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Interaction Between a Laser Beam and Semiconductor Nanowires: Application to the Raman Spectrum of Si Nanowires

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ABSTRACT

One presents in this work the study of the interaction between a focused laser beam and Si nanowires (NWs). The NWs heating induced by the laser beam is studied by solving the heat transfer equation by finite element methods (fem). This analysis permits to establish the temperature distribution inside the NW when it is excited by the laser beam. The overheating is dependent on the dimensions of the NW, both the diameter and the length. When performing optical characterization of the NWs using focused laser beams, one has to consider the temperature increase introduced by the laser beam. An important issue concerns the fact that the NWs diameter has subwavelength dimensions, and is also smaller than the focused laser beam. The analysis of the thermal behaviour of the NWs under the excitation with the laser beam permits the interpretation of the Raman spectra of Si NWs, where it is demonstrated that temperature induced by the laser beam play a major role in shaping the Raman spectrum of Si NWs.

KEYWORDS: Si, Nanowires, Raman spectroscopy, thermal conductivity, laser heating, phonons

1 INTRODUCTION

Semiconductor nanowires (NWs) have a strong potential in electronics and optoelectronics; e.g. field effect transistors, interconnects, heterostructured devices, thermoelectric converters, photovoltaics, and sensors (Cui et al 2001, Kayes et al 2005, Hochbaum et al 2008). Besides the quantum confinement effects, relevant for NWs with diameter below -20 nm (Campbell et al 1986, Piscanec et al 2003), the high aspect ratio of NWs with diameters sensibly larger than the limit for quantum effects also present noteworthy changes of the physical properties as compared to the corresponding bulk semiconductors. In particular, the thermal conductivity of NWs is strongly reduced with respect to their bulk counterparts (D.Li et al 2003). This is a very important issue, because the thermal transport is critical for the devices based on NWs. The size reduction leads to an increase in the power density to be dissipated in spite of the lower current operation; therefore, the heat extraction from the active zones of the devices is crucial for achieving high performance and reliable nanodevices.

Characterization techniques are necessary in order to the understanding of the physical properties of semiconductor NWs, which are limited not only by the dimension, but also, by other features as the surface roughness. Optical characterization tools with submicrometer spatial resolution are necessary to boost the understanding of these structures in view of the development of new nanodevice concepts. Lasers focused down to submicrometric probe beams are non invasive excitation sources allowing for the study of the fundamental properties of semiconductors. In particular, photoluminescence (PL) (Liu et al 2010), optical absorption (OA) (Xie et al 2011), Raman scattering (RS) (Piscanec et al 2005, Adu et al 2006, Doerk et al 2009, Torres et al 2010, Soini et al 2010, Alarcón et al 2011), and photocurrent (PC) (Ahn et al 2005), are characterisation tools for diverse semiconductor nanostructures. NWs are one dimensional structures with diameter below the size of the focused laser beam; besides, the laser wavelength is also larger than the NW diameter. Therefore, the interaction between the laser beam and the NWs does not follow the same trends as described for bulk semiconductors. In order to interpret the spectroscopic data, one needs of the understanding of the interaction between the NWs and the laser beam. Another relevant point concerning the dimensions of the NWs, is the small amount of matter probed by the laser beam; which is detrimental to the signal detection. RS is a second order optical process, for which the efficiency is very low, as compared to first order processes, as OA and PL. Usually, the Raman signal of a single NW is insufficient; therefore, one performs the Raman spectrum on ensembles of NWs. When several NWs are studied the optical response arises from a non-homogeneous medium, composed by NWs that can have different dimensions, and are also excited under different conditions as we will discuss later on. On the other hand, recording the Raman spectrum of an individual NW is very challenging, because of the very small volume of scattering.

The semiconductor NWs are systems with poor thermal conductivity, therefore, they are heated by the laser beam, which hinders the interpretation of the experimental results as reported in the literature (Piscanec et al 2005, Adu et al 2006, Doerk et al 2009, Torres et al 2010, Soini et al 2010, Alarcón et al 2011). Therefore, an exhaustive study of the interaction of the NWs with the laser beam, taking account of the energy transferred to the NW is necessary to interpret the spectral data in experiments in which NWs are excited by focused laser beams. Furthermore, the optical measurements can be used as contact less methods for the measurement of the thermal conductivity, for which one needs to have a full description of the interaction between the laser beam and the NW.

