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Electromagnetic interaction between a laser beam and semiconductor nanowires deposited on different substrates: Raman enhancement in Si Nanowires

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ABSTRACT

Raman scattering of Si nanowires (NWs) presents antenna effects. The electromagnetic resonance depends on the electromagnetic coupling of the system laser/NW/substrate. The antenna effect of the Raman signal was measured in individual NWs deposited on different substrates, and also free standing NWs in air. The one phonon Raman band in NWs can reach high intensities depending on the system configuration; values of Raman intensity per unit volume more than a few hundred times with respect to bulk substrate can be obtained.

INTRODUCTION

Semiconductor NWs are attracting a great deal of attention as the building blocks of the future nanodevices. Most of the useful properties of NWs arise from the dimension of its diameter, as compared to characteristic lengths, e.g. exciton Bohr radius, phonon mean free path, wavelength of the incident electromagnetic waves… One of their most interesting properties concerns its ability to enhance the optical absorption; because of the dielectric mismatch between the NW and its surrounding medium, it gives absorption resonances for certain NW diameters when interacting with an incident electromagnetic wave, e.g. illumination with an external light source [1,2]. Recently, antenna effects have been reported in the photocurrent response of Ge NWs [3]. This means that the electromagnetic field inside the NW can be enhanced for predetermined wavelengths by tuning the NWs diameter. This behaviour should permit the development of devices, such as efficient light sources, solar cells, and sensitive photodetectors among other. The NWs diameter dependent absorption resonances occur when the light wavelength is commensurate with the diameter size of the NW [3,4]. The study of this interaction is very important for selecting the NWs dimension for the desired application. The problem can be treated by numerical methods solving the Maxwell equations inside the NW. The measurement of the absorption of NWs is not an easy task; however, Raman spectroscopy can be an alternative method to experimentally study the optical resonances in semiconductor NWs, as well as the response of the NWs to the light polarization [4,5]. We present herein an analysis of the interaction of a laser beam with Si NWs using finite element methods for solving the Maxwell equations, considering different substrates supporting the NWs. Experimental microRaman measurements on individual Si NWs deposited on different substrates are used for illustrating the resonance effects.

METHODS AND SAMPLES

The Si NWs were grown by the vapor-liquid-solid (VLS) method, using colloidal Au as a catalytic metal, in a low pressure chemical vapor deposition (LPCVD) reactor. The growth was carried out at 500 ºC using Si_2H_6 as a precursor gas with a 10 sccm flow; N_2 was used as the carrier gas with a flow of 90 sccm, and a total pressure of 400 mTorr was kept in the chamber.

The interaction between the Gaussian laser beam power distribution and the NW was modelled by solving the Maxwell equations using finite element methods (fem). Raman spectra of the NWs were recorded with a Labram UV-HR 800 Raman spectrometer from Jobin Yvon. The excitation and the scattered light collection were performed by means of a confocal metallographic microscope with a high magnification objective (X100) with 0.95 numerical aperture (NA). Either He-Ne (632 nm) or frequency doubled YAG laser (532 nm) excitation sources were used. The laser beam diameter at the focal plane is ≈1 μm, which is several times larger than the typical NW diameter. The NWs were mounted in rotating stages for studying the light polarization effects. Different substrates supported the NWs, e.g., Al, Ge, and also free standing NWs were studied.

RESULTS AND DISCUSSION

The Raman signal was observed to be strongly dependent on the substrate on which the NWs were deposited for the measurements, also the orientation of the NW axis with respect to the light polarization was observed to have a strong influence on the Raman signal. The NWs were first observed at the scanning electron microscope (SEM), from where they were transferred to the optical microscope attached to the Raman spectrometer. The diameter of the NWs ranges from 30 to 150 nm, well below the diffraction limit for the observation in the optical microscope, and also significantly smaller than the wavelength of the laser used in the Raman experiments.

