

Plasmon Excitation of Coherent Interface Phonons in Si-SiO₂ Systems

M.S. Choi, N. Zhang, M. Dutta, and M.A. Stroschio¹

Dept. of Electrical and Computer Engineering, University of Illinois at Chicago, Chicago, IL 60607, USA

¹Email: stroschio@uic.edu

C.O. Aspetti and R. Agarwal

Dept. of Materials Science and Engineering, University of Pennsylvania, Philadelphia, PA 19104, USA

INTRODUCTION

Herein, the creation of interface phonons in silicon is modeled in metal-SiO₂-Si heterostructures based on plasmon-excited generation of longitudinal optical phonons in polar SiO₂ which in turn evanesces into the silicon via a remote polar phonon mechanism [1]–[3]. Recent study also suggests that large-amplitude acoustic pulses can be generated by ultrashort surface plasmons [4] These studies presented herein are stimulated, in part by the hot luminescence with relatively high efficiency in bulk silicon with a silver nanocavity and 5 nm thick silicon dioxide (SiO₂) interlayer [5], of which the internal quantum efficiency is very low by silicon itself. In this abstract, we will demonstrate coherent interface phonon production with high phonon occupation numbers. The generation of such nonequilibrium phonons with high occupation number is of interest in many areas including: carrier transport, studies of phonon decay rates, studies of phonon-assisted processes [6]–[8], and second order processes requiring both phonons and photons.

THEORY

When the electromagnetic field pulse is sufficiently short comparing to the phonon mode of a material, impulsive stimulated scattering (ISS) can occur [9]. Unlike phonons generated from ordinary scattering processes, the phonons generated as a result of ISS are in the same mode. These "coherent" phonons, which can be "selected" [10], would have high phonon occupation number. On the other hand, laser incident on metal creates plasmons at the metal-SiO₂ interface. Since surface plasmon polaritons can be defined as a specific set of surface electromagnetic modes [11], we assume the plasmons as an amplified electric field in this abstract; the magnitude of triggering pulse will be treated as simply the Purcell enhancement factor times the magnitude of the electric pulse. The polar phonons generated by laser-induced plasmons evanesce into the silicon. The coherent optical phonon displacement can be calculated from the equation of motion as following [10]:

$$Q(z > 0, t > 0) = Q_0 e^{-\gamma(t-zn/c)} \sin[\omega_0(t - zn/c)]$$

where

$$Q_0 = 2\pi I N \alpha' e^{-\omega_0^2 \tau_L^2 / 4} / \omega_0 n c$$

where I is the magnitude of the triggering pulse, α' is the differential polarizability, τ_L is the pulse duration. The optical phonon displacement is plotted in Figure 1 for silicon inside the nanocavity in 140 fs laser pulse at 4.11×10^{15} Hz. For the given phonon displacement, the energy loss due to the emission of coherent optical phonons is given by [10]:

$$\Delta E = \frac{\Delta I}{I} = -\frac{2\pi N \omega_0 l \left(\frac{\partial \alpha}{\partial R}\right) R'_0}{n c}$$

$$\text{where } R'_0 = \frac{4\pi I \left(\frac{\partial \alpha}{\partial R}\right)_0 e^{-\frac{\omega_0^2 \tau_L^2}{4}}}{\omega_0 n c}$$

PHONON POTENTIAL

The screening length of the metal in the structure under study is δ and the thickness of oxide is d (from 0 to d). Let the phonon potentials (Φ) for the given structure be defined as follow:

$$\begin{cases} \Phi = A e^{-q(z-d)} & \text{when } z > d \\ \Phi = B e^{qz} + C e^{-qz} & \text{when } 0 \leq z < d \\ \Phi = D e^{z/\delta} & \text{when } z < 0 \end{cases}$$

By using appropriate boundary conditions and the normalization condition, we can then get the secular equation for this structure whose solution will be the interface phonon potentials of this system [12]:

$$\frac{\partial \epsilon_{metal}(\omega)}{\partial \omega} D^2 \frac{\delta}{2} \left(1 + \frac{1}{\delta^2}\right) + \frac{\partial \epsilon_{ox}(\omega)}{\partial \omega} q (B^2 (e^{2qd} - 1) + C^2 (1 - e^{-2qd})) + \frac{\partial \epsilon(\omega)}{\partial \omega} q A^2 = \frac{4\pi \hbar}{L^2}$$

DISCUSSION

We found that the energy loss due to the emission of coherent optical phonon in SiO₂ to be about 5 % of the plasmons, which is about 135 meV with 2.7 eV. Parameters used in the calculation are summarized in Table I. Simultaneously, the phonon potential calculation shows in Figure 2 that the 143 meV Si-SiO₂ interface phonon is produced. This indicates that almost every plasmon produces a coherent optical phonon at Si-SiO₂ interface: this mechanism could be a possible source of massive nonequilibrium phonon production which would lead to many areas.

