetic structure. The dashed line connecting $(\pi, 0)$ and $(0, \pi)$ in (c) is the antiferromagnetic Brillouin zone. The crystal structure at $x = 0.07$ and $0.15$ is orthorhombic, while that at $x = 0.22$ is tetragonal at 10 K. (d) and (e) The Fermi surfaces for the stripes parallel to $b$ and $a$, respectively. The insets show the stripe structure. The black and white areas have different spin arrangement. The boundaries between the black and white areas are the charge stripes. (f) The Fermi surface at $x = 0.063$ obtained by Zhou et al in ARPES [218].

dependence of the pseudogap is naturally explained in the restricted charge transfer direction to $a$ or $b$. The boundary of the anisotropic–isotropic excitations is 4000–5000 cm$^{-1}$ in the overdoped phase. The thin solid curves indicate the $E = \pm 2500$ cm$^{-1}$ positions. A one-dimensional conductor has a flat Fermi surface. The tight binding Fermi surface for the stripes parallel to $b$ is rounded near $(0, \pi)$ at $x = 0.07$ in figure 25(a). If the Fermi surface near $(0, \pi)$ becomes flat and perpendicular to the $a$ axis as shown by the red line, the charge transfer increases and the kinetic energy decreases, because the group velocity is perpendicular to the Fermi surface. The Fermi surface for the stripes parallel to $b$ is shown in figure 25(d). The flat region near $(\pi/2, \pi/2)$ comes from a different mechanism, as discussed later. In the same way the Fermi surface near $(\pi, 0)$ becomes flat in figure 25(e) to decrease the kinetic energy for the stripes parallel to the $a$ axis. In the crystal of mixed stripe directions the observed Fermi surface is the average of figures 25(d) and (e). In fact the flat Fermi surface was observed near $(0, \pi)$ and $(\pi, 0)$ at $x = 0.063$ and 1/8 in ARPES [217–219]. Figure 25(f) shows the Fermi surface at $x = 0.063$ obtained by Zhou et al [218]. The one-dimensional charge transfer along the stripe was considered in ARPES [219], but the present experiment revealed that it is perpendicular to the stripe. The Fermi surface measured by ARPES has four-fold rotational symmetry, because the stripe direction is fluctuating in space and time. But the Fermi surface of the stripe phase has no four-fold rotational symmetry as shown in figures 25(d) and (e). The four-fold rotational symmetry breaking was observed in tunnel spectroscopy [194] and Nernst effect [196].

Another model to break the rotational symmetry is the d-wave Pomeranchuk instability [220, 221]. Yamase and Zhyher [222] calculated the Raman susceptibility near the d-wave Pomeranchuk instability. The d-wave Pomeranchuk instability couples to the B$_{1g}$ electronic and phononic excitations. A central peak emerges at the energy shift zero for each of the electronic and phononic B$_{1g}$ spectra, as temperature decreases in the carrier density below the critical value ($x \leq x_c$). The central peaks change into two low-energy peaks for the electronic and phononic channels at $x > x_c$. The soft mode energies increase with broadening, as the carrier density increases. The B$_{1g}$ spectra in figure 15(b) have not such a central peak nor the low-energy peak whose energy increases with increasing the carrier density. The B$_{1g}$ phonon of 218 cm$^{-1}$ has the satellite peak on the high-energy side. It
is the opposite side of the prediction from the Pomeranchuk model. The present Raman scattering experiment seems to give a negative result for the Pomeranchuk instability. Further investigation is necessary.

The electron–phonon coupled hump below 180 cm$^{-1}$ and the magnetic hump from 1000 to 3500 cm$^{-1}$ are strongly enhanced near the insulator–metal transition at low temperatures in the B$_{2g}$ spectra of figures 6 and 15. The electronic states near $(\pi/2, \pi/2)$ strongly interact with the A$_2$, A$_3$, and A$_4$ phonons at $(\pi, \pi)$ as discussed in section 3.4. The momentum $(\pi, \pi)$ is the reciprocal lattice vector to form the orthorhombic structure from the tetragonal structure and also the antiferromagnetic structure. If the electronic states with the velocity parallel to $(\pi, \pi)$ are preferable to stabilize the system through the electron–phonon interactions, the Fermi surface changes to increase the part in which the velocity is parallel to $(\pi, \pi)$. It is shown by the green line in figure 25(a).

