## Phonon anomalies in the infrared conductivity of the RuSr<sub>2</sub>GdCu<sub>2</sub>O<sub>8</sub> ferromagnetic superconductor

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Many aspects of the *c*-axis transport properties of superconducting (SC) bilayer compounds such as YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> (Y-123) have been explained with a model invoking the onset of inter- and intrabilaver Josephson currents. In particular, this model explains a strong anomaly of the oxygen bond-bending phonon mode accompanied by the formation of an additional broad absorption peak in the *c*-axis optical conductivity of Y-123 [1]. The layered ruthenate-cuprate compound RuSr<sub>2</sub>GdCu<sub>2</sub>O<sub>8</sub> (Ru-1212) [2] presents a unique opportunity to develop the model of the bilayer Josephson plasmon for the case when a ferromagnetic moment is in the insulating plane. The present work aims to address this issue by studying the critical behavior in the infrared (IR) dielectric response of Ru-1212 near the superconducting  $T_{SC} = 45 \text{ K}$  and magnetic  $T_M = 133 \text{ K}$ transition temperatures.

Polycrystalline Ru-1212 samples were synthesized by solid-state reaction from high purity RuO<sub>2</sub>, SrCO<sub>3</sub>, Gd<sub>2</sub>O<sub>3</sub>, and CuO powders. The ellipsometric measurements in the region from 50 to  $3000 \text{ cm}^{-1}$  have been performed using a homebuilt setup attached to a "Bruker" IFS 133v spectrometer.

The measured real part of the IR conductivity  $\sigma_1(\omega)$ and that of the dielectric function  $\epsilon_1(\omega)$  of Ru-1212 for different temperatures are shown in Fig. 1. We applied a quantitative dispersion analysis in a Kramers-Kronig consistent way by fitting to the both  $\sigma_1(\omega)$  and  $\epsilon_1(\omega)$  a sum of Lorentzian functions. At room temperature the five distinct IR-active phonon modes at 128, 151, 190, 288, and 650 cm<sup>-1</sup> are superimposed on a featureless electronic background of  $100 \,\Omega^{-1} \,\mathrm{cm}^{-1}$ , consistent with the DCconductivity value. The IR-active phonons observed in the anisotropic ceramics are believed to correspond to the modes polarized along the *c*-axis and can be assigned similarly as reported for the c-axis optical phonons in Y-123 crystals. A comparison with the IR-spectra of Y-123 suggests that the phonon mode at 654 cm<sup>-1</sup> involves primarily the apical oxygen vibrations, whereas that at  $288 \text{ cm}^{-1}$  is related to the

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Fig. 1. Real part of the IR conductivity  $\sigma_1(\omega)$  and the dielectric function  $\varepsilon_1(\omega)$  of Ru-1212 measured at different temperatures.



Fig. 2. Temperature dependence of  $\sigma_1(\omega)$  at the phonon frequencies: successive curves are offset by  $5 \Omega^{-1} \text{ cm}^{-1}$  for clarity.

Cu–O bending mode which involves vibrations of the oxygen ions of the CuO<sub>2</sub> planes. The three low-frequency modes at 128, 151, and 190 cm<sup>-1</sup> are suggested to be assigned to the displacements of Cu, Gd, and Ru, respectively. Fig. 2 shows  $\sigma_1(\omega)$  at the

phonon frequencies in detail for different temperatures. Among all phonon modes, only the apical oxygen mode exhibits the classical inharmonic increase of the eigenfrequency on decreasing temperature, revealing anomalies neither at  $T_{\rm SC}$  nor at  $T_{\rm M}$ . At the same time the low-frequency phonon modes show a characteristic softening at  $T_{\rm SC}$  as observed in Y-123 related compounds.

The most apparent feature related to the onset of superconductivity is the transformation of the bending mode at  $288 \text{ cm}^{-1}$  to a broad band centered at  $308 \text{ cm}^{-1}$ . The transformation of the band profile starts at around 90 K being well below  $T_{\rm M}$ , but also well above  $T_{\rm SC}$ . The most pronounced changes are observed at  $T_{SC}$ . Comparing the temperature dependence of the asymmetric peak in Ru-1212 at  $288 \,\mathrm{cm}^{-1}$  with the data reported by Munzar et al. [1] for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6,45</sub> we find a striking similarity. The gradual onset of the anomalies above  $T_{SC}$ is explained within the model of the bilayer Josephson plasmon as due to the persistence of a coherent superconducting state within the individual copperoxygen bilayers, whereas the steep and sudden changes at  $T_{\rm SC}$  occur when the macroscopically coherent superconducting state forms. The present observation suggests a strongly underdoped character of the bulk superconductivity in Ru-1212 originated in weakly coupled cuprate biplanes.

The most remarkable feature associated with the onset of ferromagnetic order within the Ru–O layers at  $T_{\rm M}$  is the anomalous softening of the mode at 190 cm<sup>-1</sup> observed on decreasing temperature below  $T_{\rm M}$ . This fact indicates that the Ru-related phonon mode is strongly affected by the interaction with the electronic system. The onset of this effect at the magnetic transition temperature can be explained by the renormalization of the electron–phonon coupling when the kinetic energy of the itinerant electrons increases due to the onset of the ferromagnetic correlations in the Ru–O layers. In favor



Fig. 3. Temperature dependences of  $\sigma_1$  measured at  $\omega = 75 \text{ cm}^{-1}$  (solid squares) and  $\omega = 3000 \text{ cm}^{-1}$  (solid circles). The solid line shows the derived function of  $\sigma_1(T)$  at  $\omega = 3000 \text{ cm}^{-1}$ .

of this scenario an increase of  $\sigma_1(\omega)$  spectral weight in the mid-IR range at frequencies  $\omega > 1000 \text{ cm}^{-1}$  is observed on lowering temperature below  $T_{\rm M}$ . Fig. 3 shows indeed that the temperature dependence of  $\sigma_1$ measured at  $\omega = 3000 \text{ cm}^{-1}$  (solid circles) exhibits maximum negative derivative (solid line) right at  $T_{\rm M}$ while the dependence of  $\sigma_1$  at  $\omega = 75 \text{ cm}^{-1}$  (solid squares) reproduces well that of the DCconductivity at  $T > T_{\rm SC}$ . Finally, we note that the behavior of the optical conductivity of Ru-1212 observed in the present study is in a good agreement with the previous report based on conventional reflection measurements [3].

## References

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