We present herein the analysis of the interaction between the laser beam and Si NWs, and the application to the interpretation of the Raman spectra of NWs.

2 ABSORPTION EFFICIENCY OF NWS

The measurement of the optical response of NWs is conditioned by the fact that the exciting laser beam diameter at the focus largely exceeds the NW diameter. To get a reasonably signal, one measures ensembles of NWs, which often present diameter and length distributions. On the other hand, NWs have poor thermal conductivity compared to the bulk semiconductors; besides, the poor thermal contact to the substrate suppresses the heat dissipation and the NWs can reach temperatures well above room temperature, to the extreme that this NW heating by the laser beam can provide the main contribution to the shape of the optical response of the NWs, e.g. the Raman phonon bands are broadened and downshifted as a consequence of the laser induced heating. Besides, one has to consider that the laser beam presents a Gaussian power profile, the diameter of the laser beam being larger than the NWs diameter; therefore, the energy transferred to the NW by the laser beam must depend on the position of the NW inside the laser beam spot; which means that when exciting with the laser beam a bundle of NWs, all of them are not under the same excitation conditions.

On the other hand, because of the sub-wavelength dimension of the NW diameter, and the dielectric mismatch between the NW and the surrounding media, absorption resonances for NW diameters commensurate with the wavelength occur (Doerk et al 2010). It is possible to calculating the absorption efficiency using Mie solutions of the Maxwell equations, allowing us to handle the true energy absorbed by the NW, depending on its diameter. Fig.1 shows the absorption efficiency calculated for two different laser wavelengths, showing resonances for certain diameters.

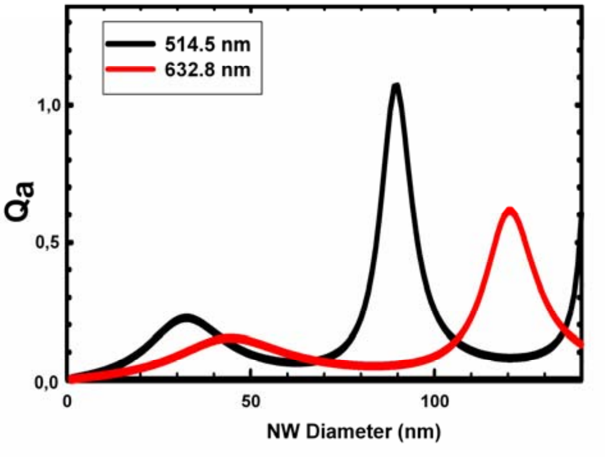

Figure 1: Absorption efficiency for two laser lines (514.5 nm and 632 nm) as function of the Si NW diameter

3 THERMAL TRANSPORT IN THE NW UNDER THE LASER BEAM EXCITATION

The laser beam interacting with the NW acts as a local heat source. The heat generated at the zone of laser impact is distributed over the NW, which the overheating depends on the effective power absorbed, and the thermal conductivity of the NW, which both of them depending on the NW diameter, and the heat dissipation, which is determined by the thermal contact between the NW and the supporting substrate, and the immersion medium; therefore, one should consider the dimension of the NWs and the surrounding medium. All these aspects need to be considered when solving the heat transport equation inside the NW, in order to give the estimation of the temperature inside the NW excited by the laser beam.

The heat transfer equation is solved by finite element methods (fem). The heat source is constituted by the laser energy locally absorbed by the NW. One assumes steady state behaviour, with natural convective/radiative heat exchange between the NWs and the immersion medium, Neumann boundary conditions for free standing NWs, and Dirichlet boundary conditions in the case of NWs with heatsinks. The heat distribution over the NW was calculated as function of the NW dimensions, both length and diameter. First, we study the temperature reached by a Si NW of 37 nm diameter as a function of the incident laser power. The temperature represented in Fig.2 corresponds to the temperature estimated in the zone of impact of the laser beam. One observes that even at very low incident laser powers the temperature is significantly enhanced. This suggests that the micro-Raman spectra of NWs present, even when the laser power is reduced, a non negligible contribution of temperature. It should be noted that the laser beam can hardly reduce down to the limit for negligible heating of the NW, because of the very low Raman signal, which is especially true when measuring individual NWs

Once the laser impacts on the NW, the energy absorbed constitutes a hot source, being the heat distributed over the NW until reaching the equilibrium between the absorbed and dissipated energy, which depends on the NW dimensions. The temperature at the laser impact zone in a free standing NW is plotted in Fig.3 as a function of the NW diameter. Note that the temperature reached by the NW is modulated by the absorption efficiency.

