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Elucidating Energy Relaxation in Single Nanostructures with Ultrafast Microscopy

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The frontier in renewable energy research now lies in designing mesoscale functionalities from nanostructured and molecular components. One key research challenge is to integrate functional entities across multiple length scales to achieve optimal energy flow. To attack the above-mentioned frontier, we aim at unraveling multiscale energy flow across both multiple length and time scales, coupling simultaneous high spatial, structural, and temporal resolution. In my talk, I will focus on our recent progress on unraveling energy relaxation and propagation pathways in single nanostructures with ultrafast optical microscopy. The research presented here is supported by the Solar Photochemistry Program at the Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences.

We have probed environmentally-dependent energy relaxation pathways in single nanostructures by single-particle transient absorption microscopy (TAM).[1-3] Figure 1 shows an example of TAM experiments performed on suspended and substrate-supported graphene on SiO_2.[1] We observed that the hot phonon effect occurs at much lower excitation intensity for suspended graphene compared to substrate-supported graphene. These results show the importance of the environment in controlling the properties of graphene.

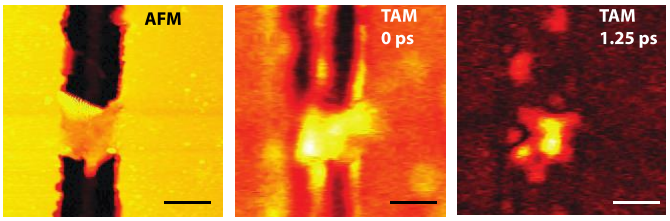

Figure 1 *Correlated AFM and TAM images for suspended graphene at two different delay times. Scale bars: 2 μm*

We have also initiated an investigation into exciton dynamics in graphene-like 2D atomically thin crystals.[2] Femtosecond transient absorption spectroscopy and microscopy were employed to study exciton dynamics in suspended and Si_3N_4 substrate-supported monolayer and few-layer MoS_2 2D crystals. Exciton dynamics for the monolayer and few-layer structures were found to be remarkably different from those of the bulk. Fast trapping of excitons by surface trap states was observed in monolayer and few-layer structures, pointing to the importance of controlling surface properties in atomically thin crystals such as MoS_2 in addition to controlling their dimensions.[2]

Another challenge in nanotechnology research is inhomogenous distributions of size, shape, and surface properties of nanoparticles in as synthesized samples. We have successfully demonstrated structure-specific transient absorption imaging of single-walled carbon nanotubes as a way to investigate intrinsic relaxation pathways.[3] The results show a ~ 40 meV red-shift of the lowest exciton transition, which is attributed to dielectric screening effects by the substrate. Energy relaxation in individual metallic nanotubes was observed with decay constants of a few hundred fs and about 10 ps. We attributed the fast and slow decay components to carrier scattering by optical and acoustic phonons, respectively.

(1) Gao, B.; Hartland, G.; Fang, T.; Kelly, M.; Jena, D.; Xing, H. G.; Huang, L. *Nano Lett* **2011**, *11*, 3184.
(2) Shi, H.; Huang, L.; *et.al.*; *ACS Nano* **2013**, *7*, 1072.
(3) Gao, B.; Hartland, G. V.; Huang, L. *J Phys Chem Lett* **2013**, *4*, 3050.