c-Axis Optical Response in the Static Stripe Ordered Phase of the Cuprates

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The c-axis far-infrared reflectivity spectra in the superconducting state were investigated for $La_{1.85-y}Nd_ySr_{0.15}CuO_4$. The Josephson plasma edge rapidly shifts towards lower frequency with increasing y, and almost disappears when y exceeds a critical value ($y_c \approx 0.12$) above which the low-temperature-tetragonal (LTT) deformation switches on, stabilizing the spin/charge ordered stripe phase. In the vicinity of y_c, a "normal state" reentrant behavior was observed with lowering temperature below T_c. This clearly demonstrates that the static stripe order pinned by the LTT distortion suppresses the interlayer phase coherence.

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Since Tranquada *et al.* discovered the incommensurate splitting of the neutron diffraction spots in $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ [1], the stripe ordering, which is also observed in the manganites [2] and the nickelates [3], has also come up for active discussions in the cuprates. Although it seems to be a common characteristic in strongly correlated electronic systems [4], there are many unresolved problems specific to the cuprates. The most debated and important problem is whether the stripe order or its fluctuation plays a crucial role in the superconductivity pairing mechanism [5] or acts only as a competitor.

The best documented control parameter suppressing superconductivity and/or promoting the stripe order is the rare earth (Nd or Eu) substitution in the cuprates of composition $La_{2-x-y}Nd_ySr_xCuO_4$ (LNSC) [6]. LNSC shows the low-temperature-tetragonal (LTT) distortion due to the tilt of CuO_6 octahedra. The LTT deformation is regarded as a collective pinning potential stablizing the static stripe order [1]. The superconducting transition temperature T_c is considerably reduced in the static stripe phase, and hence the static stripe order is apparently competing with the superconducting order. Nevertheless, evidences have been accumulated for the existence of a state in which the stripe order coexists with superconductivity in LNSC with $y = 0.4$ and x larger than $1/8$ [1,7,8]. From the optical and microwave measurements, it was demonstrated that a reduction in T_c in LNSC is associated with the elongation of the in-plane penetration depth (λ_{ab}) [7].

Compared with the in-plane properties of the stripe phase which have so far been intensively studied, little is known about the c-axis properties. The optical spectrum for a polarization parallel to the c axis is an alternative probe for superconductivity in the cuprates. When the system goes into the superconducting state, a sharp plasma edge appears in the c-axis reflectivity spectrum due to the establishment of c-axis phase coherence [9], while it is overdamped in the normal state, reflecting incoherent motion of carriers. Since it is known that superconducting CuO_2 layers are weakly coupled via the Josephson tunneling [10], this reflectivity edge is called Josepshon plasma edge. In this paper, we report the c-axis reflectivity spectra of $La_{2-x-y}Nd_ySr_xCuO_4$ single crystals for various y's mainly with $x = 0.15$ that illustrate how the c-axis coherence is suppressed as the stripes become static with increasing Nd content.

The single crystals of LNSC, grown by a traveling solvent floating zone method, were well characterized by neutron diffraction, magnetic susceptibility, resistivity and Hall coefficient measurements [11,12]. The Nd substitution reduced the superconducting transition temperature T_c. For example, at $x = 0.15$, T_c is decreased from 36 K for $y = 0$ down to 12 K for $y = 0.4$. The samples for optical study were cut from the as-grown crystal rods, carefully examining crystal axes by x-ray Laue diffraction patterns. After annealing in flowing oxygen gas, the *ac* face of the samples were polished to obtain optically flat surfaces. Reflectivity spectra were measured with polarization parallel to the c axis using a Fourier transformation-type spectrometer ($\omega > 30$ cm^{-1}) equipped with a bolometric detector and a He-gas flow cryostat.

Figure 1 shows a comparison of the c-axis reflectivity spectra of Nd-free and Nd-substituted ($y = 0.4$) LNSC for $x = 0.15$ and 0.2. The Nd-substituted samples show static spin order [13] (for $x = 0.15$ the charge order also has been observed in the hard x-ray diffraction measurement [14]), probably coexisting with the superconducting order at low temperatures. In all of the samples, the normal state spectra below 150 cm^{-1} are almost flat, indicating an extremely low conductivity due to incoherent charge transport in the c direction. For the Nd-free crystals, when temperature is lowered below T_c, a sharp plasma edge develops (shown by the dotted curves in Fig. 1). As is well known, a plasma edge shifts towards higher frequency with hole doping. The rounded plasma edge (the shallow dip) seen for $x = 0.20$ is suggestive of a finite conductivity remaining within the superconducting gap region. It was argued that the remaining conductivity may originate from quasiparticles associated with a nodal d-wave gap, or from

