Josephson Plasmon and Inhomogeneous Superconducting State in $La_{2-x}Sr_xCuO_4$

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We report on the interlayer far infrared response for a series of $La_{2-x}Sr_xCuO_4$ crystals with $0.08 < x < 0.20$ focusing on the survey of the Josephson plasmon resonance (JPR). The analysis of the JPR mode provides information on the local variation of the superfluid density within the CuO_2 planes thus empowering one with a tool for "microscopy" on the superconducting condensate. Our results uncover the presence of regions with characteristic length of ~100–200 Å within which superconductivity is strongly depressed or completely depleted. An examination of the doping trends suggests that development of superconducting inhomogeneities is triggered by the formation of the unidirectional spin density wave state in $La_{2-x}Sr_xCuO_4$ at $x = 1/8$.

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High-T_c superconductivity is achieved when a moderate density of conducting holes is introduced in the CuO_2 planes. Numerous experiments indicate that doping can be highly nonuniform leading to a formation of hole-rich and hole-depleted regions within the CuO_2 planes [1–3]. Such electronic phase separation occurring on diverse length scales appears to be an intrinsic attribute of doped Mott-Hubbard (MH) insulators [4] and also offers new approaches to elucidate high-T_c superconductivity. However, it has yet to be determined if the *superconducting condensate* in cuprates is characterized by similar local disproportions. Here we attempt to extract information on condensate inhomogeneities through an examination of the Josephson plasma resonance (JPR) in a series of $La_{2-x}Sr_xCuO_4$ (LSCO) single crystals: a prototypical doped MH insulator. The JPR mode originates from tunneling of Cooper pairs across the CuO_2 layers. An analysis of the JPR resonance shows that the local variations of the superfluid density within the layers are most prominent near the doping regime where the neutron scattering experiments indicate development of quasi-static unidirectional spin density waves in LSCO [5].

We applied infrared spectroscopy to examine the JPR mode and the interlayer ($\mathbf{E} \parallel c$) response of several well-characterized LSCO crystals at seven different doping levels, with Sr content varying between $x = 0.08$ and $x = 0.2$ [6,7]. The near-normal-incidence reflectance $R(\omega)$ was collected over a broad temperature (7–300 K) and frequency (10–48 000 cm^{-1}) range using a combination of interferometers and a grating monochromator. The absolute values of $R(\omega)$ were determined using an *in situ* Au- or Al-coating procedure described in Ref. [8]. The optical conductivity $\sigma_1(\omega) + i\sigma_2(\omega)$ and complex dielectric function $\epsilon_1(\omega) + i\epsilon_2(\omega)$ were calculated from $R(\omega)$ using Kramers-Kronig analysis. The strength of the $\delta(0)$ function in $\sigma_1(\omega = 0, T < T_c)$ was quantified with the plasma frequency $\omega_s^2 = 4\pi e^2 n_s/m^*$, where n_s is the density of superconducting carriers and m^* is their mass. The magnitude of ω_s was extracted from the imaginary part of the conductivity using a technique proposed by Dordevic *et al.* [9] that corrects for screening due to unpaired charge carriers in the conductivity data at $T < T_c$.

Figure 1 displays the raw reflectance data (black lines) for all samples in the superconducting state at 7 K. All spectra show well-defined plasma edges. The frequency positions of plasma edges systematically increase with doping in accord with earlier studies [9–11]. The simplest model that captures the gross features of the c-axis response of cuprates, including the JPR mode, is the "two fluid" model which yields the following form for the dielectric function:

$$\epsilon(\omega) = \epsilon_\infty - \frac{\omega_s^2}{\omega^2} - \frac{\omega_n^2}{\omega^2 + i\gamma_{sr}\omega}, \qquad (1)$$

where ϵ_∞ is the high frequency dielectric constant, ω_n is the plasma frequency of unpaired carriers, and γ_{sr} is their scattering rate. The gray lines in Fig. 1 show the best fits of reflectance calculated using Eq. (1). For consistency the values of $\epsilon_\infty = 27$ and $\gamma_{sr} = 5000$ cm^{-1} were kept the same for all samples and only ω_s and ω_n were varied. As can be seen from Fig. 1 the fit is adequate for $x = 0.08$, 0.10, 0.17, and 0.20 samples but not for $x = 0.12$, 0.125, and 0.15 crystals. In these latter materials the plasma minimum is much broader than the model predicts. Especially poor are the fits for both $x = 0.125$ compounds which signals a breakdown of the two-fluid description for $x \simeq 1/8$.

