Resonance Raman Signature of Intertube Excitons in Compositionally-Defined Carbon Nanotube Bundles

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Electronic interactions in low-dimensional nanomaterial heterostructures can lead to novel optical responses arising from exciton delocalization over the constituent materials. Similar phenomena have been suggested to arise between closely interacting semiconducting carbon nanotubes of identical structure. Such behavior in carbon nanotubes has potential to generate new exciton physics, impact exciton transport mechanisms in nanotube networks, and place nanotubes as one-dimensional models for such behaviors in systems of higher dimensionality. Here we use resonance Raman spectroscopy to probe intertube interactions in (6,5) chirality-enriched bundles. Raman excitation profiles for the radial breathing mode and G-mode display a previously unobserved sharp resonance feature. We show the feature is evidence for creation of intertube excitons and is identified as a Fano resonance arising from the interaction between intratube and intertube excitons.[1] The universality of the model suggests that similar Raman excitation profile features may be observed for interlayer exciton resonances in 2D multilayered systems.

[1] J. R. Simpson, O. Roslyak, J. G. Duque, E. H. Hároz, J. J. Crochet, H. Telg, A. Piryatinski, A. R. Hight Walker, and S. K. Doorn , Nature Comm. **9**, 637 (2018).
[2] J. Cannon, *et al.*, J. Phys. Chem. B, **116**, 9812 (2012).

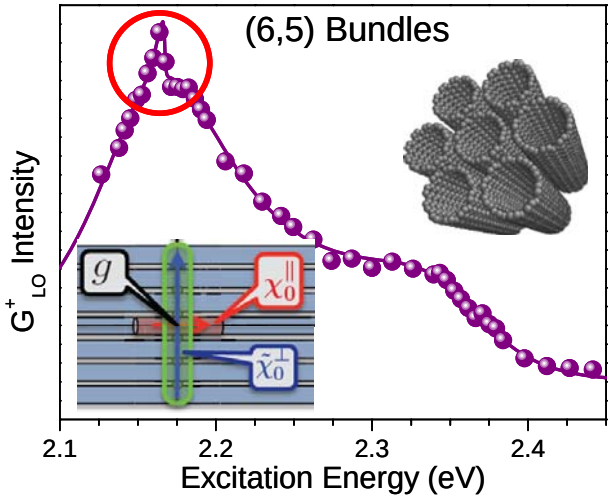

Fig.1 Raman resonant excitation profile of the G^+ mode for the largest bundle size of (6,5)-enriched SWCNTs.[1] Data shown as symbols and fits to the Fano resonance model shown as a solid line. The sharp additional feature at ~2.16 eV (circled) appears for bundled samples and increases with bundle size. Lower-left inset shows a bundle cross-section with the intertube exciton (right arrow) and the intertube exciton (upward arrow). Local coupling *g* between the two types of excitons at the intercept point gives rise to their scattering. A schematic of bundled SWCNTs is illustrated in the upper-right inset.[2]