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Coherent anti-stokes spectroscopy as a probe of chemical disorder in isolated carbon nanotubes TATYANA SHEPS, JORDAN BROCIOUS, ERIC O. POTMA, PHILIP G. COLLINS, University of California, Irvine — We use a third-order coherent anti-stokes (CAS) optical technique to study chemical disorder in individual carbon nanotubes. The CAS response is highly sensitive to this disorder, to the extent that a few chemical defects can appreciably decrease the overall signal. The experiments are performed on individual single- and multi-walled carbon nanotubes (SWNTs and MWNTs) connected in a transistor geometry and subjected to varying degrees of controlled, electrochemical oxidation. The overall CAS intensity can be used to probe the extent of chemical modification, and inhomogeneities along a nanotube resolve local coherent electron density fluctuations. We find that the CAS signal is also strongly affected by substrate interactions: aligned SWNTs grown on single crystal quartz are quenched compared to SWNTs on fused quartz. Finally, CAS spectroscopy on individual SWNTs and MWNTs using picosecond pulses resolves the third-order vibrational signal component at the G-band frequency. The ratio of electronic to vibrational CAS signal components is diameter dependent and in small diameter SWNTs the vibrational component is dominated by the electronic CAS signal. However, in MWNTs, this technique is a first step toward chemically sensitive CARS imaging on a single nanotube level.

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