An anomalous response of the samples near $x = 1/8$ doping is also evident in the behavior of the optical constants (Fig. 2). Crystals in the vicinity of $x = 1/8$ show an additional absorption feature in the dissipative part of the conductivity $\sigma_1(\omega)$, as well as a strongly asymmetric loss function $-\mathrm{Im}[1/\epsilon(\omega)]$. The frequency position of the absorption resonance in $\sigma_1(\omega)$ tracks the location of the plasma edge in $R(\omega)$. At $T \ll T_c$ the magnitude of the conductivity at the resonance center dramatically exceeds the background conductivity (1–1.5 Ω^{-1} cm^{-1}). No such peak is detected in $\sigma_1(\omega)$ for 0.08, 0.10, 0.17, and

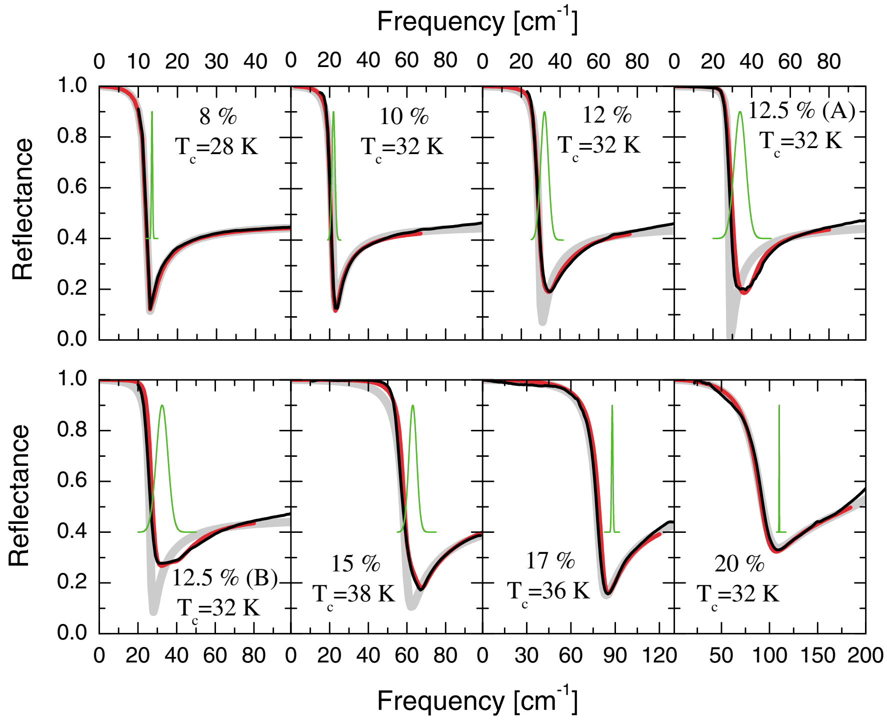

FIG. 1 (color). Infrared reflectance of LSCO samples (black lines) at 7 K. The gray lines are the fits using a two fluid model [Eq. (1)]. Fitting parameters ω_s and ω_n in cm^{-1}, for different dopings x: $(x,\ \omega_s,\ \omega_n)$ = (0.08, 65, 390), (0.1 115 550), (0.12 155 500), (0.125 160 800), (0.125 150 800), (0.15 320 800), (0.17 418 1150), (0.2 550 2200). The red lines are the fits using Eq. (2). Gaussian distributions of plasma frequencies [Eq. (3)] used for each particular sample are also shown with green lines.

0.20 samples. Inspection of the loss function spectra is also instructive since $-\mathrm{Im}[1/\epsilon(\omega)]$ uncovers the line shape of the longitudinal modes, including the JPR resonance. Within the two-fluid scenario the JPR mode produces a Lorentzian peak at $\omega_0 = \omega_s/\sqrt{\epsilon_\infty}$. The width of the peak is determined by the quasiparticle current [third term in Eq. (1)]. A Lorentzian-like mode in the $\mathrm{Im}[1/\epsilon(\omega)]$ spectra for $x = 0.08$, 0.1, 0.17, and 0.2 crystals attests that the JPR response of these materials is fully consistent with the two-fluid description, implying homogeneous interlayer coupling between uniform superconducting layers. This behavior is to be contrasted with the asymmetric JPR mode in crystals with x near $1/8$.

