Optical melting of the transverse Josephson plasmon: A comparison between bilayer and trilayer cuprates

W. Hu,^{1,*} D. Nicoletti,¹ A. V. Boris,² B. Keimer,² and A. Cavalleri^{1,3,†}

¹Max Planck Institute for the Structure and Dynamics of Matter, Center for Free Electron Laser Science, 22761 Hamburg, Germany

²Max Planck Institute for Solid State Research, 70569 Stuttgart, Germany

³Department of Physics, Clarendon Laboratory, University of Oxford, OX1 3PU Oxford, United Kingdom

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We report on an investigation of the redistribution of interlayer coherence in the trilayer cuprate $Bi_2Sr_2Ca_2Cu_3O_{10}$. The experiment is performed under the same apical-oxygen phonon excitation discussed in the past for the bilayer cuprate $YBa_2Cu_3O_{6.5}$. In $Bi_2Sr_2Ca_2Cu_3O_{10}$, we observe a similar spectral weight loss at the transverse plasma mode resonance as that seen in $YBa_2Cu_3O_{6.5}$. However, this feature is not accompanied by the light-enhanced interlayer coherence that was found in $YBa_2Cu_3O_{6+x}$, for which the transverse plasma mode is observed at equilibrium even in the normal state. These new observations offer an experimental perspective in the context of the physics of light-enhanced interlayer coupling in various cuprates.

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I. INTRODUCTION

In various cuprate superconductors, resonant excitation of specific phonon modes was found to induce transient states with exotic optical properties. These have been interpreted as indicative of light-induced superconductivity far above the equilibrium transition temperature. For single-layer compounds like La_{1.675}Eu_{0.2}Sr_{0.125}CuO₄, the excitation of an in-plane Cu-O stretching phonon was shown to enhance *c* axis coherent coupling [1]. Importantly, a plasma mode appeared at approximately the same frequency (~65 cm⁻¹) as that seen in the superconducting state of the optimally doped single-layer cuprate La_{1.84}Sr_{0.16}CuO₄. The transient *c* axis coherence in La_{1.675}Eu_{0.2}Sr_{0.125}CuO₄ was observed up to the charge-ordering temperature [1] and was interpreted as a sign of light-induced superconductivity [2] after optical melting of stripes [3].

Similarly, in the bilayer cuprate YBa₂Cu₃O_{6.6}, for which hole doping is also near 1/8, excitation of the apical oxygen phonon resulted in a strengthened interlayer superconducting coupling below the equilibrium transition temperature ($T_c =$ 61 K) and in the appearance of a plasma resonance above T_c [4]. This phenomenon was directly correlated to the melting of charge order in the same material [5], in analogy with the physics of La_{1.84}Sr_{0.16}CuO₄.

More difficult to understand are the results at lower doping levels, in YBa₂Cu₃O_{6.5} and YBa₂Cu₃O_{6.45}, for which the lightinduced plasma edge appeared at even higher temperatures, even above the equilibrium charge-ordering temperature. Light-induced interlayer coupling was detected up to temperatures that are in fact rather reminiscent of the equilibrium pseudogap (T^*). Further experimental work in YBa₂Cu₃O_{6.5} has associated the appearance of a low frequency interbilayer plasma mode with a partial melting of the intrabilayer plasmon at high frequency, as if coherence were transferred from the CuO₂ bilayer to the interbilayer region of the unit cell [6]. This hypothesis was qualitatively corroborated by observations made with ultrafast x-ray scattering, which indicated a rearrangement of the unit cell atoms that involve transient expansion (compression) of CuO_2 intrabilayer (interbilayer) distances [7].

Hence, the physics of such light-induced superconductivity appear to be more general than optical melting of stripes. This notion was further confirmed by the observation of a related transient phase in the organic material K_3C_{60} [8].

