MID-INFRARED THIRD HARMONIC GENERATION IN HIGHLY-ALIGNED SINGLE-WALLED CARBON NANOTUBES

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Optical properties of carbon nanotubes (CNTs) have been extensively investigated during the last decade, and much basic knowledge has been accumulated on how light emission, scattering, and absorption occur in CNTs. However, their nonlinear optical properties remain largely unexplored. Due to inversion symmetry, the lowest observable nonlinear processes in achiral ('armchair' and 'zig-zag') CNTs are third order processes. In chiral nanotubes, while $\chi^{(2)}$ is finite, the 50%-50% mixture of right-handed and left-handed nanotubes in a typical macroscopic ensemble of CNTs precludes the observations of second-harmonic generation. Here, we have observed third harmonic generation (THG) from samples consisting of highly aligned CNTs on sapphire with linearly-polarized intense mid-infrared femtosecond radiation. A third harmonic signal was not measured from the sapphire substrate, thus the induced third harmonic signal is generated in the highly aligned CNT sample. Through polarization-dependent third harmonic generation experiments, the nonzero tensor elements of $\chi^{(3)}$ have been extracted. The contribution of the weaker tensor elements to the overall $\chi^{(3)}$ signal has also been calculated to be approximately 1/3 of that of the dominant $\chi^{(3)}_{zzzz}$ component, which is consistent with theory and other measured values.