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Thickness dependent phonon renormalization in ultrathin silicon nanomembranes

Two-dimensional (2D) like single crystal silicon nanomembranes (Si NM) has been received huge spotlight in advance of nanoelectronics as well as optoelectronics devices.[1-2] It has been shown that as the thickness of Si NMs is reduced to a few nanometers, the quantum confinement effects (QCEs) would cause the electron mobility reduction owing to the increased phonon scattering.[3] Furthermore, a number of optical analysis on the QCE in Si NM, such as strain/ surface-orientation dependence of phonon confinement effect was reported by studying photoluminescence (PL) and Raman spectroscopy.[4] However, most previous optical investigations of Si NM were made using only the silicon on insulator (SOI) wafer (Si NM / SiO2 / bulk Si), where the signal intereferences of the bulk Si substrate to the observed PL and Raman spectra could not be avoided using any focused laser beam between UV to NIR region. In fact every focused laser beam has large penetration depth (few nanometers to 10 microns) compared to the thickness of the ultrathin Si NM. Since among them UV laser has the smallest penetration depth (~5nm for 325 nm laser), most investigations of the optical properties from the ultrathin Si NM using SOI wafer has been done with a 325 nm laser still creating a limitation in using ultraviolet (UV) laser for few nm (<5 nm) Si NM due to impact of bulk Si substrate. Additionally, to our best knowledge, there is still limited study over the low-frequency Raman scattering from the Si-NMs and subsequent study on thickness dependent electronic band structure and phonon vibrations. Low-frequency Raman scattering is one of the most useful method for the direct observation of acoustic phonon. Mainly the lack of suitable reflective volume holographic filters in the UV region essential for ultralow frequency Raman scattering measurement limits the detailed research of acoustic phonon behavior using SOI wafer. Thus, in some cases, measurements were made using a visible light (Vis) laser having longer penetration depth compared to UV laser but it was not available below 28 nm Si NM because of unwanted effect from bulk Si. [5] In addition, it was also suggested that resonant Raman scattering of optical mode in Si NMs (i.e., 520cm-1 for c–Si) might occur with 3.81 eV excitation (i.e. UV laser).[4] However, detailed studies proving the resonant Raman scattering have not been investigated yet. Thus these limitations such as understanding of the acoustic phonon vibration mode and excitation laser wavelength dependency on Raman spectra for a few nanometer Si NM could not be solved by just using SOI wafer.

In this work, to avoid any further optical impact of bottom Si even in use of visible and near-infrared (Vis-NIR) laser, we have experimentally fabricated thickness-controlled Si NMs on sapphire by transferring a (100) Si NMs from SOI wafer to sapphire substrate. We measured the thickness-dependent Raman spectrums of Si NM using various excitation laser wavelengths. We observed the enhanced Raman intensity at 520 cm-1 from the thinnest Si membrane when excited with 325 nm laser and this result is attributed to the resonant Raman scattering as proven by the shift in transmittance peak of the various thickness films. To further identify resonance Raman scattering, we obtained the thickness dependent band structure modulation utilizing first-principles calculation within generalized gradient approximation (GGA)+U and it was confirmed the transmittance results were in excellent agreement with theoretical results. Our analysis also focused on thickness dependency in ultralow frequency Raman scattering even under a few nanometers. Using photoelastic model, we simulated spectrums and found that they were well matched with measured ones. In addition, we also investigated the uniaxial tensile strain effect on the ultralow frequency Raman modes. This obtained results enabled the effective observation of changes in acoustic mode of Si NM depending on thickness and strain.

References

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Figures

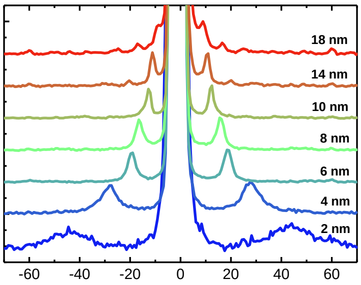

Figure 1: Ultralow frequency Raman spectra of Si NM from 18 nm (top) to 2 nm (bottom) measured with 2.81 eV excitation