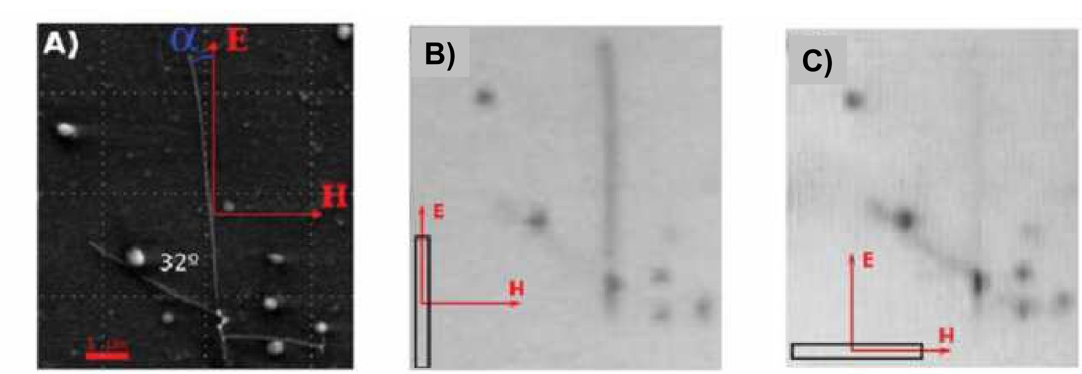

Fig.1. A) SEM image of Si NWs, B) optical microscope image of the NWs for TM polatization with respect to the NW axis, C) optical microscope image for TE polarization.

The NWs standing on the Al substrate were observed in the optical microscope when oriented parallel to the light polarization, while when rotated 90º they were almost unappreciable, Fig.1. Note that the diameter observed in the optical microscope is larger than the true diameter observed in the scanning electron microscope (SEM), because it is a light dispersion image.

Modelling the electromagnetic interaction between the laser beam and the NW demands the solution of the Maxwell equations for the electromagnetic wave propagating across the NW and its surrounding medium. In this context the medium surrounding the NW is critical to the solution of the problem. Normally, the NWs are separated from their growth substrate and are placed in a hosting substrate. One has to consider the role played by this substrate in the solution of the interaction of the NW with the electromagnetic field. Instead of using discrete dipole approximation (DDA) or finite difference on time domain (FDTD) methods we used finite element methods (fem), using COMSOL multiphyisics software package. The problem is modelled in two dimensions, for this one considers ideally long NWs neglecting the influence of the NW ends. The incident laser is polarized in TEM00 mode, and focused onto the NW with a large numerical aperture objective, in the same conditions as a typical microRaman experiment; therefore the laser at focus presents a Gaussian distribution power profile with a diameter ≈1 μm. The light is linearly polarized with the electric field aligned parallel to the NW axis (TM). The NW was either deposited on a substrate or standing in air. The domain of simulation was large enough to avoid the influence of the borders; furthermore, it was surrounded by cartesian perfect matching layers (PMLs), which absorb the totality of the incoming radiation, thus eliminating the secondary reflections. Two wavelengths were studied, 532 nm and 632 nm, which are typical excitation laser lines used in the Raman experiments.

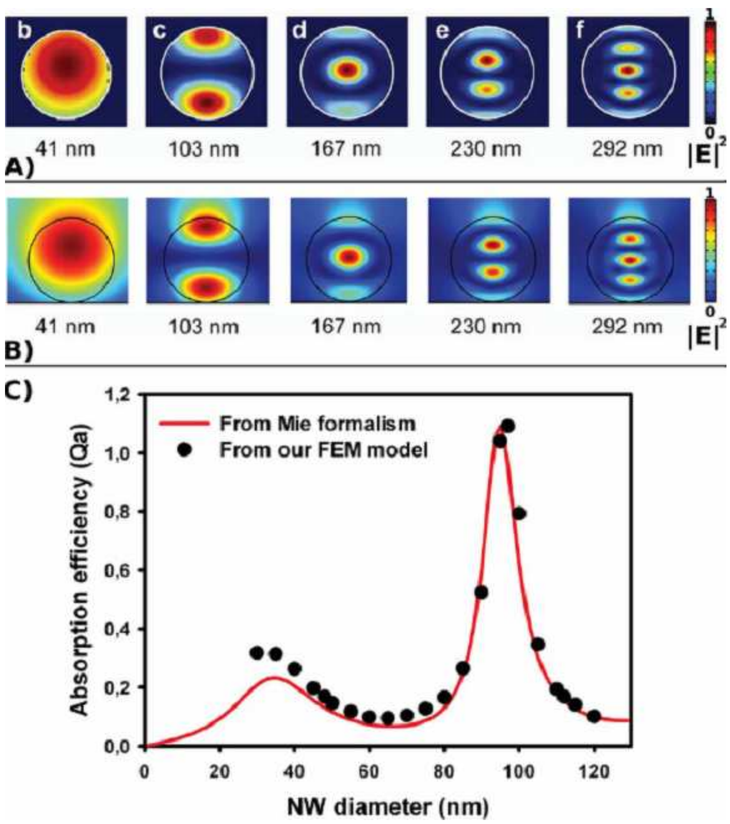

Fig.2. A) Normalized $|E|^2$ calculated by FDTD in ref [5] for different NW diameters deposited on Au, λ=532 nm, B) Normalized $|E|^2$ calculated by FEM, C) Absorption efficiencies calculated by FEM, and using the Mie formalism.