All these experimental observations are now the basis for a number of theoretical interpretations of the light-stabilized superconductivity in cuprates, including transient suppression of competing order [9,10], redistribution of phase fluctuations between interbilayer and intrabilayer Josephson junctions [11], parametric cooling [12,13], electron-phonon coupling [14], and phonon squeezing [15,16], for which no comprehensive picture exists.

Here, we extend our analysis to another non-chargeordered, multilayer cuprate, a useful comparison for the data in YBa₂Cu₃O_{6.5}. We study the response of trilayer Bi₂Sr₂Ca₂Cu₃O₁₀ (underdoped, with midpoint $T_c = 99$ K and $\Delta T_c = 10$ K), under similar driving.

We report a transient high-frequency spectral weight loss at the transverse plasma mode. However, in this compound, the intertrilayer coherence was not enhanced at any temperature, and the response at the transverse plasma mode disappeared immediately above the superconducting transition temperature. Different scenarios are analyzed to rationalize this observation.

II. CRYSTAL STRUCTURE AND EQUILIBRIUM OPTICAL PROPERTIES

In Fig. 1(a), we show the structure of $YBa_2Cu_3O_{6.5}$, which exhibits two CuO_2 planes in one unit cell. Each copper atom in the plane is coordinated with four oxygen atoms in the same layer and with one apical-oxygen atom in the perpendicular direction. In $Bi_2Sr_2Ca_2Cu_3O_{10}$, the structure contains three closely spaced CuO_2 planes and no Cu-O chains. The copper atoms in the two outer CuO_2 planes are fivefold coordinated with oxygen atoms as in $YBa_2Cu_3O_{6.5}$, whereas the copper

^{*}wanzheng.hu@mpsd.mpg.de

[†]andrea.cavalleri@mpsd.mpg.de



FIG. 1. Multilayer cuprates: crystal structure and *c* axis optical response at equilibrium. Crystal structures of (a) bilayer YBa₂Cu₃O_{6,5} and (e) trilayer Bi₂Sr₂Ca₂Cu₃O₁₀, displaying different CuO₂ layers and apical oxygen atoms per unit cell. (b) and (f) Equilibrium reflectivity, (c) and (g) energy loss function, and (d) and (h) optical conductivity of both samples, displayed in different frequency ranges to highlight the interbi(tri)layer, intrabi(tri)layer, and transverse Josephson plasma modes, respectively. Cartoons show the currents involved for each of these modes [23]. Data in (c) and (d) are taken from Ref. [30], while those in (g) and (h) have been measured with ellipsometry on the same Bi₂Sr₂Ca₂Cu₃O₁₀ sample at the infrared beamline of the synchrotron radiation source ANKA in Karlsruhe. Reflectivities in (b) and (f) were determined by time-domain terahertz spectroscopy in the same setup used for our pump-probe measurements. The purple line in (f) is a fit to the data with a JPR at $\omega = 5 \text{ cm}^{-1}$.

atoms at the inner CuO_2 plane only bind to four oxygens [Fig. 1(e)]. Previous studies have shown that the doping level is not homogeneous in these three planes, with the outer CuO_2 planes being more doped [17,18] and exhibiting smaller superconducting gaps than the inner CuO_2 plane [18].

The *c* axis equilibrium reflectivity of both samples is shown in Figs. 1(b) and 1(f) in the spectral range below 80 cm^{-1} . In YBa₂Cu₃O_{6.5}, the superconducting transition is evidenced by the appearance of an interbilayer Josephson plasma resonance (JPR) at ~30 cm⁻¹, as already reported in Refs. [4,6]. In Bibased cuprates, the highly anisotropic crystal structure strongly suppresses the interlayer Josephson coupling strength [19], thus resulting in typical plasma edges in the gigahertz frequency range. This is also the case for Bi₂Sr₂Ca₂Cu₃O₁₀, where signatures of a JPR are only found at <10 cm⁻¹.

