Comment on "Phase Diagram of La$_{2-x}$Sr$_x$CuO$_4$ Probed in the Infrared: Imprints of Charge Stripe Excitations"

Recently Lucarelli et al. have reported [1] temperature-dependence of the in-plane optical reflectivity of La$_{2-x}$Sr$_x$CuO$_4$ over a wide doping range, focusing on the infrared peaks at 30 cm$^{-1}$ for $x=0.12$, 250 cm$^{-1}$ and 510 cm$^{-1}$. They interpreted the first peak (30 cm$^{-1}$) as a signature of charge stripe ordering, while the latter two (250 cm$^{-1}$ and 510 cm$^{-1}$) are attributed to the polaronic charge excitations. However, careful readers would notice that the reported spectra are largely different from those so far measured on the same system. As we illustrate below, all these peaks are caused by an uncontrolled leakage of the $c$-axis reflectivity into the measured spectra.

First, we show that the absorption peaks at 250 cm$^{-1}$ and 510 cm$^{-1}$ are nothing but the $c$-axis phonon modes ($A_{2u}$). The reported TO-phonon frequencies for two of the three $A_{2u}$-modes [2] coincide well with the above two frequencies. In Fig.1 we compare the inset spectra of Fig.1c in ref.[1] with the purely $c$-polarized spectrum of La$_{1.96}$Sr$_{0.04}$CuO$_4$. This clearly demonstrates that Lucarelli et al. observed the spectra mixed with the $c$-axis component. A similar mixing is more or less observed in most of their samples except for $x=0$ and 0.26.

Second, the $c$-component mixing seriously affects the reflectivity values below $\sim$200 cm$^{-1}$. At low temperatures, the reflectivity is close to unity for $E \parallel ab$, while it decreases with lowering $\omega$ to less than 0.5 for $E \parallel c$, reflecting the incoherent charge dynamics in the $c$-axis direction. If a $c$-component is mixed into the measured in-plane spectrum, then the measured reflectivity tends to decrease with decreasing $\omega$, creating an artificial new absorption peak in the Kramers-Kronig transformed conductivity spectrum. Note that the spectra reported by the other groups [3,4] do not show such a pronounced peak below 100 cm$^{-1}$. Although Lucarelli et al. mentioned that their spectrum for $x=0.12$ is consistent with the result by Dumm et al. [4], the gigantic peak at 30 cm$^{-1}$ is not seen in the latter.

Finally, we point out a non-systematic doping dependence of reflectivity spectrum seen in Fig.1 of ref.[1]. For example, the 510 cm$^{-1}$ peak that is substantially weakened at $x=0.15$ develops again for $x=0.19$, which strongly suggests an accidental mixing of $c$-component [5]. As to the origin of $c$-component mixing, there are several possibilities such as polarizer leakage, mis-cutting and multi-domain structure of crystals grown by traveling-solvent-floating-zone method. From the non-systematic spectral change with $x$, it is speculated that the source of $c$-component and the mixing rate may be different from sample to sample in ref.[1]. It should be noted that even a small amount of admixture of the $c$-component seriously affects the optical spectrum, whereas neutron scattering and/or transport measurements are more robust against a few percent mixture of different crystal angles.

In summary, the three infrared peaks observed in ref.[1] in addition to the in-plane phonon peaks do most certainly originate from the $c$-axis component mixed into the in-plane spectra. Therefore, neither the $d$-band scenario nor the charge stripe dynamics can be deduced from these experimental results.

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[5] Also, the magnitude of mid-infrared conductivity for $x=0.03$ exceeds that of both $x=0.05$ and 0.07, in stark contrast with the established doping dependence[6].  
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![Fig. 1. Comparison of the reflectivity spectrum of La$_{1.96}$Sr$_{0.04}$CuO$_4$ (thick curve) from Fig.1c inset of ref.[1] with the $c$-polarized spectrum of La$_{1.96}$Sr$_{0.04}$CuO$_4$ (thin curve)[7]](image-url)