1-5-4

Application of Femtosecond Shaped Optical Pulses for Manipulation of Surface Phenomena with Atomic Scale Resolution

Don Futaba, Naoshi Morita, Ryuji Morita, Akira Suguro* and Mikio Yamashita
Department of Applied Physics, Hokkaido University & CREST・ JST, *Citizen Watch Co.

For the purpose of mediating surface processes through the generation of selective phonons and locally probe the electronic structure, we are constructing a femtosecond (fs) laser-scanning tunneling microscope (STM). Through the use of femtosecond (fs) shaped optical pulses using a spatial light modulation of fs-laser pulses, we will generate coherent lattice vibration, which will then be used to mediate surface processes. The motivation to mediate reactions or structural changes stems from the desire to explore hidden reaction pathways. In addition, with the ability of the STM, local probing of the surface electronic structure at the atomic level is possible.

Pulse trains or timed pulses have previously been used in exciting resonances in such materials as molecules, molecular crystals[1], BiSb mixed crystals,[2] and GaAs[3] by adjusting the repetition rates to match the vibrational mode of interest.

In our first experiment, we are pursuing the selective desorption of silicon adatoms from the Si(111)-7x7 surface. The less strongly bonded adatom sites serve as localization sites of electronic excitation energy that makes them excellent sites to attempt to excite localized vibrations. Electron energy loss spectroscopy experiments on the (7x7) surface[4] found distinct losses at 570 cm^{-1} and a broader structure at 200-270 cm^{-1} which agreed well with molecular dynamics simulations[5]. Furthermore, these calculations have found that both the center and corner adatoms have distinct out-of-plane vibrational modes. The energies of these modes have been calculated to be 6, 10, and 15 THz for the faulted half (FH) corner sites and 8, 10, and 16.3 THz for the corresponding modes of the FH center sites. The vibrational frequencies for unfaulted half (UH) adatoms were found to be similar to those of FH center adatoms. Using this as a guideline, out intent is to selectively excite adatom vibrations and thus induce desorption.

The experimental arrangement (illustrated in the figure below) consists of a mode-locked Ti:Sapphire laser (repetition rate: 100 MHz, pulse width: 100-fs, pulse energy: 1 nJ, center wavelength: 800 nm) a 4-f pulse-shaping apparatus with two concave mirrors (focal length: 30 cm), two gratings (grating constant 1/1200 mm), and a liquid crystal spatial light modulator (648 pixels).

Currently we are working to utilize a 2-D pulse-shaper to enable control over the pulse shape as well as relative phase control between successive pulse-trains.

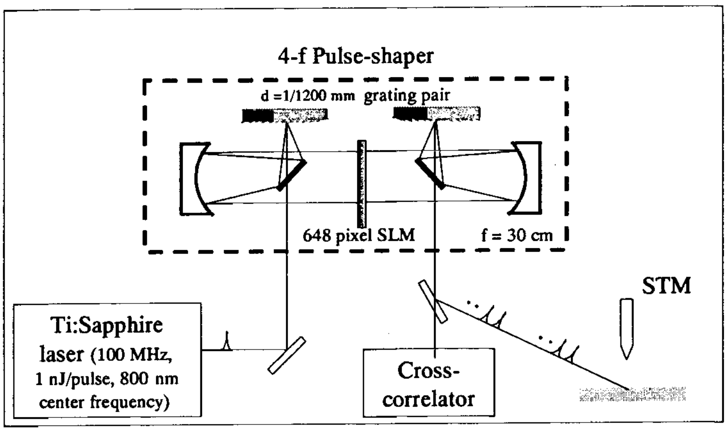

Figure of the experimental arrangement. Pulses from a mode-locked Ti:Sapphire laser are input into a 4-f pulse-shaping apparatus with two concave mirrors, two gratings, and a spatial light modulator at the Fourier plane. Shaped pulses are directed onto the sample surface.

---

[1] A.M. Weiner, D.E. Leaird, G.P. Weiderrecht, and K.A. Nelson, Science **247** (1990) 1317.

[2] M. Hase, T. Itano, K. Mizoguchi, and S. Nakashima, Japn. J. Appl. Sci. **37** (1998) L281.

[3] T. Dekorsy, W.A. Kutt, T. Pfeifer, and H. Kurz: Europhys. Lett. **23** (1993) 223.

[4] W. Daum, H. Ibach, and J.E. Müller, Phys. Rev. Lett. **59** (1987) 1593.

[5] J. Kim, M-L. Yeh, F. S. Khan, and J.W. Williams, Phys. Rev. B **52** (1995) 14709.