The broadband *c* axis optical conductivity and energy loss functions, measured for both samples at equilibrium, are reported in Figs. 1(c), 1(d), 1(g), and 1(h). Both bilayer YBa₂Cu₃O_{6.5} and trilayer Bi₂Sr₂Ca₂Cu₃O₁₀ show incoherent *c* axis responses with low conductivity. Infrared-active phonons appear as peaks in $\sigma_1(\omega)$, at frequencies ranging from 100 to 650 cm⁻¹. These modes sharpen with decreasing temperature. In YBa₂Cu₃O_{6.5}, two inequivalent apical oxygen positions result in two apical oxygen phonons around 550 and 630 cm^{-1} [Fig. 1(d)]. A broad conductivity peak appears around 400 cm⁻¹ with decreasing temperature [shaded area in Fig. 1(d)], causing a strong reduction in the oscillation strength of a 320 cm⁻¹ planar-oxygen bending phonon.

In Bi₂Sr₂Ca₂Cu₃O₁₀, a single apical oxygen phonon is observed at 575 cm⁻¹ [Fig. 1(h)] [20]. In this case, it is the spectral weight of this apical oxygen mode which gets depleted below T_c and transferred to a ~500 cm⁻¹ peak. Similar to the 400 cm⁻¹ mode in YBa₂Cu₃O_{6.5}, this 500 cm⁻¹ peak in Bi₂Sr₂Ca₂Cu₃O₁₀ is associated with a transverse JPR [21], which has been also found in various multilayer cuprates [22].

Analogous modes of nonphononic character are also found in the energy loss function of both $YBa_2Cu_3O_{6.5}$ and $Bi_2Sr_2Ca_2Cu_3O_{10}$ at ~500 cm⁻¹ and ~550 cm⁻¹, respectively [Figs. 1(c) and 1(g)]. These can be identified as intrabi(tri)layer longitudinal plasma modes.

The physics of longitudinal and transverse plasmons can be understood by considering multilayer cuprates as a stack of Josephson junctions, with Cooper pairs tunneling between neighboring CuO_2 layers.



FIG. 2. Apical oxygen excitation in YBa₂Cu₃O_{6.5} and Bi₂Sr₂Ca₂Cu₃O₁₀. (a) and (d) Cartoons of oxygen motions [20,31] under midinfrared resonant excitation. (b) and (e) Energy loss function and (c) and (f) the real part of optical conductivity are displayed for both cuprates at the lowest temperature, along with the spectrum of the midinfrared pump pulses (orange).

In the bilayer case (YBa₂Cu₃O_{6.5}), one can think of two Josephson junctions in one unit cell: An intrabilayer junction between closely spaced planes and an interbilayer junction. Each junction is associated with a longitudinal Josephson plasma mode, which is detected as a peak in the loss function $-\text{Im}[1/\varepsilon(\omega)]$, or equivalently, as an edge in the optical reflectivity. In addition to these two modes, out-of-phase oscillations of the Josephson plasma within and between pairs of copper oxide layers give rise to the transverse plasma mode, revealed by a peak in the real part of the optical conductivity $\sigma_1(\omega)$ [23]. For underdoped YBa₂Cu₃O₆₅, the transverse mode, as well as the intrabilayer longitudinal mode, develops at temperatures much higher than the superconducting transition temperature $T_{\rm c}$, which is suggestive of preexisting interlayer coherence in the normal state [24]. As shown in Figs. 1(c) and 1(d), both plasma modes in YBa2Cu3O6.5 are clearly observed even at $T = 1.2T_{\rm c}$, indeed almost as large as those observed deep in the superconducting phase $(T = 0.2 T_c)$.

In trilayer $Bi_2Sr_2Ca_2Cu_3O_{10}$, the physics of longitudinal and transverse plasma modes is similar to that described above. Indeed, despite the larger unit cell, one can think of the trilayer block composed of three equally spaced CuO_2 planes as a single Josephson junction. The tunneling dynamics is that of a single mode, with the charge in the middle CuO_2 plane not changing significantly in an infrared field [21,25]. Hence, one finds only two longitudinal plasma modes and a single transverse plasma mode in trilayer $Bi_2Sr_2Ca_2Cu_3O_{10}$ [Figs. 1(f)-1(h)], which unlike in YBa₂Cu₃O_{6.5}, all appear exactly at the superconducting transition [21].