As we detail below, the JPR anomalies in samples with $x \simeq 1/8$ give evidence for an inhomogeneous superconducting condensate. For the purpose of concrete discussion, we focus on two models addressing the issue of inhomogeneous condensate from somewhat different (albeit complementary) prospectives. van der Marel and Tsvetkov (vdMT) have been able to show that an

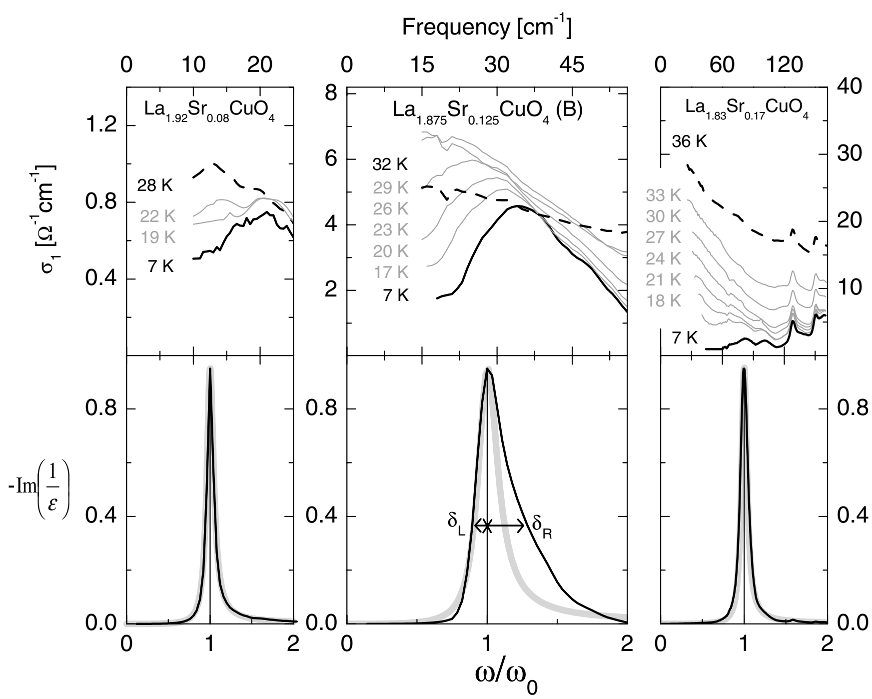

FIG. 2. The optical conductivity $\sigma_1(\omega)$ (top panels) and the normalized loss function spectra (bottom panels) for 0.08, 0.125, and 0.17 samples, at $T \le T_c$. The frequency axes are displayed both in cm^{-1} and in normalized units ω/ω_0 where ω_0 is the frequency position of a JPR mode in the loss function spectra. A prominent absorption resonance in the $\sigma_1(\omega)$ spectrum near the frequency position of the JPR mode is observed only for the $x = 0.125$ sample. The loss function spectrum for the $x = 0.125$ sample reveals a pronounced asymmetry inconsistent with the two-fluid description. The loss function data for both $x = 0.08$ and 0.17 crystals are in accord with the two-fluid model (gray lines).

additional absorption feature in the response of a layered superconductor can be linked to a nonuniform distribution of Josephson coupling constants between the layers [11]. They have suggested the following equation for the complex dielectric function:

$$\frac{1}{\epsilon(\omega)} = \int_0^\infty \frac{1}{\epsilon_i(\omega, \omega_s)} \rho(\omega_s) d\omega_s, \tag{2}$$

