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# Manipulation of Optical Phonon Polaritons in Patterned $SiO_2$ Thin-Films

Kartik Venkatraman[1*], Sunny Situ[1], Katia March[2], Peter Rez[3] and Peter A. Crozier[1]

1. SEMTE, Arizona State University, Tempe, Arizona, USA.
2. Eyring Materials Center, Arizona State University, Tempe, Arizona, USA.
3. Department of Physics, Arizona State University, Tempe, Arizona, USA.
* Corresponding author: kvenka16@asu.edu

The detection of vibrational excitations at high spatial resolution with monochromated vibrational electron energy-loss spectroscopy (EELS) can be used to perform local materials characterization on complex geometries. Recent experimental work on a $SiO_2$ thin-film showed that for typical TEM sample geometries, coupling between the entrance and exit surfaces influences the energy-position of the Si-O bond-stretch vibrational mode [1]. Recent theoretical work showed that this coupling between the entrance and exit surfaces manifests itself as an optical surface phonon polariton (SPhP) mode which is supported by $SiO_2$. It was also demonstrated that the probability of excitation of such modes is strongly influenced by the probe position and as the electron beam was positioned closer to the $SiO_2$/vacuum interface, the strength of the SPhP decreases while that of the edge phonon polariton (EPhP) increases [2]. In this paper, we experimentally explore changes in the vibrational energy-loss spectrum when we introduce nanostructural features such as holes in a thin-film of $SiO_2$. We hypothesize that changing the nanostructure of the sample will influence the intensity and energy position of the SPhP and EPhP leading to a change in shape of the vibrational energy-loss spectrum. Simulations were performed based on the classical dielectric theory and finite element method in COMSOL Multiphysics to guide future experiments.

A 3 μm layer of $SiO_2$ on a Si wafer was prepared for STEM EELS analysis by lifting out a focused ion beam (FIB) sample using a Nova 200 NanoLab (FEI) FIB. Holes of different diameters were drilled in $SiO_2$ using the Ga-ion beam in the FIB. A NION UltraSTEM 100 aberration-corrected electron microscope equipped with a monochromator was used to acquire energy-loss spectra. The microscope was operated at 60 kV, with probe convergence and collection semi-angles of 20 and 40 mrad respectively. The experimental EELS energy-resolution was 8 meV. Background subtraction and signal integration were performed using the Gatan Microscopy Suite.

Figure 1a shows the background subtracted vibrational energy-loss spectrum from a pristine $SiO_2$ thin-film. The spectrum consists of a strong asymmetric signal peaking at 142 meV with a strong shoulder that peaks at 154 meV. These are associated with the Si-O SPhP and the bulk asymmetric Si-O vibrational stretch respectively. (The weak peak near 100 meV is associated with the symmetric Si-O vibrational stretch [1, 2]). Fig. 1b shows the background subtracted energy-loss spectra when the electron beam is positioned near the holes at points A and B marked in the annular dark field (ADF) image (insert). It is observed that in comparison with the pristine thin-film spectrum, there is an increase in the intensity and a shift in the energy position of the 142 meV peak to 138 meV at both positions. The shift in the energy-position of the peak is associated with an increase in the excitation of the EPhP and a simultaneous decrease in that of the SPhP.

Simulations were performed using COMSOL Multiphysics to determine the influence of the size of the holes, the separation between two holes and the position of the electron beam relative to the holes on the vibrational energy-loss spectrum. Fig. 2a shows the simulated energy-loss spectrum from a pristine $SiO_2$ thin-film. The lower energy peak at 140 meV is associated with the $SiO_2$ SPhP while the higher energy peak at 155 meV is associated with the bulk Si-O vibrational stretch. Fig. 2b shows the change in the energy-loss spectrum as the radii of the circular holes in a 2x2 array is varied. It is observed that if the position of the electron beam is fixed at the center of the array, an increase in the size of the holes leads to a peak shift in the phonon polariton signal to lower energy-losses. This is associated with an increase in the excitation of the EPhP at the expense of the SPhP. Controlled experiments based on these simulations will be discussed [3].

References:

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[3] The support from National Science Foundation CHE-1508667 and the use of (S)TEM at Eyring Materials Center at Arizona State University is gratefully acknowledged.

