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Thermal Effects on the Phonon Polariton Response of Nanoscale Cavities

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Infrared phononic cavities are prototype platforms for polaritonic chemistry and nanoscale heat transfer and they have offered unique opportunities to investigate novel chemical and physical processes in extremely confined spaces [1]. Despite recent progress in these research areas, there is still a lack of experimental studies on the surface phonon polariton (SPhP) of a single nanoscale cavity due to limits in spatial resolution of light-base techniques. In this work, we use spatially-resolved vibrational EELS [2] to locally study the polaritonic properties of nanoscale cavities of different sizes, seeking to understand the role of cavity size and temperature in the behavior of coupled SPhP modes.

We fabricated nanoscale cavities (fig. 1a inset) in polycrystalline SiC membranes using a FIB equipped with a Xe^+ plasma source. The fabricated structures mainly consist of a suspended rod-like bridge cut in the middle to form a nanoscale cavity (or gap). The gap distances vary from 5 to 280 nm. Each side of the gap was subjected to uniform temperatures ranging from room temperature to about 1000°C. More complex structures were also fabricated to generate temperature gradients across the gaps. Local temperature was determined by measuring the ratio between energy gain and loss - a method described in reference [3]. The whole imaging and spectroscopy work was performed using a Nion UltraStem equipped with an aberration corrector and monochromator using a ~1.5 - 2 Å probe with an energy resolution of ~10 meV, at 60 kV.

We found that the spectral response within the cavities is dominated by the excitation of several coupled SPhP modes (fig. 1a) appearing between 95 – 120 meV. Scattering modulation in the EELS spectra can be visualized within the cavity due to the changes in the loss probability associated with the excitations of different coupled modes. Temperature leads to inelastic scattering enhancement and apparent shift as the temperature increases (fig. 1b). This effect is primarily driven by the behavior of phonon number population (Bose-Einstein distribution). As the gap distance decreases, we found that for extremely small cavities (gap of 5 nm) the phonon scattering signal increases on the higher energy side of the peak, between 110-115 meV, which suggests the presence of new exotic modes driven by extreme proximity effects. Our simulations of model systems consisting of two infinite planar surfaces and two cylindrical nanorods predict a large variety of coupled SPhP modes and phonon energy splitting, which gets stronger when reducing the gap distance. Coupling between excitations in neighbouring structures is ubiquitous in nanophotonics but studies of coupling between IR polariton excitations using electron probes [4, 5] has just started.

We also explored the polaritonic response as function of cavity temperature. The surface scattering probability from SPhP modes across cavity gaps exhibits a symmetric parabolic distribution for cavities at uniform temperature of the cavity boundaries ($\Delta T = 0$). Figure 2a shows this parabolic profile for cavities with different gap sizes. The parabolic behavior is quite evident for large gap distances and it tends to flatten for small gap distances. For the cavities with temperature gradients across the gaps (ΔT ≠ 0), the parabolic distribution displays a higher increase in scattering on one side of the gap relative to the other side. The intensity increases towards the side with higher temperature (fig. 2b). This surface scattering variation is not associated with only one side of the cavity because the coupled SPhP modes result from the interaction between two modes at different thermal states. Theoretical developments for inelastic electron scattering from phonons sustained in objects at different thermal states are not yet developed. Progress along this direction will permit to provide an accurate interpretation of scattering EELS data and determine the thermal properties of coupled nanosystems.

Finally, we measured locally the temperature of the suspended structures using the bulk acoustic phonon scattering signal collected from regions in the suspended rods, quite near to the cavity (fig. 2c). The measured temperature values agree with values provided by the heating system. Interestingly, the temperature determined using the polaritonic signal acquired with the probe parked within the cavity exceeds the measured bulk temperature by ~ 200 ºC. This temperature variation is evident in the slope change of the linear curves in Figure 2c. This finding seems to suggest that localized SPhP on the cavity surfaces might be subjected to a higher temperature condition resulting in local heating within the cavity. An explanation of the physical mechanisms driving this anomalous effect remains to be presented.

In summary, we studied the coupling of SPhP modes within nanocavities of varying sizes and at different temperatures. Our spatially-resolved EELS measurements demonstrated the existence of a large variety of active channels (or SPhPs) for radiative heat transfer and polaritonic chemistry processes. The scattering variation across the cavity is symmetric for systems at uniform temperature across the cavity, and asymmetric for cavities subjected across temperature gradients. Our work brings fundamental insights into the polaritonic and thermal response of cavities, representing progress on light matter interaction processes at different temperatures [6].