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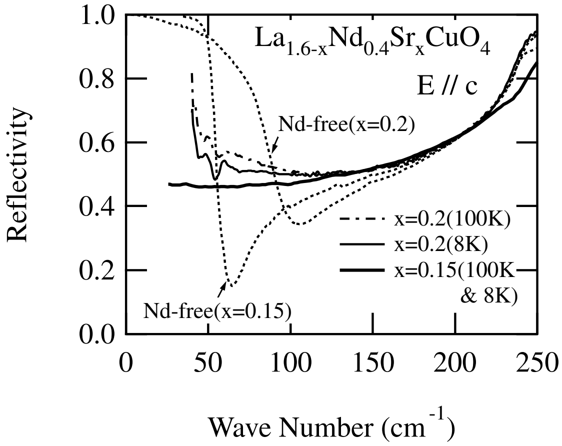

FIG. 1. The c-axis reflectivity spectra of $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ for $x = 0.15$ and 0.20 above and below T_c. The 8-K spectra of Nd-free crystals with the same Sr contents are presented by dashed curves as references [9].

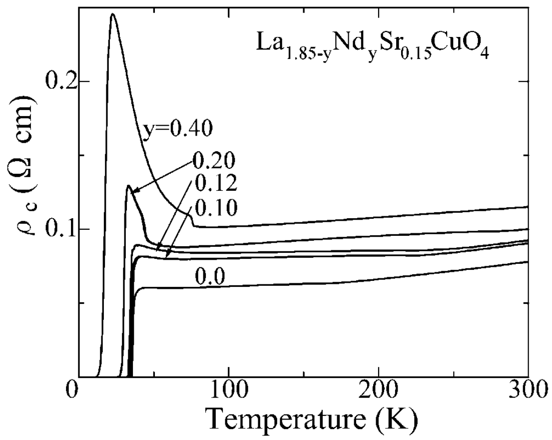

FIG. 2. T dependence of the c-axis resistivity for $La_{1.85-y}Nd_ySr_{0.15}CuO_4$ with various y's.

some amount of uncondensed carriers appearing as a result of possible coexistence with the normal metallic region in the highly doped compounds [15].

In the crystals with $y = 0.4$, no clear Josephson plasma edge can be seen at 8 K lower than T_c (12 K for $x = 0.15$ and 16 K for $x = 0.20$). Although the low-ω limit of the present experiment is 30 cm^{-1}, the reflectivity above 30 cm^{-1} should show an incipient decrease below T_c if the plasma edge is located not far below 10 cm^{-1}. Therefore, the absence of such a decrease in Fig. 1 indicates that the plasma edge is suppressed far below 10 cm^{-1} even if it may exist. On the other hand, the spectrum for $x = 0.2$ shows a slight decrease in reflectivity below T_c, suggesting a plasma edge located about 30 cm^{-1} in a heavily damped form due to residual conductivity within the superconductivity gap. In either case, the static stripe order radically suppresses the interlayer phase coherence between the superconducting CuO_2 planes.

The question is if the collective pinning potential could be switched on continuously or discontinuously with increasing y. Motivated by this problem, we investigated the Nd-content (y) dependence of the spectrum for Sr content fixed at $x = 0.15$. Figure 2 shows the temperature dependence of resistivity in the c direction for various y's. With increasing y, resistivity increases and T_c decreases. When y exceeds 0.12, resistivity exhibits a small jump at T_d ($\approx$40 K for $y = 0.20$, for example), certainly corresponding to the phase transition from the low-temperature-orthorhombic (LTO) to the low-temperature-tetragonal structure [12]. According to Ref. [16], the onset temperature of static stripe order coincides with, or is slightly lower than, T_d. For the Nd content slightly larger than 0.12 ($y = 0.12 + \delta,\ \delta < 0.01$), the phase transition was found below T_c in the $\rho_c(T)$ measurement when suppressing superconductivity in a high magnetic field ($\approx$7 T) [12]. The superconducting transition is sharp for any compound, indicating homogeneous distribution of Sr and Nd atoms in a crystal. Changes of the phase transition temperatures T_d and T_c with y are plotted in Fig. 3. It is seen in the figure that the LTT phase rapidly develops above $y = 0.12$, while the change in T_c is steepest at $y \approx 0.12$ but is overall rather moderate.

