Comment on "Evidence for a-b-Plane Coupling to Longitudinal c-Axis Phonons in High-T_c Superconductors"

In a recent Letter [1] Reedyk and Timusk analyzed infrared optical data of a variety of layered high-T_c superconductors. They demonstrated that in these materials a striking correlation exists between the position of "notches" in the a-b reflectivity and the peak positions of the c-axis energy loss function. The authors attribute this correlation to strong coupling of optically forbidden c-axis longitudinal phonons to the in-plane midinfrared continuum. As an alternative explanation we propose that this correlation results from a nonideal optical arrangement.

In the commonly used reflectivity geometry the incident light is not strictly normal to the surface. Also one is usually working with an unpolarized light beam strongly focused onto the sample, with angles of incidence (ϕ) distributed up to 1 sr. As a result the light probes the dielectric functions both in the c direction and in the planar directions. This leads to particularly pronounced effects in anisotropic materials, where strong c-axis oscillators leak into the a-b response. This leakage is further enhanced if the crystals are nonperfect, e.g., due to diffraction or multiple scattering events on a diffusely reflecting surface. To estimate the importance of such effects, a detailed analysis of the optical arrangements and the morphology of the sample surfaces is required. None of the experimental papers cited in Ref. [1] offered such an analysis.

Using the Fresnel equations for anisotropic media [2] we calculated the reflectivity of polarized light with a ϕ of 15, 30, and 45 degrees. We have chosen phonon frequencies such as to mimic the c-axis loss function of Fig. 2(a) of Ref. [1] [3]. The result is displayed in Fig. 1(a). It is striking that dips occur in the reflectivity at the positions of the *longitudinal* modes in the c direction. In Fig. 1(b) we display experimental room temperature reflectivities of an epitaxial thin film of $Tl_2Ba_2Ca_2Cu_3O_{10}$. The polarization dependence of the minima strongly suggests that leakage of the c-axis dielectric function into the planar response is indeed responsible for the minima observed in these materials. The absence of minima for the s-polarized geometry explains in a natural way why no notches were found in the spectra taken in the $\mathbf{q} \perp c$ geometry in Fig. 1(b) of Ref. [1].

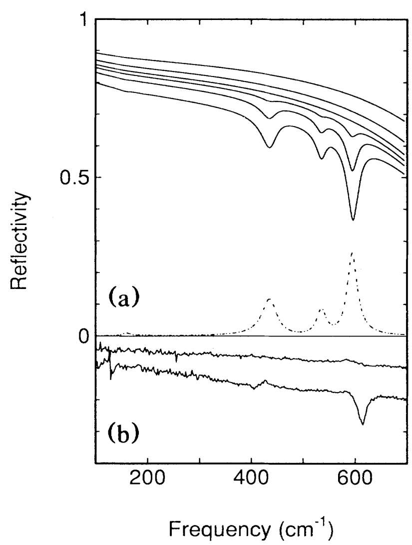

FIG. 1. (a) Theoretical c-axis loss function (dotted, arbitrary units along the vertical axis) and reflectivity (solid). From top to bottom: s-polarized light with a ϕ of 45, 30, and 15 degrees relative to the surface normal. Lowest three curves: p polarized, same angles in reverse order. (b) Reflectivity of $Tl_2Ba_2Ca_2Cu_3O_{10}$ for s (upper curve) and p-polarized light (lower curve) taken with $\phi = 45$ degrees.

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[1] M. Reedyk and T. Timusk, Phys. Rev. Lett. **69**, 2705 (1992).
[2] R. M. A. Azzam and N. M. Bashara, in *Ellipsometry and Polarized Light* (North-Holland Publ. Co., Amsterdam, 1977).
[3] For the planar response we used $\epsilon_\infty = 5.0$, and a plasmon frequency of 2000 cm^{-1} with a decay rate of 200 cm^{-1}. For the c-axis response we used $\epsilon_\infty = 6.2$, and the following phonon transverse frequencies, decay rates, and oscillator strengths: $\omega_j = 329, 506$, and 560 cm^{-1}, $\gamma_j = 40, 20$, and 20 cm^{-1}, and $s_j = 6.5, 0.56, 0.22$.

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