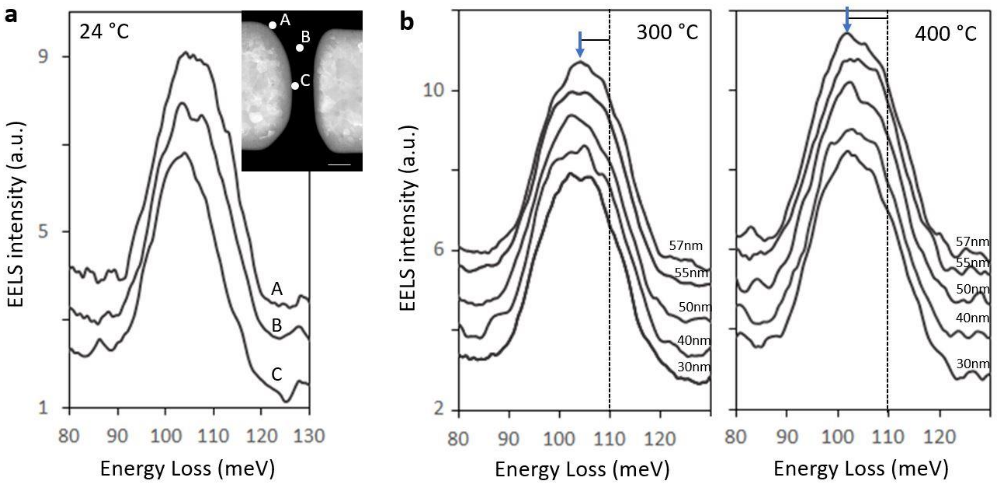

Figure 1. Spectral behavior of phononic cavities as a function of temperature. (a) EEL spectra acquired at different locations (A, B and C) within a cavity at room temperature. Inset shows an ADF-STEM image of the cavity. Scale bar is 100nm (c) EEL spectra acquired in a cavity across the shortest gap distance at 300 and 400 °C. Notice the variations in the EELS intensity and apparent shift imposed by temperature changes.

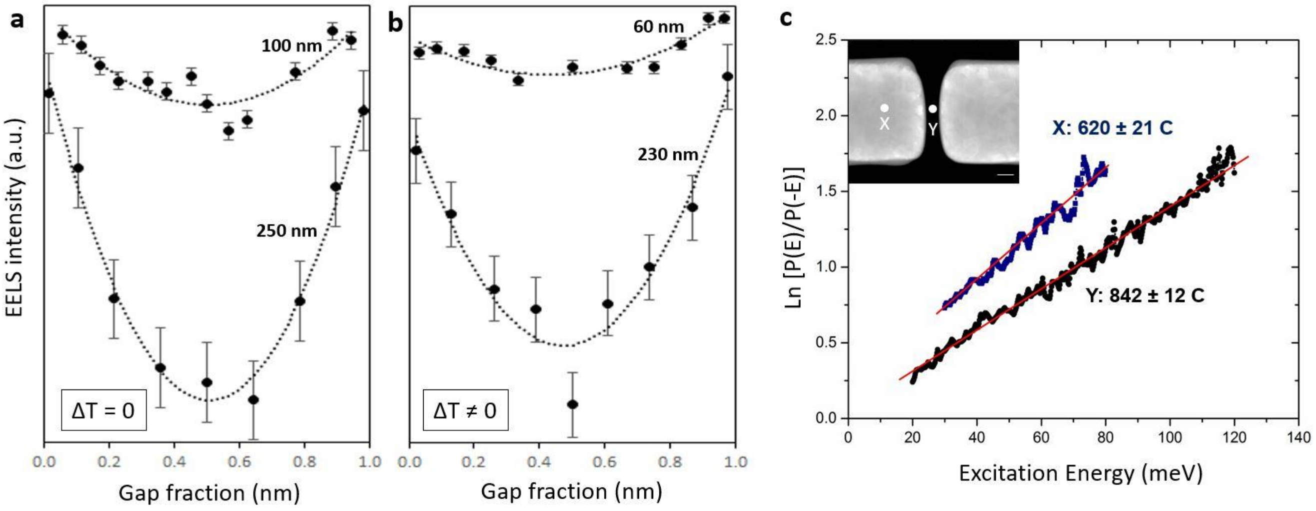

Figure 2. EEL scattering distribution across the cavity gaps at different thermal conditions. (a) For cavities at uniform temperature (ΔT = 0) the distributions exhibit a symmetric parabolic behavior. Gap distances are indicated next to each curve (b) For cavities with temperature gradients across the gaps (ΔT ≠ 0), there is an asymmetry in the parabolic profile. (c) Temperature measurements at different locations (X and Y) within/near the cavity. Temperature values are indicated. ADF image of the cavity is shown in the inset. Scale bar is 65 nm.

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