Figure 4 shows the c-axis spectra of $La_{1.85-y}Nd_y$-$Sr_{0.15}CuO_4$ with various y's at 8 K. For larger y's ($= 0.2$ and 0.4), where $T_d > T_c$, i.e., the superconducting state is in the LTT phase, the Josephson plasma edge can hardly be seen. For the same reason described earlier, we can conclude that the screened plasma frequency $\omega'_{ps} = \omega_{ps}/\epsilon_\infty^{1/2}$ ($\epsilon_\infty = 25$ in the present case) would be reduced to, at most, 10 cm^{-1} in the LTT phase. For smaller y's in the LTO phase, as y increases, the plasma edge shifts towards lower frequency, accompanied by an increase of a dip reflectivity. The low-ω shift of the edge implies weakening of the Josephson coupling strength between the layers, while the dip shallowing is indicative of a little increase in inhomogeneity such as an increase of residual conductivity or distribution of Josephson plasma frequency. Note that a precursory decrease of ω'_{ps} with y is observed even in the LTO phase at $y < 0.12$. This

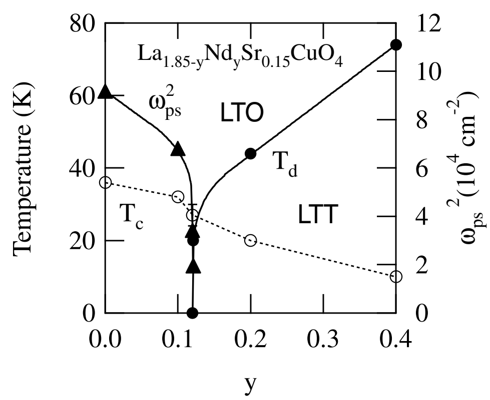

FIG. 3. Nd-content (y) dependence of T_c (open circle), T_d (solid circle), and ω_{ps}^2 (triangle) for $La_{1.85-y}Nd_ySr_{0.15}CuO_4$. Solid curves are guides for the eye.

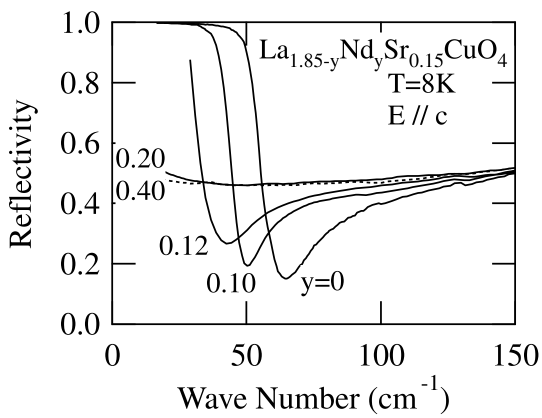

FIG. 4. c-axis reflectivity spectra of $La_{1.85-y}Nd_ySr_{0.15}CuO_4$ at 8 K for various Nd contents.

suggests that even a small amount of Nd substitution can slow down the stripe fluctuation, the presence of which in even Nd-free LSC has been suggested by recent experiments [7,17,18]. The Josephson plasma frequency is plotted as a function of y in Fig. 3 [19].

For $y = 0.12$, the Josephson plasma edge gradually develops below T_c ($\approx$30 K) and the dip position shifts to higher frequencies with lowering temperature, as in the case of LSCO ($y = 0$). However, a dramatic phenomenon is observed when the Nd content is slightly higher than 0.12 ($y = 0.12 + \delta$) but very close to the critical value y_c, where $T_d \approx T_c$. In Fig. 5 the c-axis spectra for the compound with $y = 0.12 + \delta$ are shown at various temperatures. In this sample, T_c ($\approx$25 K) is higher than T_d ($\approx$20 K), as described above. Below T_c, a reflectivity edge shows up and develops at about 35 cm^{-1} at 20 K. When the sample is cooled below T_d, the plasma edge starts to degrade—the dip in reflectivity becomes shallow and shifts to lower frequencies, as if the sample goes back to the normal state. This "normal state reentrant" behavior is further evidence for the loss of c-axis coherence in the LTT phase. With further cooling below 10 K, the plasma edge grows again, recovering the c-axis coherence. To illustrate the nonmonotonic behavior, the plasma frequency is plotted as a function of T in the inset of Fig. 5(b).