Within the Josephson superlattice model, the transverse plasma frequency is defined by $\omega_T^2 = (d_2\omega_{Jp1}^2 + d_1\omega_{Jp2}^2)/(d_1 + d_2)$, where d_1 and d_2 are the thickness of the interbi(tri)layer and intrabi(tri)layer junctions, respectively, and ω_{Jp1} and ω_{Jp2} are the corresponding longitudinal Josephson plasma frequencies.

III. PUMP-PROBE EXPERIMENT

For our pump-probe experiments, we used midinfrared pulses centered at $15 \,\mu m (667 \, \text{cm}^{-1})$ and polarized along the direction perpendicular to the CuO₂ planes to drive the apical oxygen phonons. The excitation pulses were generated by difference-frequency mixing in an optical parametric amplifier, with a 300 fs pulse duration and 4 mJ/cm² pump fluence. Figures 2(a) and 2(d) illustrate the atomic motions in the phonon driven state for YBa₂Cu₃O_{6.5} and Bi₂Sr₂Ca₂Cu₃O₁₀, respectively. A typical spectrum of the midinfrared excitation pulse is plotted together with the equilibrium *c* axis optical response functions in the superconducting state in Figs. 2(b), 2(c), 2(e), and 2(f).

To probe the transient optical properties, we used broadband terahertz pulses generated by laser-ionized gas plasma [26] and detected by electrooptical sampling of the terahertz field either in a gas plasma [6,26] or in a 50 μ m thick *z*-cut GaSe



FIG. 3. Dynamical redistribution of interlayer coherence: the high-frequency response. The real part of the optical conductivity at equilibrium (gray) and at the maximum transient response (dark blue), measured at the lowest temperature, are shown for both samples in panels (a) and (b). Frequency-resolved light-induced changes ($\Delta\sigma_1$) are also displayed as color plots throughout their dynamical evolution. The transient spectral weight redistribution around the transverse plasma mode, quantified as $\int \Delta |\sigma_1(\omega)| d\omega$, shows a longer lifetime in YBa₂Cu₃O_{6.5} (7 ps) than in Bi₂Sr₂Ca₂Cu₃O₁₀ (3 ps). Temperature-dependent measurements are displayed in panels (c) and (d). Data for YBa₂Cu₃O_{6.5} are adapted from Ref. [6].

crystal [27]. The polarization of the terahertz probe pulses was also set to be perpendicular to the CuO_2 planes.

With this technique, we could directly determine the reflected terahertz field for different pump-probe time delays t after photoexcitation. By separately modulating midinfrared pump and terahertz probe and simultaneously recording the signals using two lock-in amplifiers, we retrieved both the pump-induced terahertz field change $\Delta \tilde{E}(\omega,t)$ and the stationary terahertz field $\tilde{E}(\omega,t)$. The complex reflection coefficient of the photoexcited sample $\tilde{r}'(\omega,t)$ was calculated using the relation

$$\frac{\Delta \tilde{E}(\omega,t)}{\tilde{E}(\omega,t)} = \frac{\tilde{r}'(\omega,t) - \tilde{r}^{0}(\omega)}{\tilde{r}^{0}(\omega)}$$

where $\tilde{r}^{0}(\omega)$ is the equilibrium complex reflection coefficient obtained from steady-state optical spectroscopy. In order to take into account the pump-probe penetration depth mismatch, we used a multilayer model [28] for $\tilde{r}'(\omega,t)$, involving a fully photoexcited layer, with thickness equal to the midinfrared pump penetration depth (4 μ m for YBa₂Cu₃O_{6.5} and 6 μ m for Bi₂Sr₂Ca₂Cu₃O₁₀) and complex refractive index $\tilde{N}^{\text{photoexcited}}(\omega,t)$, and a bottom layer in which the refractive index retains the equilibrium value $\tilde{N}^0(\omega)$. By extracting $\tilde{N}^{\text{photoexcited}}(\omega,t)$ from $\tilde{r}'(\omega,t)$ using this multilayer model, we could retrieve the transient optical response functions of the photoexcited layer.