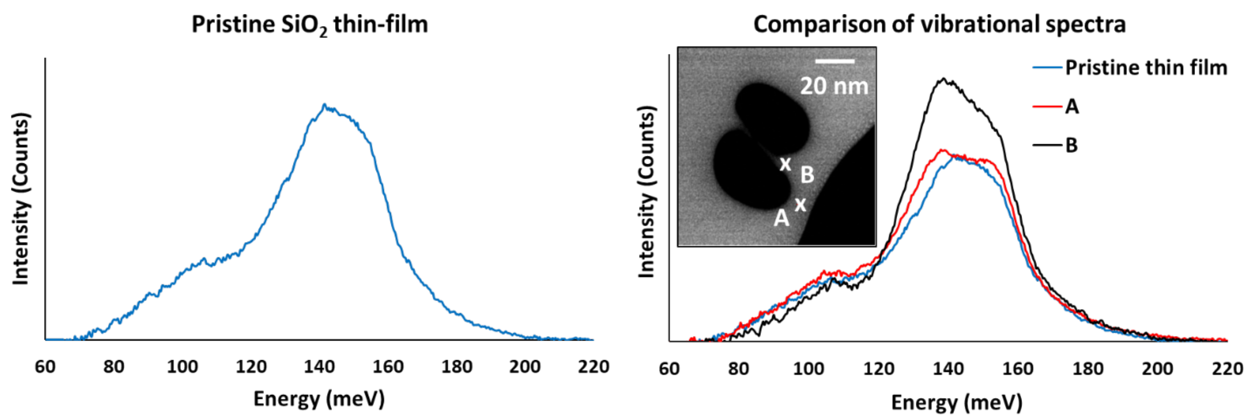


**Figure 1.** a) Background subtracted energy-loss spectrum from a pristine $SiO_2$ thin-film. b) Comparison for two particular positions of the electron beam relative to the holes.

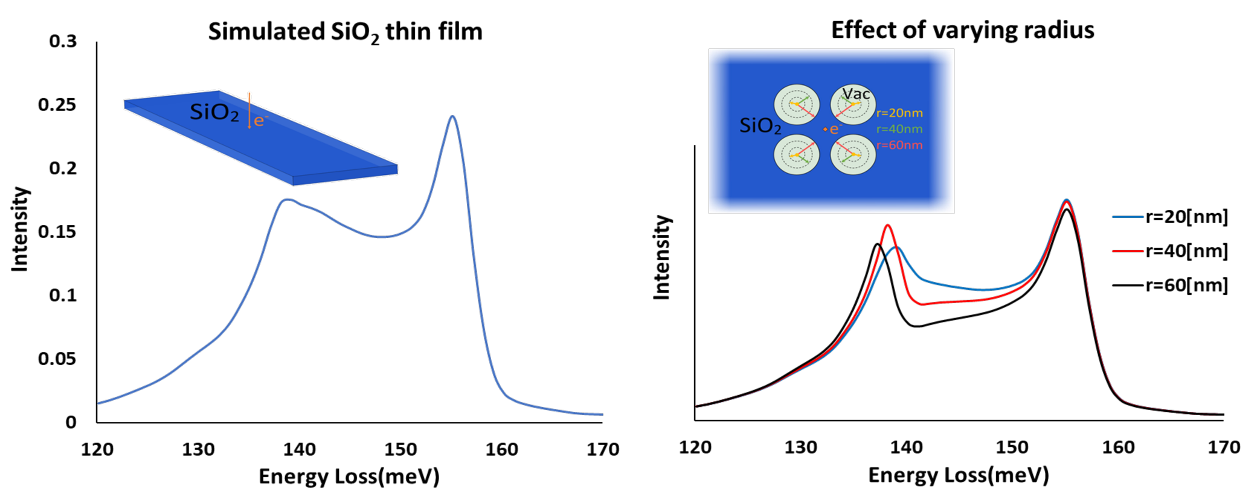


**Figure 2.** a) Simulated energy-loss spectrum from a pristine $SiO_2$ thin-film. b) Simulations showing the influence of probe position relative to the array of holes on the energy-loss spectrum.

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