

Nonlinear Effects with Plasmonics at IR Wavelengths

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Abstract: Raman spectroscopy is a well-established tool providing for molecular finger prints. The frequency of light scattered off a vibrating molecule is shifted with respect to the incident light frequency. That shift is related to the vibration frequency and reveals a great deal of information on the vibration modes of the molecule. We demonstrated efficient nonlinear coupling between local vibrating molecules and propagating polariton modes at the vibration frequency (namely, mid-IR frequencies) using suspended graphene waveguides.

Summary: Graphene is a monolayer thick crystal of carbon. Graphene is bio compatible and does not absorb light in the 2-20 micron wavelength range, thus very suitable as IR/THz propagation medium. We fabricated free-standing graphene on periodic metallic structures at the micron scale.

Periodic metallo-dielectric structures have long been used as optical filters in the IR/THz range. Metal mesh structure portray negative refractive index in various frequency ranges. For example, the structure shown in Fig. 1 exhibits an effective negative refractive index at the resonance frequency. The resonance frequency is determined by the structure pitch.

In the experiment, a pump Ar ion laser at 514.5 nm was used to excite the test molecules (B. Megaterium) which were situated on the suspended graphene waveguides. The peak Raman intensity of the test molecules is located at 900 cm^{-1} . The graphene waveguide (approximately 3-4 layers) was coating a copper mesh screen with a $12\text{ }\mu\text{m}$ pitch and $8\times 8\text{ }\mu\text{m}^2$ openings. The laser was focused at the center of the screen opening to a spot much smaller than the screen opening or the screen's pitch.

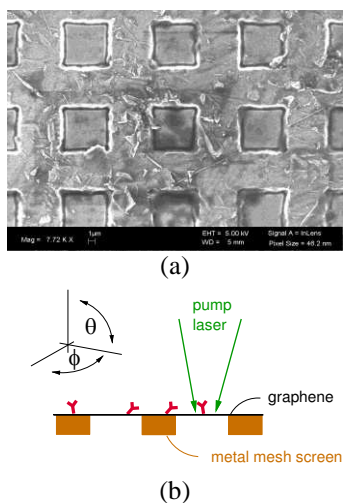


Fig. 1. (a) Graphene-coated metal mesh screen with square openings. (b) The experimental configuration: a test molecule (B. Megaterium) was deposited on the graphene waveguide. We used the 900 cm^{-1} Raman line for interrogation. The sample was tilted and rotated as necessary.

While Raman is quite a local phenomenon, the excitation of a specific vibration mode may be coupled to surface plasmon polariton (SPP) mode in the graphene-waveguide. This surface mode is at the IR wavelength range. The mode is sensitive to the structured waveguide and, hence at resonance, will be reflected back and forth

by the periodic structure and contribute to the Raman signal.

In Fig. 2 we plot the peak Raman intensity at 900 cm^{-1} as a function of the laser pump intensity for various tilt angles. The data exhibit nonlinear dependence at tilt angle of 4 degrees while the dependence at all other angles exhibited linear behavior. Similar data, albeit for different set of tilt angles, could be measured for the graphene itself at 1600 cm^{-1} .

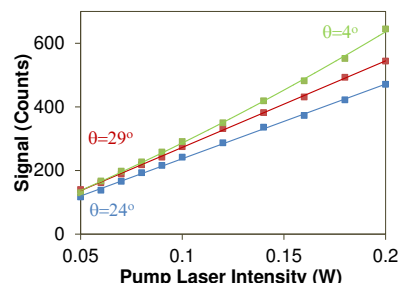


Fig. 2. Peak Raman intensity as a function of pump laser intensity for various tilt angles.

In summary, graphene was shown to be a useful waveguide, which supports polariton modes in the infrared wavelength range.