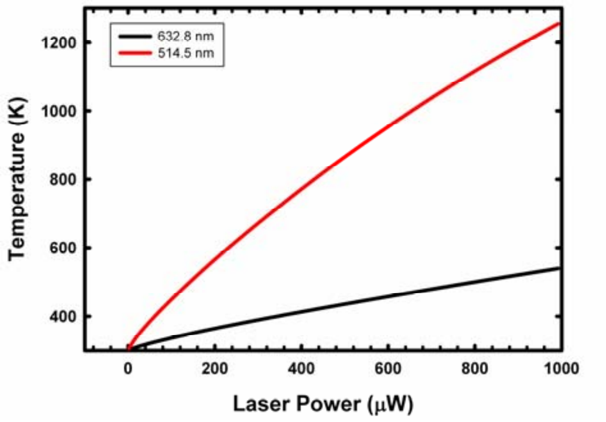

Figure 2: Temperature at the laser impact zone in a NW with 37 nm diameter and 5μm length, for two different laser lines (514 nm and 632 nm)

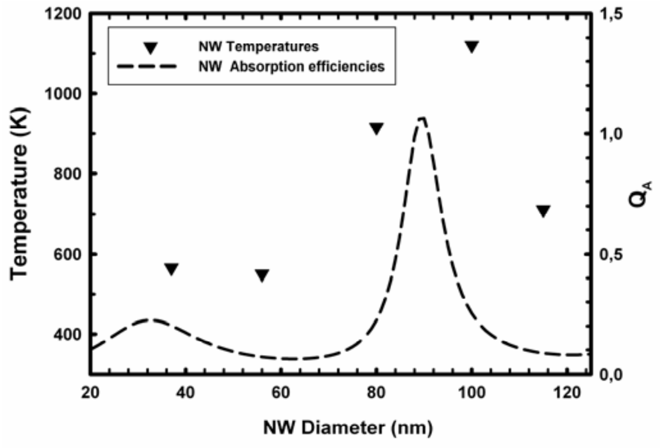

Figure 3: Temperature (symbols) at the laser impact zone as function of the diameter (fixed length of 5 μm), right axis represents the absorption efficiency (discontinuous line), Q_A

If one considers the role of the NW length, one observes that the temperature decreases for longer NWs at equal diameter, because of the larger volume of long NWs. Furthermore, the temperature distribution inside the NW depends on the position along the NW where the laser beam impacts.

A very important aspect concerns the Gaussian power distribution of the laser beam. Because the laser beam spot at focus is larger than the NW diameter, the laser energy absorbed by the NW must strongly depend on the position of the NW inside the focused laser beam spot. This is very important because the NWs diameters are below the diffraction limit, therefore, they cannot be resolved in the optical microscope of the micro-Raman apparatus, which makes difficult to achieve a perfect alignment of the NW with the laser beam. This is illustrated in the temperature reached by the NW when the laser beam is scanned transversally across the NW, Fig.4. The maximum temperature is reached when the NW is in the center of the laser beam spot at the focus plane, being notably reduced when the NW is in the periphery of the laser beam spot, or it is slightly out of focus. This makes sense when measuring ensembles of NWs, because each of the NWs forming the ensemble is excited in a different way in terms of the absorbed energy.