A possible explanation would be the following. At first, superconductivity sits in the LTO phase in which the Josephson plasma mode develops with lowering temperature. When the temperature goes below T_d ($\approx$20 K), a substantial portion of the crystal undergoes the transition to the LTT phase in which the superconducting order would be destroyed. As a result, the overall c-axis phase coherence is severely damaged. However, some portion of the crystal (or some of the layers) remains in the LTO phase, where the Josephson plasma continues to develop and dominates again at low temperatures below 15 K, probably when the superconducting order is also induced in the LTT phase by the proximity effect.

The mixed phase behavior in the vicinity of y_c ($\approx$0.12) suggests that there might be a first-order transition line at

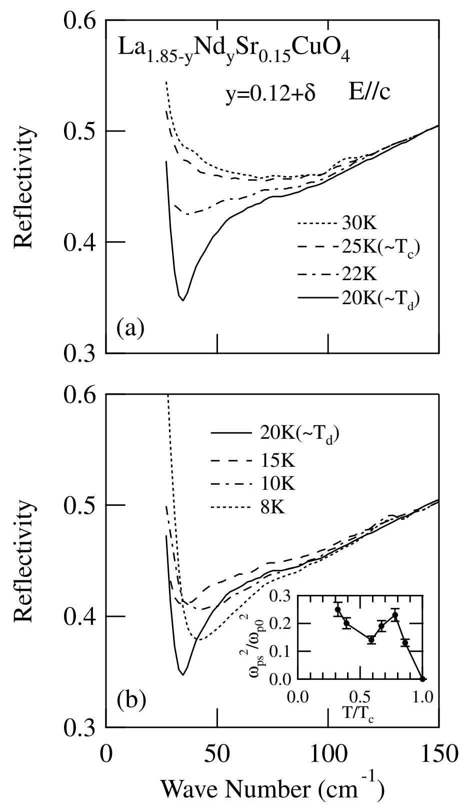

FIG. 5. T dependence of the c-axis reflectivity spectrum for $y = 0.12 + \delta$ with $T_c \approx 25$ K above (a) and below (b) $T_d \approx 20$ K. Inset: normalized square of the Josephson plasma frequency ω_{ps} versus normalized temperature. $\omega_{p0}(= 300\ cm^{-1})$ is ω_{ps} for the Nd-free sample at $T = 0$ K.

$y = y_c$. In fact, as illustrated in Fig. 3, $\omega_{ps}^2(y)$ steeply decreases as y approaches y_c, while $T_d(y)$ shows an almost vertical rise when y exceeds y_c. As nonzero ω_{ps} signals the long-range c-axis phase coherence, the $y = y_c$ line separates two distinct superconducting states with and without c-axis coherence, and the transition between the two is likely in first order. Concerning the role of lattice distortion in the pinning mechanism of stripes, it is known that when the tilt angle of CuO_6 octahedra due to the LTT distortions exceeds the critical value at $y = y_c$ (y_c is dependent on x [6]), the pinning potential becomes strong enough to pin the stripes collectively. Thus, it is plausible that in the superconducting state for $y > y_c$ the static stripe order coexists but destroys the c-axis coherence.

The observed stripe effect on the optical spectra looks similar to the pair-breaking effect due to Zn substitution. Zn also gives rise to a rapid reduction in the Josephson plasma frequency [20] as well as an enhancement of the in-plane penetration depth [21]. Recently the scanning tunneling microscope observation revealed that superconductivity is destroyed locally around the doped Zn atom [22]. As in the case of static stripe order where antiferromagnetic (AF) domains are formed between the charge stripes, AF correlation is enhanced or a short-range AF order is induced around Zn [23]. Therefore, in both cases it is expected that the superconductivity order parameter is spatially modulated. Such a superconducting state has also been suggested by the magnetization (MH) characteristics [12]. For the static stripe state, the superconducting order might be strongly suppressed in the spin domains and survive on the charge stripes. A reduction in the interlayer Josephson coupling strength would result from the fact that the stripe orientation is alternately rotated by 90° layer by layer [24], which restricts an effective superconducting area where the Josephson current can flow.

In summary, we have demonstrated that the static stripe order stabilized by Nd substitution has a dramatic effect on the c-axis optical spectrum in the superconducting state. The Josephson plasma frequency is radically reduced, when the Nd content approaches the critical value ($y_c \approx 0.12$) above which the stripes become static, due to the collective pinning of the LTT lattice distortion. The superconductivity coexisting with the static stripe order is a state with a suppressed c-axis phase coherence.

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