Measurement of phonon damping by nanostructures

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Abstract. The understanding of phonon lifetime and scattering rates is attracting an increasing interest due to the major role of phonon in thermal and electrical conductivity which are key properties for technological applications. The infrared complex dielectric function of a crystal is determined by the harmonic characteristics of the phonon together with the intrinsic and extrinsic phonon scattering rates. In order to investigate the interplay between the phonon intrinsic scattering and the scattering of the phonon by a nanostructured surface, infrared reflectivity measurements from SiC nano-pyramids on SiC substrate have been analysed using a Kramers-Kronig conversion technique to deduce the infrared complex dielectric function. Then, the real and imaginary parts of the dielectric function were fitted simultaneously by using a theoretical model for the dielectric constant that considers frequency-dependent phonon damping at the center of the Brillouin zone. It has been found that surface nanostructuring strongly enhances the overall scattering rate of the phonon at the Brillouin zone center.

1. Introduction
Nanostructured SiC has been recently receiving a lot of attention from the materials and device community. Due to its wide potential applications ranging from microelectronics, optoelectronics, high temperature semiconductor nanocomposites, photocatalysts, electron field emitting devices to biomedical engineering and functional ceramics. All said applications rely on the unique attractive properties of SiC, including high temperature material stability, high thermal conductivity, low thermal expansion coefficient and thermal shock endurance [1–4]. As well as, the high mechanical strength of SiC enables its use in harsh environment. In order to better use and control the properties of the nanostructured SiC, a quantitative understanding of phonon lifetime and scattering rates is mandatory [5] since the phonon response controls the thermal and electrical conductivity in the infrared spectral range. For this purpose SiC nanostructures of diameter ranging between 70 nm and 100 nm were fabricated on SiC. In this contribution the complex infrared dielectric function of nanostructured SiC was calculated and fitted to experimental data. The permittivity was calculated by a model that considers the material as coupled damped oscillators. Perturbation theories were applied to determine the anharmonic components of interatomic forces associated with interchanges of energies between phonon modes as function of frequency and temperature [6]. Kramers-Kronig conversion technique was employed to deduce the infrared complex dielectric function [7].
2. Experimental Details
We have purchased good-crystalline quality SiC wafers from Cree, Inc. The sample was cut into two pieces, where the nanostructures were fabricated on one of them. The pattern of the structures was drawn by electron-beam lithography, preceded by the deposition of positive tone PMMA resist, followed by lift-off procedure. An Al layer was then deposited as a mask for Reactive Ion Etching (RIE) of the patterned SiC substrate. RIE was operated with the use of inductively coupled plasma in a mixture of 60% SF6 and 40% O2 at 3 mTorr pressure. The residual mask was removed in aqueous solution of KOH.

The reflectivity from each sample is normalized to that of a gold coated mirror. The measurements were conducted at room temperature in the 400-1500 cm\(^{-1}\) spectral range. Near normal incidence angle was fixed with uncertainty of 15°. The FTIR setup is Nicolet 4700 spectrometer from the Thermo Electron Corporation with a DTGS detector and a KBr beamsplitter.

3. Results and Discussion
Figure (1) shows the fabricated nanostructured SiC with diameter ranging from 70 nm to 100 nm. We investigated the surface phonons in SiC and the effect of the structuring on their lifetime. We measured the intact surface SiC reflectivity and that of the surface with nanostructures. Figure (2) is the unpolarized infrared reflectivity measurements made on SiC unmodified wafer and on the fabricated nanostructured SiC. A clear difference is noticed between the two reflectance spectra. This difference can not be due to light diffusion because the size of the nanostructures is about two orders of magnitude smaller than the light wavelength.

The reflectivity measurements have been converted into real and imaginary parts of the dielectric functions. These functions are fitted simultaneously with a Lorentz model that considers the material as coupled oscillators with frequency-dependent damping constants [6]. Figure (3) and Figure (4) show the obtained results. The data from the nanostructured SiC is plotted along with that of the intact SiC. We see in symbols the experimental results and in solid lines the simulated ones. The best fit is mostly affected by considering the 3-phonon decays, which demonstrates that at room temperature the 3-phonon decay processes are the dominant mechanism.

A remarkable difference in the permittivity was observed due to the introduction of the nanostructures. From the fit we deduce that the presence of nanostructure has increased significantly the phonon damping. Figure (5) compares the damping constants of the two
investigated samples. We found that the damping constants related to the intact sample is ranging from 6 times to an order of magnitude smaller than that of the surface with nanostructures depending on the frequency range. Since the two measured samples come from the same epitaxial layer, we attribute the enhancement of the scattering rate to collisions between the surface phonons and the structures. Furthermore, these results add support to the hypothesis that in nanostructures the phonon are confined and have no interaction with the external environment. Unfortunately, the reflectivity of a single nanostructure could not be measured because of the difficulty in focusing the infrared light on such a small region; however the measurements of the confined phonon on a single nanostructure is being pursued in our laboratory using near-field optical techniques.

4. Conclusion
Phonon scattering by surface structuring is illustrated in the case of SiC. Infrared reflectivity measurements have been converted into real and imaginary parts of the material dielectric function. The results have been fitted to Lorentz model that considers coupled oscillators and frequency-dependent damping coefficient. We have found that the surface structuring enhances the phonon scattering mechanisms up to an order of magnitude depending on the frequency
range. We hope that these results stimulate further experiments on tailoring the harmonicity and anharmonicity of the phonons for controlling the SiC infrared properties for the use in SiC-based devices. More analysis taking into account the localized phonon modes and the effect of surface structuring on the decay channels for optical phonons will be the subject of another paper.

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