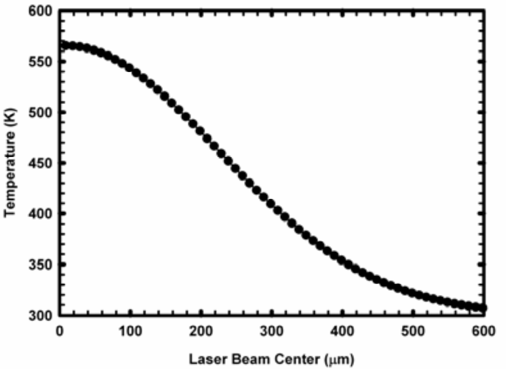

Figure 4: temperature as function of the position of the NW with respect to the laser beam center (the origin is the center of the laser beam)

4 RAMAN SPECTRUM OF NWS

The Raman spectrum of NWs is sensitive to the diameter of the NWs (Campbell et al 1986); the phonon confinement shifts down and asymmetrically broaden the one phonon bands. However, very often shifts of several cm^{-1} have been reported for the LO-TO phonon band of Si NWs, even with diameters larger than 25 nm, which does not match the expected shift predicted by the Richter, Campbell and Fauchet (RCF) model (Campbell et al 1986). According to the discussion of paragraph 3 one can claim that the Raman spectrum of NWs is shaped by the NW laser induced heating. Several Raman spectra obtained on ensembles of Si NWs are shown in Fig.5

One observes spectra with a shift of several cm^{-1} with respect to the control spectrum of a bare Si substrate. Furthermore, one observes an asymmetrical broadening, and even some of them present band splitting. The asymmetry of the Raman bands has been associated with inhomogeneous laser heating (9). The Raman signal arises from the volume where the laser impacts, therefore the temperature gradient of interest to the Raman signal is restricted to the laser beam diameter. This very local gradient has been assumed by some authors as above 300

K/µm (Adu et al 2006); however, there is not evidence that could support this assertion, other than the anomalously large Raman bands observed when measuring ensembles of NWs. One has calculated the temperature distribution inside the NW, varying the thermal conductivity down to the lowest thermal conductivity reported for rough NWs (Hochbaum et al 2008). The temperature gradient at the scale of the laser beam diameter is only a few degrees/µm, even for the NWs with the lowest thermal conductivity; this result rules out the existence of abrupt temperature gradients at the micrometric scale. This is confirmed when one measures the Raman spectrum of an individual Si NW, which is heated by the laser beam, but the Raman band appears symmetric, Fig.6; which rules out the existence of significant temperature gradient inside the scattering volume, in agreement with the calculations.

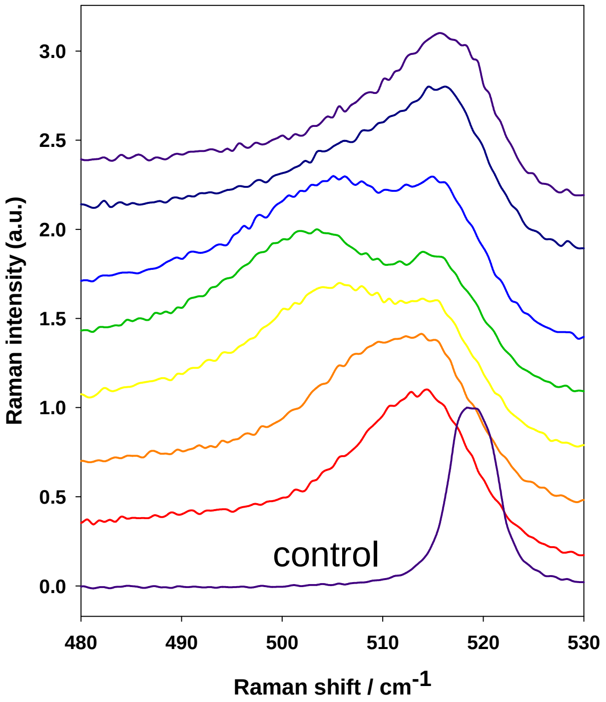

Figure 5: Raman spectra of ensembles of NWs. The control spectrum from a bare Si is also included

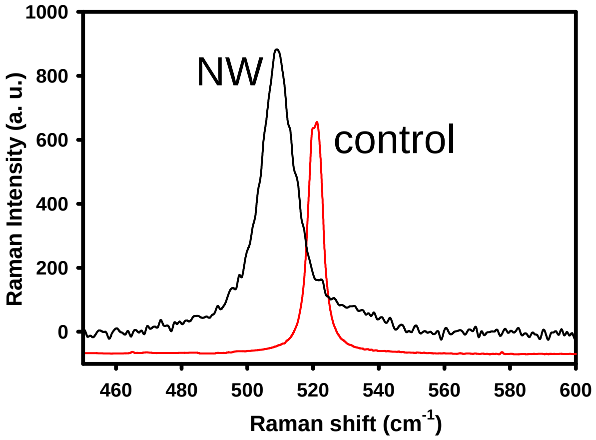

Figure 6: Raman spectrum of an individual NW, and the control spectrum from a bare Si substrate