IV. RESULTS AND DISCUSSION

The pump-induced changes to the real part of the optical conductivity $\sigma_1(\omega, t)$, measured in the superconducting state $(T < T_c)$, are plotted for both samples in Figs. 3(a) and 3(b), for frequencies $\omega \gtrsim 150 - 250 \text{ cm}^{-1}$. For YBa₂Cu₃O_{6.5}, the 400 cm⁻¹ transverse plasma mode exhibits a prompt shift to lower frequencies upon excitation, while in Bi₂Sr₂Ca₂Cu₃O₁₀, the 500 cm⁻¹ peak is completely depleted and its spectral weight gets transferred to the apical oxygen phonon at 575 cm⁻¹.

These data suggest that, for both cuprates, pumping the apical oxygen phonon results in a weakening of the intrabi(tri)layer Josephson coupling. The color plots in Figs. 3(a) and 3(b) display the light-induced changes in $\sigma_1(\omega)$ as a function of pump-probe time delay for both samples.



FIG. 4. Dynamical redistribution of interlayer coherence: the low-frequency response. Complex optical conductivities at equilibrium (gray) and at the peak of the light-induced response (dark blue) for (a) YBa₂Cu₃O_{6.5} and (c) Bi₂Sr₂Ca₂Cu₃O₁₀, measured at $T < T_c$. The finite lifetimes of the transient states (7 and 3 ps, respectively) set frequency cutoffs for the transient optical response (outside the shaded areas). Differential complex optical conductivity $\Delta \sigma_{1,2} = \sigma_{1,2}^{\text{transient}} - \sigma_{1,2}^{\text{equilibrium}}$ for (b) YBa₂Cu₃O_{6.5} and (d) Bi₂Sr₂Ca₂Cu₃O₁₀ at $T < T_c$ (dark blue) and $T > T_c$ (red).

As discussed previously, in YBa₂Cu₃O_{6.5}, the plasma mode redshift is observed up to temperatures far above equilibrium T_c [Fig. 3(c)], while for Bi₂Sr₂Ca₂Cu₃O₁₀, this transient spectral weight redistribution occurs only when the experiment is performed below T_c [Fig. 3(d)]. The relaxation dynamics for the two samples are also different: While the transient conductivity changes in YBa₂Cu₃O_{6.5} relax back to equilibrium within about 7 ps, the time scale found in Bi₂Sr₂Ca₂Cu₃O₁₀ is faster (~3 ps).

The dynamics of the inter(bi)trilayer coupling strength was probed in both sets of experiments by measuring the transient optical response at lower frequencies, under the same excitation conditions. Single-cycle terahertz probe pulses were generated using either a ZnTe crystal (for $YBa_2Cu_3O_{6.5}$)

or a photoconductive antenna (for Bi₂Sr₂Ca₂Cu₃O₁₀). These pulses were shone onto the samples with polarization perpendicular to the CuO₂ planes and were then detected by electrooptical sampling in a ZnTe crystal, thus being able to probe the dynamical evolution of the optical response for $\omega \lesssim 80 \,\mathrm{cm}^{-1}$.