ACKNOWLEDGEMENT

This material is based upon work supported by, or in part by, the U. S. Army Research Laboratory and the U. S. Army Research Office under contract/grant number W911NF0810114.

REFERENCES

- [1] K. Hess and P. Vogl, *Solid State Comm.*, Vol. 30, pp. 807-809, 1979.
- [2] B.T. Moore and D.K. Ferry, *J. Appl. Phys.*, 51(5), 2603, 1980.
- [3] J.P. Leburton and G. Dorda, *Solid State Comm.*, Vol. 40, pp. 1025-1026, 1981.
- [4] V. V. Temnov, C. Klieber, K. A. Nelson, T. Thomay, V. Knittel, A. Leitenstorfer, D. Makarov, M. Albrecht, and R. Bratschitsch, *Nature Communications*, 2013.
- [5] C.-H. Cho, C. O. Aspetti, J. Park and R. Agarwal, *Nature Photonics*, 2013.
- [6] M. A. Stroschio, "Interface-Phonon-Assisted Transitions in Quantum Well Lasers", *J. Appl. Phys.*, 80, 6864, 1996.; see also M. Stroschio and M. Dutta, *Phonons in Nanostructures*, Cambridge University Press, 2001.
- [7] M. V. Kisin, V. B. Gorfinkel, M. A. Stroschio, G. Belenky, and S. Luryi, "Influence of Complex Phonon Spectra on Intersubband Optical Gain", *J. Appl. Phys.*, 82, 2031, 1997
- [8] M. A. Stroschio, M. Kisin, G. Belenke, and S. Luryi, "Phonon enhanced inverse population in asymmetric double quantum wells", *Appl. Phys. Letts.*, 75, 3258-3260, 1999.
- [9] Y.-X. Yan, and K. A. Nelson. "Impulsive stimulated scattering I. General theory." *J. Chem. Phys.*, 87:6240-56, 1987.
- [10] Y.-X. Yan, E. B. Gamble, Jr., and K. A. Nelson. "Impulsive stimulated scattering: General importance in femtosecond laser pulse interactions with matter, and spectroscopic applications." *J. Chem. Phys.*, 83:5391-9, 1985.
- [11] E. Le Ru, and P. G. Etchegoin. *Principles of Surface-enhanced Raman Spectroscopy and Related Plasmonic Effects*. Amsterdam: Elsevier, 2009.
- [12] A. R. Bhatt, K. W. Kim, M. A. Stroschio, G. J. Iafrate, Mitra Dutta et al, "Reduction of interface phonon modes using metalsemiconductor heterostructures", *Journal of Applied Physics* 73, 2338, 1993.

Parameter	SiO ₂
Density of Oscillator	$2.3 * 10^{22}$ atoms/cm ³
Amplitude of the field	7420 V/cm ²
Frequency of the field (ω)	$6.52 * 10^2$ THz
Refractive index	1.55181 at $4.11 * 10^2$ THz
Pulse duration	50 fs
Damping constant	$0.37 * \omega$
Scattering cross section	$2.8 * 10^{-14}$ cm ²

TABLE I: Parameters used to calculate the energy loss due to the emission of coherent optical phonon in SiO₂

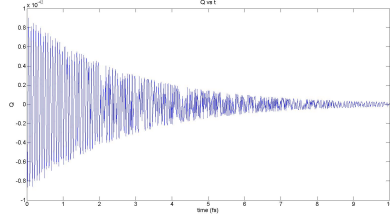


Fig. 1: Coherent optical phonon displacement in silicon

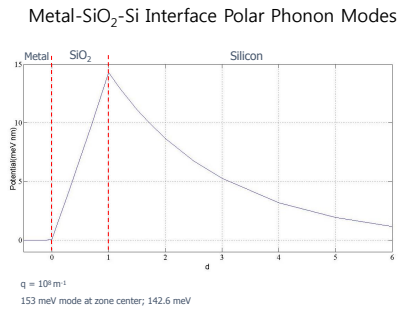


Fig. 2: Interface phonon potential of Metal-SiO₂-Si system