

Journal Name

ARTICLE TYPE

Cite this: DOI: 10.1039/xxxxxxxxxx

Supplementary information: Seeing the invisible plasma with transient phonons in cuprous oxide

Laszlo Frazer^{*a}, Richard D. Schaller^{b,c}, Kelvin B. Chang^c, Aleksandr Chernatynskiy^d and Kenneth R. Poeppelmeier^{c,e}

1 Amplitudes of transitions are substantial

To demonstrate that there is a substantial contribution to phonon-to-exciton transitions from each of the two phonons, Figures 1 and 2 show a small sample of the data. The curves show the full model and the model with one phonon deleted. The model is determined from the full transient absorption spectrum of 113,894 data points. In Figure 1, both phonons contribute. In Figure 2, only the B phonon contributes to phonon-to-exciton transitions. Since the B phonon has more energy, it induced absorption further below the bandgap.

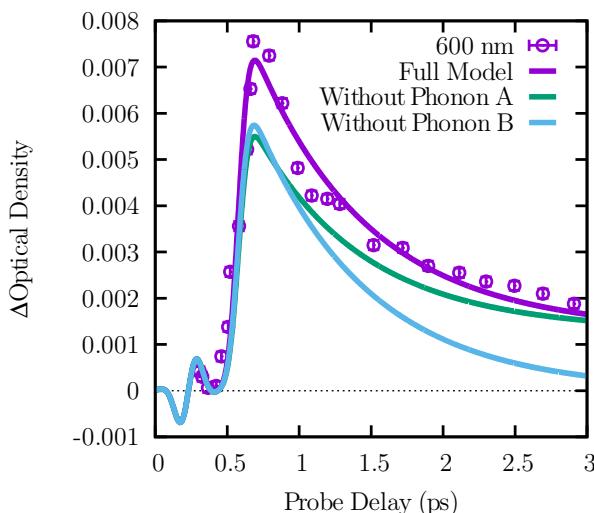


Fig. 1 Points: Transient absorption data at 600 nm taken from Fig. 3 in the main text. Curves: Model, with and without the contributions of the A and B phonons.

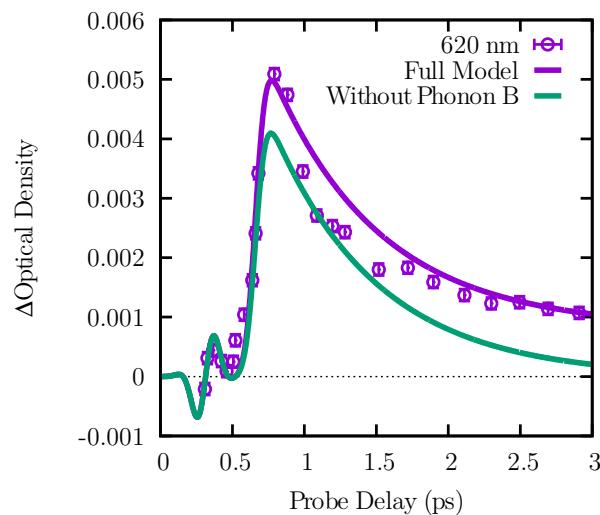


Fig. 2 Points: Transient absorption at 620 nm. Curves: Model, with and without the contribution of the B phonon.

2 Evaluation of calculated standard errors

We calculated lifetimes and standard errors using a regression and 113,894 measurements of transient absorption. To demonstrate that the calculated standard errors are reasonable, we simulated 100 experiments by recalculating the lifetimes and standard errors from random samples of 25,000 (22%) measurements out of the full data set. The results are not normally distributed because time constants are non-negative. The result from a random sample was judged to have a good standard error if the sample time constant was within two sample standard errors of the overall time constant:

$$\tau_{n=113,894} < \tau_{n=25,000} + 2\Delta\tau_{n=25,000} \quad (1)$$

and

$$\tau_{n=113,894} > \tau_{n=25,000} - 2\Delta\tau_{n=25,000}. \quad (2)$$

For τ_A , the two criteria were met 59% of the time. For τ_B , the criteria were met 95% of the time. Therefore we conclude that the standard errors are reasonably reliable. In Figure 3 we show how the calculated value of τ_A varies for different sample sizes.

^a School of Chemistry, UNSW Sydney, NSW 2052, Australia and Department of Chemistry, Temple University, 1901 N. 13th Street, Philadelphia, PA 19122, USA. Tel: 61401 648 058; pccp@laszlofrazer.com

^b Center for Nanoscale Materials, Argonne National Laboratory, 9700 South Cass Avenue, Building 440, Argonne, IL 60439, USA.

^c Department of Chemistry, Northwestern University, 2145 Sheridan Road, Evanston, Illinois 60208-3112, USA.

^d Department of Physics, Missouri University of Science and Technology, 117 Physics Building, Rolla, Missouri 65409-0640, USA.

^e Chemical Sciences and Engineering Division, Argonne National Laboratory, 9700 South Cass Avenue, Argonne, Illinois 60439, USA.

Figure 4 shows the same information for τ_B . If the sample size is less than about 20,000, the model may not converge owing to insufficient information.

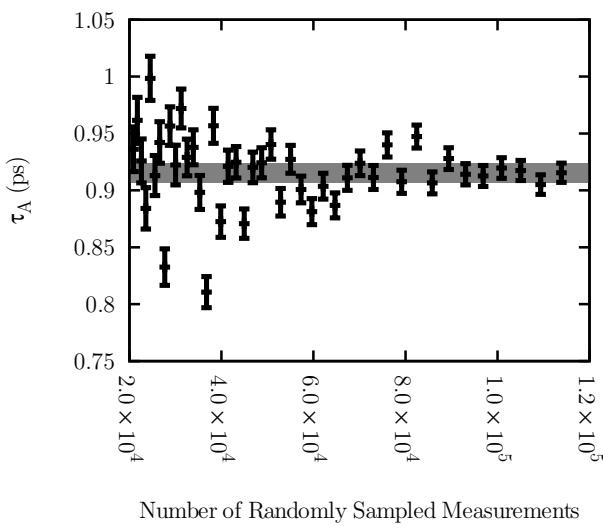


Fig. 3 The value of τ_A computed for various random sample sizes.

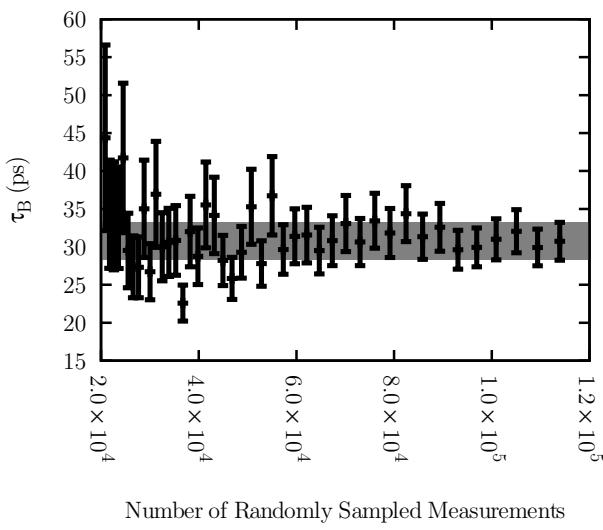


Fig. 4 The value of τ_B computed for various random sample sizes.