As displayed in Fig. 4, in the superconducting state of YBa₂Cu₃O_{6.5}, at the peak of the pump-probe response (t = 0.8 ps), the imaginary part of the optical conductivity $\sigma_2(\omega)$ changes slope and diverges more strongly toward low frequencies, thus suggesting a light-induced enhancement of interbilayer superconducting coupling. Correspondingly, the real part of the optical conductivity $\sigma_1(\omega)$ develops a peak around 50 cm⁻¹ [Fig. 4(a)]. According to our previous studies [4,6], those features can be well fitted by assuming that 20% of the sample has been turned into a transient superconducting state with a stronger interbilayer coupling. Such light-enhanced low-frequency coherence arises at the expense of the spectral weight of the high-frequency transverse plasma mode [6]. A similar effect is also detected in YBa₂Cu₃O_{6.5} in the normal state [Fig. 4(b)], where the measured $\sigma_2(\omega)$ enhancement has been interpreted in terms of a light-induced interbilayer coupling extending far above equilibrium T_c [6].

In contrast with these findings, Figs. 4(c) and 4(d) show that no low-frequency conductivity change could be detected for $Bi_2Sr_2Ca_2Cu_3O_{10}$, at all measured temperatures and time delays, although the transient spectral weight loss at the transverse plasma mode is comparable to the YBa₂Cu₃O_{6.5} case.

Note that the distance between the closely spaced CuO₂ planes in YBa₂Cu₃O_{6.5} and Bi₂Sr₂Ca₂Cu₃O₁₀ are similar, while the interbilayer separation is significantly shorter than the intertrilayer separation (8.4 vs 12 Å) [21,29]. Hence, the tunneling of Cooper pairs may take longer time in the intertrilayer junction in Bi₂Sr₂Ca₂Cu₃O₁₀ than in the case of interbilayer tunneling in YBa2Cu3O6.5. Typical time scales can be estimated from the frequencies of the plasma edge in Figs. 1(b) and 1(f), which are $\sim 30 \text{ cm}^{-1}$ for YBa₂Cu₃O_{6.5} and $\leq 5 \text{ cm}^{-1}$ for Bi₂Sr₂Ca₂Cu₃O₁₀. These correspond to tunneling times of $\sim 1 \text{ ps}$ and $\gtrsim 7 \text{ ps}$, respectively. Hence, the short lifetime (\sim 3 ps) of the spectral redistribution may not be sufficient for the two pairs of trilayer units to become coherent and the respective phases to be correlated in $Bi_2Sr_2Ca_2Cu_3O_{10}$. This observation further underscores the need for long pulse midinfrared excitation to sustain the transient state, an effort that at this stage is still frustrated by current laser technology.

However, we should also notice that the trilayer structure differs from the bilayer one in many other respects, for example, the inequivalent CuO_2 planes and the absence of chain oxygen. The transient structural rearrangements reported for $YBa_2Cu_3O_{6.5}$ [7], which include a decrease in the Cu-apical O distances and an increase in O-Cu-O buckling, leading to an effective hole doping to the planar Cu states, may not

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be reproduced in the case of $Bi_2Sr_2Ca_2Cu_3O_{10}$. In general, our results indicate that a simple stacked Josephson junction structure may not be enough for the modeling of the light-enhanced interlayer coherence in multilayer cuprates.

V. SUMMARY

In summary, we compared the *c* axis transient optical response of bilayer YBa₂Cu₃O_{6.5} and trilayer Bi₂Sr₂Ca₂Cu₃O₁₀ under the same phonon pumping conditions. Both cuprates show a transient spectral weight loss of the transverse plasma mode, suggesting a weakening of the intrabi(tri)layer coupling strength. For YBa₂Cu₃O_{6.5}, this results in a lightinduced strengthening of the interbilayer coherence for YBa₂Cu₃O_{6.5}, while for Bi₂Sr₂Ca₂Cu₃O₁₀, no enhancement in intertrilayer coherence was found. Since YBa₂Cu₃O_{6.5} and Bi₂Sr₂Ca₂Cu₃O₁₀ share many similarities in their stacked Josephson junction structure, apical oxygen phonons, and equilibrium optical properties, their different dynamical response may suggest that additional aspects have to be taken into account for theoretical modeling of light-enhanced interlayer coherence in high- T_c cuprates.

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