The results obtained in the simulations were compared to the results obtained using the the FDTD method in NWs with the same diameter deposited on gold in ref. [5]. One observes a very good agreement concerning the distribution of the electric field inside the NWs. Another test of the method was done by comparing the absorption efficiencies estimated by integrating the normalized value of $|E|^2$ for NWs standing in air, with the Mie solutions of the Maxwell equations [6], with a satisfactory result in the range of the NW diameters analyzed; Fig. 2C.

The tunability of the optical absorption by NWs requires of the control of the different aspects contributing to enhancing the optical absorption of the desired wavelengths [7]. Using the FEM model for the calculation of the electric field inside the NW permits to analyze the impact of the different factors contributing to the enhancement of the electromagnetic field; in particular, the NW diameter, the substrate, the light wavelength and polarization, and the NW nature.

The effect of the NW diameter is clearly revealed in Fig.2. It determines for a given wavelength the distribution of the electric field inside the NW. By integrating the electric field inside the NW one observes the resonances for certain diameters, which depends on the wavelength. The other relevant factor is the substrate; we solved the problem for different substrates characterized by different refractive indexes, in particular the results obtained for Au, Al, Ge and air are shown in Fig.3, where $|E|^2_{max}$ is plotted vs the NW diameter for a wavelength of 532 nm.

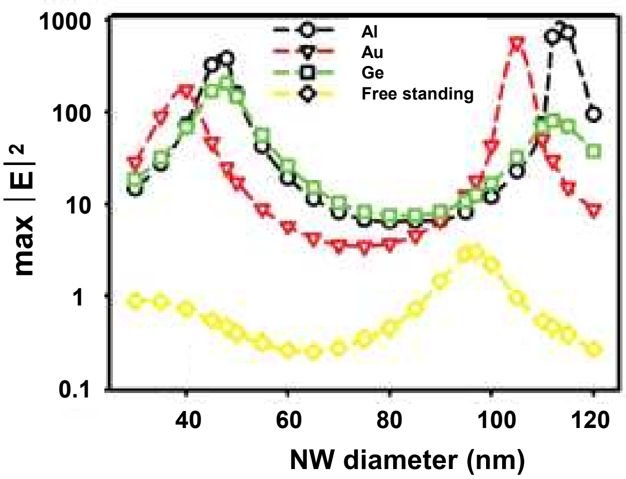

Fig.3. Plot of max $|E|^2$ vs the NW diameter using for NWs deposited on fifferent substrates, and also free standing in air

One observes amplification of $|E|^2$, up to 2 orders of magnitude under resonance conditions. Also, the overall response is enhanced when the NWs are standing on a substrate as compared to the NWs standing in air. The highest values of $|E|^2_{max}$ are obtained for Al. The diameter resonances are also dependent on the substrate where the NWs were deposited; there is a shift to the lower diameters for Au with respect to Al, and further shift to lower diameters for the free standing NW in air, as already shown by Lopez et al [5]. According to this, the optical absorption can be tuned by using different NW diameters and substrates. Therefore, the choice of the substrate is important for optimizing the optical response of the NWs, e.g. Raman scattering [8], photocurrent]3].

The Raman spectra obtained in either a free standing Si NW or a Si NW deposited on an Al substrate are shown in Figs. 4a and b respectively, compared to the spectrum obtained in bulk Si.