The electron–phonon coupled hump below 180 cm$^{-1}$ is largest at $x = 0.06$ in figure 15(c). At almost the same carrier density at $x = 0.063$ Zhou et al. [218] observed the flat Fermi surface perpendicular to $(\pi, \pi)$ at the large area around $(\pi/2, \pi/2)$ in ARPES as shown in figure 25(f). It is supposed that the orthorhombic structure is stabilized by the dynamic coupling between the electronic states near $(\pi/2, \pi/2)$ and many $(\pi, \pi)$ phonons. It is, however, not determined whether the phonon wavevector is exactly $(\pi, \pi)$ or a little shorter to nest the Fermi surfaces near $(\pi/2, \pi/2)$ and $(-\pi/2, -\pi/2)$, because the phonon dispersions near $(\pi, \pi)$ are nearly flat. In the latter case the phonons work to increase the nesting susceptibility.

The thin dashed line in figures 25(a)–(c) is the shadow Fermi surface which is the $(\pi, \pi)$ shifted primary Fermi surface. It is the folded Fermi surface in the Brillouin zone of the orthorhombic structure and also the antiferromagnetic structure. The crystal structure is orthorhombic at $x = 0.07$ and 0.15 and tetragonal at $x = 0.22$. The shadow Fermi surface is observed in ARPES of Bi2212 [223, 224], Bi2201 [225], and LSCO [169, 226]. The Fermi pocket is observed in Bi2212 [224]. The shadow Fermi surface in LSCO is observed in the underdoped phase, but not in the overdoped phase [169, 226]. The magnetic hump from 1000 to 3500 cm$^{-1}$ is small at $x \approx 1/8$ in figure 6(c), while the shadow Fermi surface is observed [226]. Therefore the shadow Fermi surface is induced by the lattice effect in agreement with Mans et al. [223].

In the underdoped insulating phase ($x < 0.055$) the stripe direction changes into the diagonal direction [20]. However, the B$_{1g}$ and B$_{2g}$ spectra at $x = 0.035$ in figures 6 and 15 do not change qualitatively from the spectra in the metallic phase. Seibold and Lorenzana [56] calculated the $k \parallel$ and $k \perp$ stripe dispersions for magnetic excitations in the site-centered and bond-centered stripe structure at $x = 0.05$. It is difficult to assign the Raman data to the dispersions, because the number of dispersion segments is too many. In the calculation the intensity of the $k \perp$ stripe magnetic susceptibility is weak at the intermediate energy range [56]. The B$_{2g}$ spectra in figure 6(c) do not show a decrease at the middle of the hump from 1000 to 3500 cm$^{-1}$. The pseudogap is observed at $(0, \pi)$ and $(\pi, 0)$ in the extrapolated shape from the metallic phase in ARPES [137]. In the diagonal stripe parallel to $(\pi, \pi)$ the Burgers vector is parallel to $(-\pi, \pi)$. The pseudogap opens near $(\pi/2, \pi/2)$, if our mechanism of the pseudogap is applied to the insulating phase. But the experimental results are different. Therefore it is supposed that the charge transfer is large in the nearest neighbor direction $a$ or $b$. The resistivity of LSCO with $x = 0.03$ decreases on decreasing temperature from high temperature to 70 K in the same way as the metallic phase and then the resistivity increases below 70 K [93, 182]. This may be explained as follows. The effect of the different directions between the charge transfer and the Burgers vector is relaxed by the thermal excitation at high temperatures, but the difference becomes crucial at low temperatures and the resistivity increases. La$_2$NiO$_{4+\delta}$ with the diagonal stripe structure [17] is an insulator, too.

The high-energy excitations come from the short range electronic excitations. The excitations in short distance are complicated by the rearrangement of spins and charges in the moving looped edge stripe in figure 22(c). This may be the origin of the isotropic energy state in $k$ space. The pseudogap energy is 2000 cm$^{-1}$, if it is estimated from the split of the B$_{1g}$ spectra from the B$_{2g}$ spectra in figures 10 and 11. This energy is independent of the carrier density and temperature in the underdoped phase. The pseudogap energy observed by ARPES is about 80 meV (640 cm$^{-1}$) at the insulator–metal transition [151, 215]. Many ARPES experiments reported that the gap energy depends on the carrier density and the gap closes at $T^*$ [149–151, 211, 212, 214]. However, ARPES also reported the example that the pseudogap survives far above $T^*$ [227]. The large energy difference comes from the fact that (1) 2000 cm$^{-1}$ is the highest energy of the different B$_{1g}$ and B$_{2g}$ spectra and not the direct gap energy and (2) Raman scattering observes the energy from the valence band to the conduction band, while ARPES observes the energy from the valence band to the chemical potential.

