Edge-enhanced Raman scattering in Si nanostructures: Single nanowire Raman detection at ~ 1 nW laser excitation power

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Specific optical properties of high-refractive-index nanostructures provide interesting experimental opportunities for studying their Raman spectra and, therefore, stress, phonon confinement and other important characteristics. Here, we report about recently discovered edge-enhanced Raman scattering (EERS) in Si nanostructures [1] and give further details related to this phenomenon. EERS takes place at the 363.8 nm excitation wavelength corresponding to very high Si refractive index N_{Si} ~ 6.5 (at the wavelength 370.8 nm corresponding to the Raman band of Si it is even higher N_{Si} ~ 7.0). In combination with well-known Si resonant-Raman enhancement in near-UV, the enhancement considered in this work makes possible Raman measurements of tiny Si structures at extremely low laser excitation power.

We studied following Si nanostructures: 1) nanostripes (rectangular cross-section with the thickness and width > 100 nm); 2) nanowires (rectangular cross-section with the thickness ~ 10 nm and width 10 - 40 nm); 3) nanopillars (circular cross-section with the diameters in the range 10 – 20 nm). In our experiments, we used 300 nm thick Si nanostripes in the shallow-trench-isolated (STI) device structures. Nanowires oriented along the [110] or [100] axes of Si wafer were fabricated from 10 nm thick semiconductor-on-insulater (SOI) structure by the electron beam lithography. High-aspect-ratio nanopillars oriented along the [001] axis of Si wafer were prepared by the same method from bulk Si. Measurements were done using confocal Raman microscope Nanofinder-30 (Tokyo Instruments Inc.) equipped with the 363.8 nm wavelength Ar laser. 1.4 numerical aperture oil-immersion lens allowed focusing light in ~ 150 nm. Diffraction grating and mirrors were optimized for the near-UV spectral range.

Our finite-difference-time-domain (FDTD) simulation showed that the electric field of the 363.8 nm light is highly concentrated at the edges of Si nanostripe when the light is polarized parallel to it. Light localization area appeared to be less than 20 nm. With decrease in the nanostripe cross-section, in the case of the nanowire, the 364 nm light electric field enhancement is observed in all volume of the nanowire. Change of the nanowire cross-section shape from rectangular to circular did not infuence the effect. For the visible light, the enhancement effect is much weaker because the refractive index of Si in the visible is lower than that in UV.

In agreement with the FDTD simulation, we observed Raman enhancement at the edges of the Si nanostripes in the configuration with both incident and scattered lights polarized parallel to the stripes. EERS allowed us to obtain Raman signal from < 20 nm wide area at the stripe edge and then to determine type and value of the stress in this area with the spatial resolution much better than ~ 150 nm provided by our objective lens.

Raman measurement of nanowires required very low excitation power (~ 100 nW) in order to avoid laser-induced heating. However, even at such low excitation power, Raman signal of single Si nanowires was not weak (Fig. 1) due to EERS (we use this term even for nanowires despite the enhancement takes place in the total volume of the nanowires). Fig. 1 shows that the

nanowire spectra taken at 8 and 85 nW display the same Raman shift 520.15 cm^{-1} and FWHM 3.2 cm^{-1}, while the spectrum taken at 9500 nW excitation display slight downshift 520.05 cm^{-1} and significant band broadening up to 4.9 cm^{-1} caused by the inhomogeneous laser-induced heating [2]. We should note that the illuminated area of a 20 nm wide nanowire is 7-8 times smaller than the area of the laser spot with ~ 150 nm diameter. Therefore, actual excitation power for 20 nm wide nanowire is only ~ 1 nW at the ~ 8 nW excitation. Taking this into account, we have found that the Raman signal of Si nanowire with the 10 nm x 15-20 nm cross section displays ~ 10^2 enhancement compared to Si wafer or SOI the nanowires were made of.

Paradoxical increase in the Raman intensity of Si nanowire with the decrease in its width (in certain range of widths) was experimentally observed (Fig. 2) and confirmed by FDTD simulation. Phonon confinement effect was clearly detected in the Raman spectra of both nanowires and nanopillars. Si nanowire Raman band broadening and downshift (compared to the Si standard FWHM ~ 2.8 cm^{-1} and Raman shift ~ 520.5 cm^{-1}) with decrease in their width was found to be in reasonable agreement with theoretical calculations.

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References:

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[2] S. Piscanec, M. Cantoro, A.C. Ferrari, J.A. Zapien, Y. Lifshitz, S.T. Lee, S. Hofmann and J. Robertson, Phys.Rev.B, **68** (2003) 241312(R)

Figures:

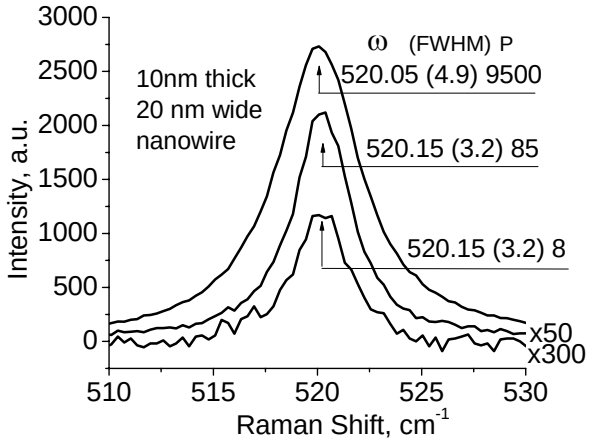

Fig. 1. Dependence of the Si nanowire Raman spectrum on the laser excitation power. ω stands for the Raman shift while P stands for the power.

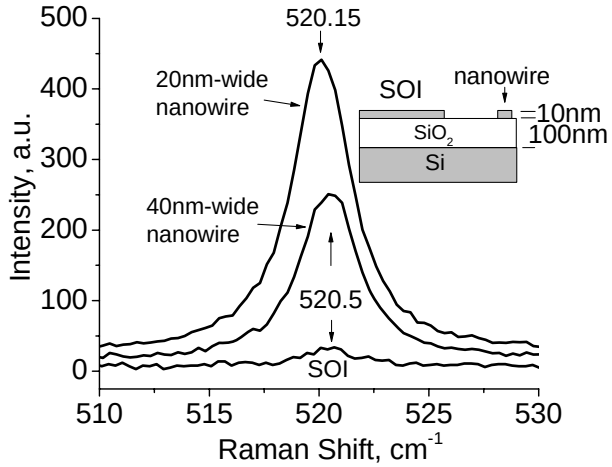

Fig. 2. Single Si nanowire Raman spectra taken in the configuration with both incident and scattered lights polarized parallel to the nanowire at the excitation power of 85 nW; and SOI Raman spectrum taken at the same condition. Cross-section of the sample with SOI and nanowire is schematically shown in the inset. (Raman signal from the Si substrate was just 1.5 times stronger than that from SOI. Therefore, one can neglect its contribution to the nanowire spectra.)