where $\rho(\omega_s)$ is the normalized distribution of Josephson plasma frequencies and $\epsilon_i(\omega, \omega_s)$ is the complex dielectric constant for each individual component. We show that this proposal remedies problems with the two-fluid description of the data for samples with $x \simeq 1/8$ provided $\rho(\omega_s)$ is substantially broadened. Koshelev, Boulaevski, and Maley (KBM) investigated broadening of the loss function associated with the JPR mode due to random distribution of normal state regions *within* the CuO_2 planes [12]. According to Ref. [12] such randomness triggers the asymmetric line shape of $-\mathrm{Im}[1/\epsilon(\omega)]$. Within the KBM scenario the high energy tail in the loss function spectra is directly related to mixing between *local* Josephson modes with different momenta and frequency produced by in-plane inhomogeneities. An advantage of the KBM approach is that it allows one to address the issue of length scales associated with the inhomogeneous superconducting state. Short range inhomogeneities are averaged out by slowly varying external field leading to narrow JPR resonances typically observed in most cuprates. However, mode mixing resulting in the asymmetric broadening of the JPR plasmon gains prominence if the length scale associated with inhomogeneities is comparable to the so-called Josephson length $\lambda_J = \gamma s$, where γ is the ratio between the in-plane and interplane condensate superfluid plasma frequencies $\gamma = \omega_{s,\mathrm{ab}}/\omega_{s,c}$ and s is the interlayer spacing [13]. Below we exploit both of these complementary models to characterize spatial disproportions in the superconducting condensate in LSCO.

In order to describe the anomalous form of the JPR plasmon near $x = 1/8$ in the spirit of Eq. (2) we used Eq. (1) for $\epsilon_i(\omega)$ and a Gaussian distribution of Josephson plasma frequencies:

$$\rho(\omega_s) = \frac{1}{\beta\sqrt{2\pi}} e^{-1/2[(\omega_s - \omega_{s0})/\beta]^2}, \tag{3}$$

where ω_{s0} is the center of distribution and β is its width. Fitting results are shown in Fig. 1 with red lines, along with the corresponding Gaussian profiles (green lines). Equation (2) provides a satisfactory account of the data for all samples. However, the width of the Gaussian distribution needed to generate successful fits reveals systematic variation with doping. For $x = 0.08$, 0.10, 0.17, and 0.20 samples the input distributions are narrow, as expected from the fact that the two-fluid model was sufficient. Conversely, for 0.12, 0.15, and both 0.125 crystals much broader distributions were required [see Fig. 3(a) where we plot the doping dependence of the reduced Gaussian width $\tilde{\beta} = \beta/\omega_0$]. In order to characterize the asymmetry of the JPR mode in the spirit of the KBM model, we estimated the difference between the half-width of the loss function on the left δ_L and on the right δ_R of the resonance at ω_0. In Fig. 3(a) we also show the JPR asymmetry defined as $W_{\mathrm{JPR}} = (\delta_R - \delta_L)/\omega_0$. It is clear from Fig. 3(a) that both W_{JPR} and $\tilde{\beta}$ reveal similar doping trends, suggestive of strong *spatial* variation of the superconducting condensate [17]. Moreover, the doping dependence of both these parameters quantifying the nonuniformity of interlayer Josephson coupling ($\tilde{\beta}$ and W_{JPR}) appears to track that of spin correlation length ξ_s extracted from neutron scattering measurements [5,15,16] and replotted in Fig. 3(b). This connection points to an interdependence between spin ordering and an inhomogeneous superconducting state in the LSCO system.

Both the KBM and vdMT approaches suggest that the superconducting condensate in the LSCO system is highly nonuniform within the CuO_2 planes with special prominence of the effect at $x \simeq 1/8$. This composition is unique within the so-called stripes picture which predicts the formation of unidirectional spin and charge density waves in doped MH systems [4]. In particular, $x = 1/8$ corresponds to a commensurate lock-in of the stripe structure that is believed to be responsible for numerous transport and superconducting anomalies [4]. Neutron scattering experiments support the relevance of the stripes hypothesis to the 1/8 anomaly in LSCO and reveal a dramatic enhancement of the correlation length associated with the stripelike spin modulations [5,15,16]. In this context, an association between the enhanced spin