6. Discussions

The large difference between the hole-doped cuprate superconductors and the electron-doped cuprates is the existence or absence of the stripe structure. Neutron scattering disclosed that the magnetic excitations are always commensurate $(\pi, \pi)$ in Nd$_{2-x}$Ce$_x$CuO$_4$ (NCCO), suggesting that the stripe is absent in electron-doped cuprates [228, 229]. The B$_{1g}$ Fleurie–Loudon type two-magnon peak softens on increasing the carrier density in the hole-doped cuprates as shown in figure 6(b) [105]. However, the two-magnon peak does not soften in electron-doped cuprate superconductors [230–233]. In the metallic phase the B$_{1g}$ two-magnon peak disappears and the spectra shifts to much higher energy than the original two-magnon peak energy [233]. The spectra are nearly the same as the B$_{2g}$ spectra. Therefore the softening of the B$_{1g}$ two-magnon peak is not a common property in a doped antiferromagnet, but the property of the $k \parallel$ stripe magnetic excitations. The center of the spectral weight is much higher than that in LSCO. This is explained by the longer mean free path of the carrier than
interactions are observed in the B$_{2g}$ may not be the bulk phonons. The strong electron–phonon interactions. The phonons localized at the edge dislocation formation energy including the electron, spin and charge not only the electron–phonon interaction but also the stripe like a bi-polaron [236, 237]. The binding energy is related to pairs are formed at the looped edge dislocations. Therefore the Cooper pairing is formed between charge stripes. The moving carriers are much longer [234, 235]. This clearly indicates that the coherence lengths of electron-doped cuprate superconductors NCCO and Pr$_2$–Ce$_{2−x}$CuO$_4$ (PCCO) are much longer [234, 235]. This clearly indicates that the pairing is formed between charge stripes. The moving carriers are only at the looped edge dislocations. Therefore the Cooper pairs are formed at the looped edge dislocations.

The paired charges moving with the edge dislocation is like a bi-polaron [236, 237]. The binding energy is related to not only the electron–phonon interaction but also the stripe formation energy including the electron, spin and charge interactions. The phonons localized at the edge dislocation may not be the bulk phonons. The strong electron–phonon interactions are observed in the B$_{2g}$ channel. The existence of the phonon contribution is known from the isotope effect of the penetration length [238], although the isotope effect of the $T_c$ is small at the optimum doping [239]. The contribution of phonons can introduce a retardation effect to the pairing so that instantaneous Coulomb repulsion is avoided [240–242].

7. Conclusion

The individual $k \parallel$ and $k \perp$ stripe excitations in fluctuating spin–charge stripes were separately detected utilizing the different Raman processes in the B$_{1g}$ and B$_{2g}$ spectra. The electronic scattering has only $k \perp$ stripe excitations, indicating that the charge hopping is restricted to the perpendicular direction of the stripe. This is the same as the Burgers vector pointing to the sliding direction of an edge dislocation. It suggests the model that holes at the edges transfer together with edge dislocations, while other holes are localized. The charge hopping perpendicular to the stripe causes the one-dimensional transport along the $a$ or $b$ axes, because the stripe direction is $b$ or $a$ in the metallic phase. The Fermi surface becomes flat near $(0, \pi)$ and opens the pseudogap near $(\pi, 0)$ in the charge stripe parallel to the $b$ direction. In fact the flat Fermi surface and the pseudogap are observed in ARPES. The charge hopping in the $a$ and $b$ directions cannot produce the Fermi surface around $(\pi/2, \pi/2)$. We found that the electronic states around $(\pi/2, \pi/2)$ are interacting with the soft mode inducing the tetragonal–orthorhombic phase transition. All the observed Raman results are consistent with the charge hopping at the edge dislocations in the direction perpendicular to the stripes. The reported superconducting coherent length is the same as the inter-charge stripe distance at $x < 0.2$. This strongly suggests that the Cooper pairs are formed at the moving edge dislocations of the fluctuating charge stripes. The coherence length is intermediate between the BCS and the BEC superconductors. The looped (bridged) charge stripe has lower energy than the single charge stripe. The binding force between the charge stripe at the sliding edge dislocation is the key of the superconductivity.

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