Nonlinear optical properties of nanostructures

H. Grebel

*Electronic imaging Center at NJIT, Newark, NJ
07102. T: (973) 506-3538; grebel@njit.edu*

Keywords: Nonlinear nanostructures;

Ordered arrays of nanosize spheres (photonic crystals) attracted the attention of many researchers for their linear and nonlinear properties. Here we briefly describe two such applications: 1. Novel periodic structures for spectral identification and in particular, use of Surface Enhance Raman Spectroscopy (SERS) and 2. Nonlinear optical properties of nanostructures imbedded with nano-size semiconductor clusters ('nano within nano').

Surface-Enhanced Raman Scattering (SERS) is a modified version of Raman spectroscopy; the usually weak signal of a non-resonant, spontaneous Raman line is amplified via coupling of the Raman-active, optical phonons to localized electric fields. The probed molecule or, solid is adsorbed on a rough metallic silver, gold or, copper surface in order to enable such amplification [1]. Gratings are often used to couple the pumping laser light to surface charge waves (surface plasmons), thus, exciting localized electric fields. Shown in Fig. 1. is a substrate made of an ordered array of silica spheres (opal) and coated with silver. A layer of Single Wall Carbon Nanotubes (SWCNT) covers the substrate.
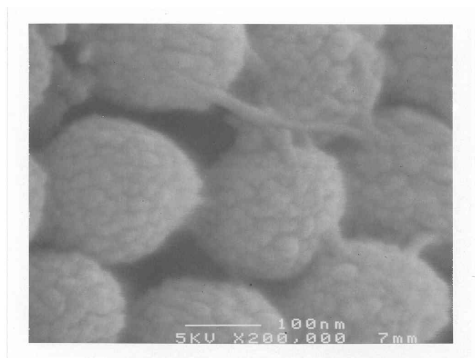

Fig. 1. SWCNT on top of silver coated substrate made of an ordered array of silica spheres.

The Raman signature of such an arrangement depends on the crystallographic orientation of the ordered array of the 'crystal' with respect to the polarized laser beam. Shown in Fig. 2 is the Raman signal at on- and off-resonance conditions.
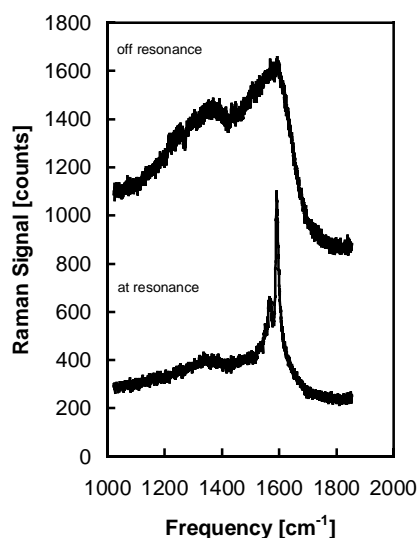

Fig. 2. Raman signal from a single wall nanotube excited at on and off-resonance condition

Using such three-dimensional, periodic structures one is able to design and realize imaging elements thinner than the propagating wavelength [2]. The optically confining environment of these structures is particularly attractive when it is implanted with semiconductor [3] or erbium [4]. Upon annealing, ion-implanted semiconductors precipitate into small nanoclusters hence the name 'nano within nano'. The combination of nonlinear properties of these quantum dots with the strong dispersion of the matrix leads to controllable pulse broadening or even pulse compression [5]. Nano-size silicon clusters (Si-nc) exhibit strong nonlinearity around 2.4 eV, which may be attributed to indirect transition to surface states. In contrast, Ge nanoclusters (Ge-nc) are featuring several direct transitions in the wavelength region between 0.5 μm to 1.8 μm. Of particular interest is the transition around 2.2 eV, which is size independent owing to mid-Brillouin transitions [6]. There exists a 'magic angle' where large dispersion occurs: the nonlinear transmission dramatically varies near this angle. Shown in Fig. 3 is the transmission change as a function of incident angle for various intensities. The nonlinear transmission is attributed to nonlinear diffraction as a result of an intensity dependent index of refraction. Photon localization in the periodic structure in addition to near resonance transition result in an index change of $\Delta n=0.25$ at relatively modest intensities of a few tens of MW/cm^2.
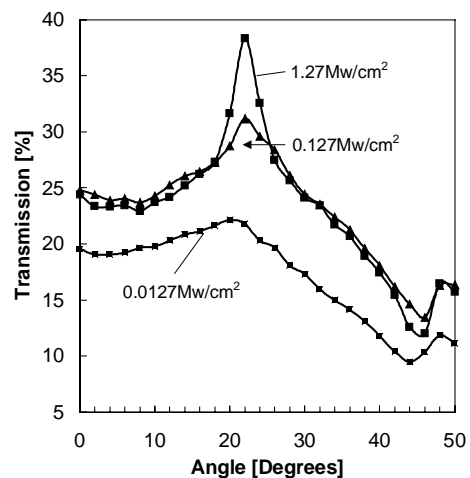

Fig. 3 Nonlinear transmission for a sample of Ge-implanted, ordered array of 300-nm size silica spheres, near the 'magic angle' of $\theta=22^o$ at various intensities.

References
[1] M. Fleischmann, P. J. Hendra, A. J. McQuillan, Chem. Phys. Letters 26, 163 (1974).
[2] J. M. Tobias and H. Grebel, *Opt. Letts.*, **24**, 1660 (1999).
[3] M. Ajgaonkar, Y. Zhang H, Grebel and C. W. White, *Appl. Phys. Letts*, **75**, 1532 (1999).
[4] M. Ajgaonkar, Y. Zhang, H. Grebel, M. Sosnowski and D. Jacobson, *Appl. Phys. Letts*., **76**, 3876 (2000).
[5] S. Vijayalakshmi, H. Grebel, G. Yaglioglu, R. Dorsinville and C. W. White, *Appl. Phys. Letts*, **78**, 1754 (2001).
[6] J. R. Heath, J. J. Shiang and A. P. Alivisatos, *J. Chem Phys*., **101**, 1607-1615 (1994).