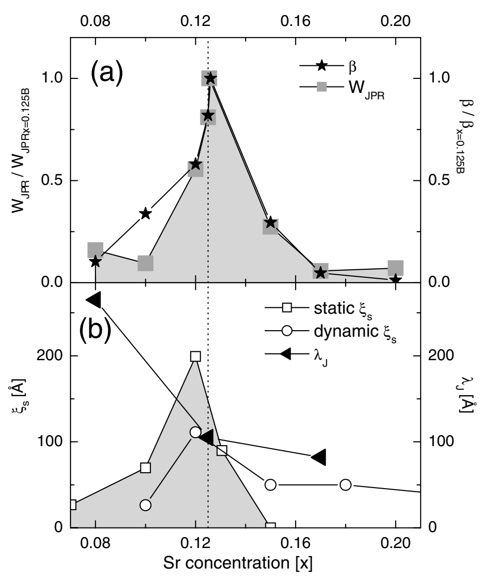

FIG. 3. (a) The doping dependence of the Gaussian distribution width $\tilde{\beta}/\tilde{\beta}_{x=0125\,\mathrm{B}}$ and of the asymmetry of the loss function $W_{\mathrm{JPR}}/W_{\mathrm{JPR},x=0.125\,\mathrm{B}}$ defined in the text; both parameters characterize the degree of inhomogeneity of superfluid density within the CuO_2 planes. (b) The correlation length ξ_s of both static and dynamic spin structure extracted from neutron scattering experiments [5,15,16], and the Josephson length λ_J determined using a procedure discussed in Ref. [13].

ordering in LSCO and parameters characterizing inhomogeneous superfluid response is quite remarkable. It is feasible that superconducting inhomogeneities also arise from the stripelike ordering in LSCO. Within this picture the superconducting condensate resides on narrow stripes separated by spacers of an antiferromagnetic insulator with the width of a few lattice spacings. However, an obvious problem with this interpretation of the JPR data is that the plasma resonance is formed over much more extended regions of the order $\lambda_J \simeq 80$–250 Å in LSCO crystals [Fig. 3(b)]. Because inhomogeneities explicitly attributable to stripes are extremely short ranged they are likely to be averaged out, as discussed above [18].

The length scales associated with the asymmetry of the JPR feature are suggestive of a formation of fairly large droplets within the CuO_2 planes where superconductivity is either suppressed or completely depleted. This picture can be readily reconciled with the neutron data assuming that the spin-ordered signal is produced by regions of depressed superconductivity. The size of spin-ordered droplets can be estimated from the correlation length extracted from the neutron data and is indeed comparable to λ_J at $x = 1/8$ doping where $\xi_s \simeq 200$ Å [Fig. 3(b)]. Further support for the scenario of phase separation into stripe-ordered and superconducting regions on the length scale $\simeq 200$ Å comes from the local spectroscopic probes [19]. An important implication of this picture to the interpretation of the JPR data is that fluctuating stripes (away from 1/8 doping [20]) do not initiate substantial local depression of the superfluid density, so that the JPR mode registers nearly homogeneous condensate distribution. However, at $x \simeq 1/8$ stripe fluctuations slow down as witnessed by elastic neutron scattering. The new quasistatic striped state appears to be destructive for superconducting pairing leading to local depression of the condensate over extended spatial regions. A role played by charge-ordering effects in LSCO crystals with Nd doping on the behavior of the JPR mode deserves further examination [21].

In conclusion, the "JPR microscopy" enabled through the analysis of the line shape of the Josephson plasmon in layered cuprates uncovers a nonuniform distribution of the superconducting condensate with the CuO_2 planes in LSCO system. In conjunction with the neutron scattering data this analysis shows that near the $x = 1/8$ doping level a spontaneous formation of superconducting and antiferromagnetic droplets overpowers the global Cooper condensation in LSCO system. Such phase separation occurring on the length scale $\simeq 100$–200 Å can be viewed as a sign of competing ground states. A well-known example of phase separation is vortices in type-II superconductors where the external magnetic field expels the condensate from regions confined within vortex cores. One novelty of this work is that the condensate is shown to be unstable against phase segregation in the absence of an applied field, even at $T \ll T_c$. Furthermore, the spatial extent of inhomogeneities is distinct from any length scales characterizing the superconducting state of cuprates. However, a competing state is also of magnetic origin and is most likely related to unidirectional spin density waves [22] which show similar doping patterns with superconducting anomalies (Fig. 3). Direct monitoring of the superconducting condensate via the Josephson probe reported here suggests the notion of inhomogeneous superconductivity in cuprates. A complete theoretical understanding of the observed effects requires a proper account of proximity effects near the boundaries of regions within the CuO_2 planes where superconductivity is depleted.

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