

## Thermal Non-equilibrium between Optical and Acoustic Phonons in Graphene Paper under Photon Excitation

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The thermal conductivity and interface thermal conductance of 2D materials are two critical properties of scientific significance and engineering design. In the last decade, Raman spectroscopy has been used widely for measuring them with localized laser heating and Raman-enabled material-specific temperature probing. Under intense photon heating, there is a strong thermal non-equilibrium between optical and acoustic phonon branches. Raman spectroscopy only measures the optical phonon temperature while acoustic phonons are the major player in heat conduction. Such critical effect has not been considered in previous experimental works. Here, the thermal non-equilibrium between optical and acoustic phonons of graphene paper under localized photon heating is studied using electro-thermal and opto-thermal techniques. The optical and acoustic phonon temperature rises are measured under different laser heating intensities. The energy coupling factor ( $E_{pp}$ ) between the in-plane (TO and LO) optical modes and acoustic branches is found in a range of  $(1.59-3.10) \times 10^{15} \text{ Wm}^{-3}\text{K}^{-1}$ , agreeing well with the quantum mechanical modeling result of  $4.1 \times 10^{15} \text{ Wm}^{-3}\text{K}^{-1}$ . Our results show a strong non-equilibrium between phonon branches under small laser spot radii. Under 100 $\times$  objective heating, the optical phonon temperature rise is over 80% higher than that of acoustic phonons. This can significantly affect any thermal characterizations based on opto-thermal methods.