Surface-enhanced Raman scattering in graphene and carbon nanotubes: Understanding plasmonic enhancement

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Surface-enhanced Raman scattering (SERS) describes the giant enhancement of the Raman cross section close to a metal surface. SERS can detect single molecules by optical spectroscopy. Its variant, tip-enhanced scattering (TERS) increases the resolution of Raman imaging down to the molecular limit. Despite the wide-spread use of plasmon-enhanced Raman spectroscopy in imaging and materials characterization its fundamental principles remain debated. The dominant mechanism for SERS is thought to be plasmonic enhancement; the large Raman cross section is explained as an enhancement by the localized surface plasmons of metal nanoparticles.

Here we present our results on plasmon-enhanced Raman scattering in graphene and carbon nanotubes. The enhancement is induced by the plasmons of well-controlled lithographically prepared gold nanodimers. Mechanically exfoliated graphene is deposited in a dry transfer process, whereas carbon nanotubes are placed by dieelectrophoresis. This setup allows to study the fundamental of SERS. We present results of the polarization and wavelength-dependence of plasmonic enhancement. The enhancement is extremely strongly localized in space (10 nm) and frequency (15 meV). We propose a new theory of SERS, where the plasmon forms an integral part of a higher-order Raman process and discuss its consequences for SERS in molecules and artificial nanostructures.