What is the origin of the strong asymmetry observed in the Raman bands of the ensembles of NWs?; as mentioned above the laser beam spot at focus is much larger than the NWs diameters, therefore, several of those NWs are being excited simultaneously, and they are disposed in different positions inside the laser beam spot, which, because of the Gaussian power distribution, are excited at different powers, thereafter they reach different temperatures; therefore, one can argue that depending on the disposition of the NWs inside the laser beam spot the different NWs can coexist at very different temperatures during the Raman measurement. We have simulated the Raman spectrum of a set of four NWs with diameters 25, 20, 20 and 20 nm respectively, in two different geometric inside the laser beam spot, Fig.7. One observes that the resultant spectra differ for the two configurations. When the temperature reached by the NWs is very different from each other one observes the band splitting experimentally seen in Fig. 5, see Fig.7b. The anomalously large width of the Raman bands is due to the large temperature inhomogeneity, which is consequence of the different temperatures reached by the NWs in different positions with respect to the laser beam axis in the focal plane.

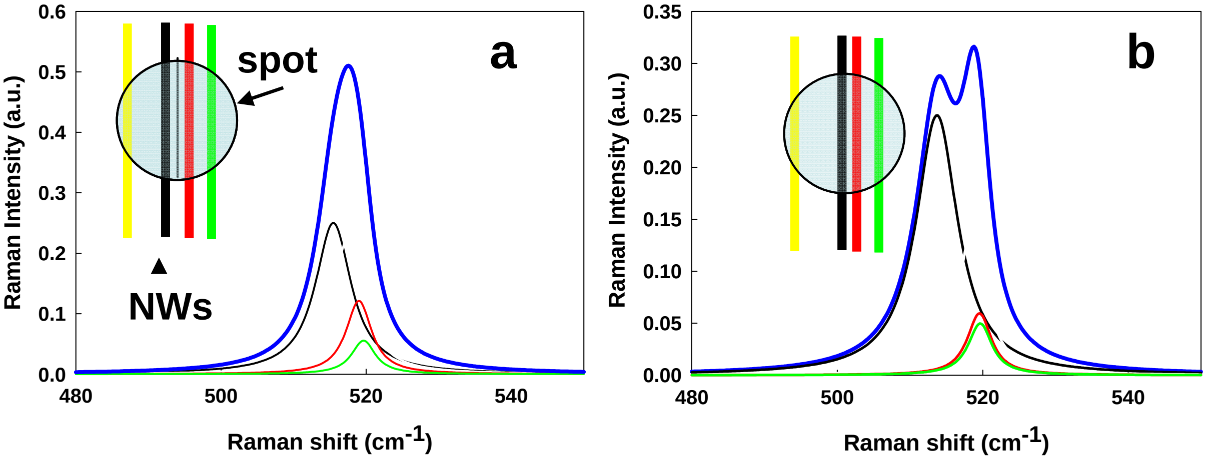

Figure 7: Simulated Raman spectra for each individual NW, and the resultant of the ensemble of four Si NWs (20, 25, 20 and 20 nm diamter from left to right in the inset), and two different distributions of the NWs inside the laser beam spot (see the insets)

5. CONCLUSION

The temperature induced by the interaction between a focused laser beam and Si NWs has been modeled using fem methods. The temperature distribution has inside the NWs has been obtained for different excitation conditions; in particular special interest has been paid to the role of the Gaussian power distribution of the laser beam, and the fact that it is larger by at least one order of magnitude than the common NWs diameters. The results obtained permit a full description of the overheating of the NWs excited by the laser beam. This analysis permits to interpret the optical spectroscopy data in NWs excited by focused laser beams. MicroRaman spectra of Si NWs are obtained and interpreted in terms of the heating induced by the laser beam.

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