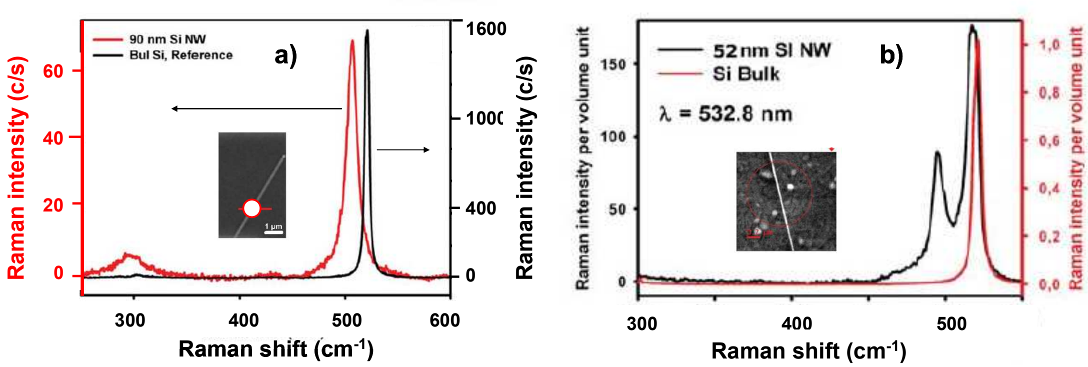

Fig.4. a) Raman spectrum obtained in a free standing Si NW, showing laser induced heating and weak electromagnetic amplification compared with the signal attained in bulk Si. B) Raman spectrum for a Si NW deposited on Al showing strong magnification with respect to the bulk signal

One observes that for the NW in air (90 nm diameter) the Raman intensity is about 30 times lower than the intensity recorded in bulk Si under the same conditions, which gives an intensity per volume unit of around 3 times with respect to bulk Si, Fig.4a; note that the laser power to reach this signal induced heating in the NW, see the broad lineshape and down frequency shift, for more details about the laser induced heating of NWs see [9]; the NW deposited on Al presents an intensity per unit volume about 150 times higher than the intensity per unit volume recorded in bulk Si, Fig.4b. The two diameters selected correspond to resonances in air (90 nm) and on Al (52 nm) according to Fig.3. Note as well that the temperature increase is minimized in this NW.

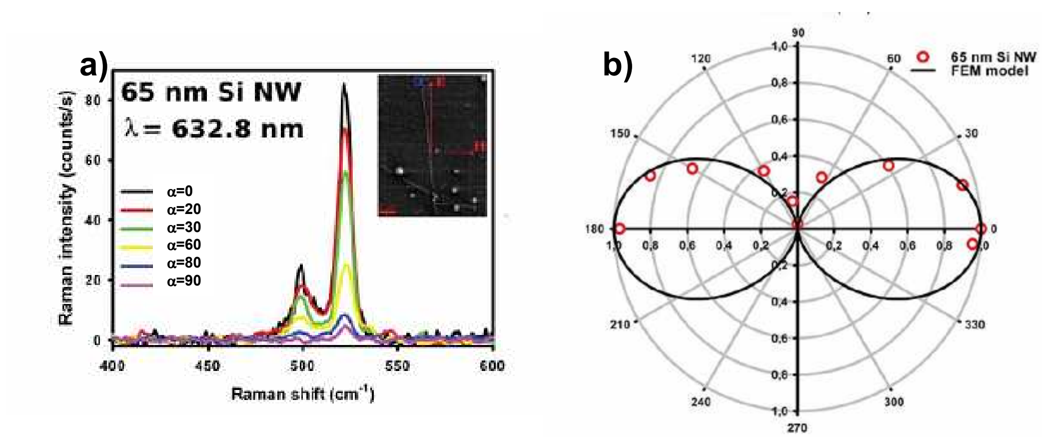

Fig.5. a) Raman spectra for different light polarization angles with respect to the NW axis. b) Polar plots of the Raman intensity / round circles, and the calculated $|E|^2$

The Raman signal was dependent on the light polarization with respect to the NW axis. The Raman spectra obtained on NWs 65 nm in diameter with 632 nm excitation wavelength for different angles between the light polarization and the NW axis are shown In Fig. 5a. The polar plot representing the Raman intensity is shown in Fig. 5b together with the plot of the $|E|^2$ calculated by FEM for such NW in Al substrate, showing the satisfactory fit to the behaviour predicted by the electromagnetic interaction model. Similar results are achieved with SiGe NWs with different compositions.

CONCLUSION

The interaction between group IV NWs and a focused laser beam has been studied by solving the Maxwell equations by FEM. Absorption resonances are demonstrated depending on the NW diameter, the substrate and the laser wavelength. The Raman spectra are used to demonstrate the calculated predictions, showing strong electromagnetic amplification under specific experimental conditions.

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