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# Time resolved hyper-Raman surface spectroscopy of (111) silicon

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We performed a time-resolved Hyper-Raman spectroscopy of a (111) silicon wafer upon its excitation by a near IR optical pulse. Upon excitation of the silicon wafer by the optical pulse, we record the broadening and increase of the Stokes and anti-Stokes bands centered around the Si lattice phonon and SiO<sub>2</sub> mode centered at ~610 cm<sup>-1</sup> and ~1100 cm<sup>-1</sup>, respectively. This unique technic makes it possible to reveal the electron-phonon scattering which thermalize the hot carriers with the lattice on time scale of 100 to 300 fs.

## I. INTRODUCTION

The dynamics of photoexcited electrons in semiconductors and metals have been extensively studied [1]. On ultrashort time, they are driven by two dominant processes: electron-electron and electron-phonon scattering. While different technics have been used to record these dynamics, time resolved X-ray diffraction [2], [3] and electron diffraction [4], [5] experiment are nearly ideal tools to study lattice deformation induced by carrier relaxation. Hereafter, we demonstrate that recording the evolution of the spectrum of THz-induced SHG versus the delay between a sub-picosecond near infrared (IR) optical pulse and an ultra-short THz pulse makes it possible to record the impact of photo-carrier generation on the evolution of the IR active phonons in a (111) Si wafer with a sub-100 fs time resolution.

## II. EXPERIMENTAL SET-UP

The experiment is performed using a 3.5 mJ Ti:Sapphire regenerative amplifier that yields at 1 kHz repetition rate, 50 fs pulses centered around 800 nm. A 1 mJ pulse is used to generate ultrashort THz pulse through a two-color plasma generation. The generated THz pulse is transmitted by a 1 mm thick silicon wafer, collimated and focused on the (111) silicon wafer by two off-axis parabolic mirrors. On the silicon wafer, set at an angle of incidence of 45°, the p-polarized THz pulse is mixed colinearly with a 2 μJ s-polarized near IR optical pulse spectrally narrow ( $\Delta\lambda \sim 40$  cm<sup>-1</sup>) centered at ~800 nm which pulse duration is ~500 fs at full-width at half maximum (FWHM). The near IR optical pulse, passing through the focusing off-axis parabolic mirror, is focused on silicon wafer by a lens which focal length  $f=125$  cm. A delay line makes it possible to delay the near IR optical and THz pulses. The reflected SHG signal generated on the silicon wafer is detected by a photomultiplier set at the exit of a monochromator. The THz pulse is chopped at 500 Hz and the SHG signal is recorded by a lock-in amplifier on which the photomultiplier is connected. The THz induced SHG spectrum is recorded by a thermally cooled CCD camera at the exit of a spectrometer. The spectral resolution for this experiment is determined by the spectrograph and the bandwidth of the near IR optical pulse.

## III. RESULTS

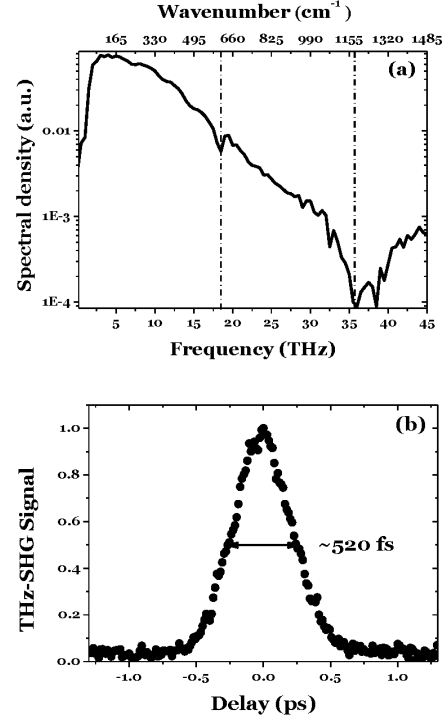


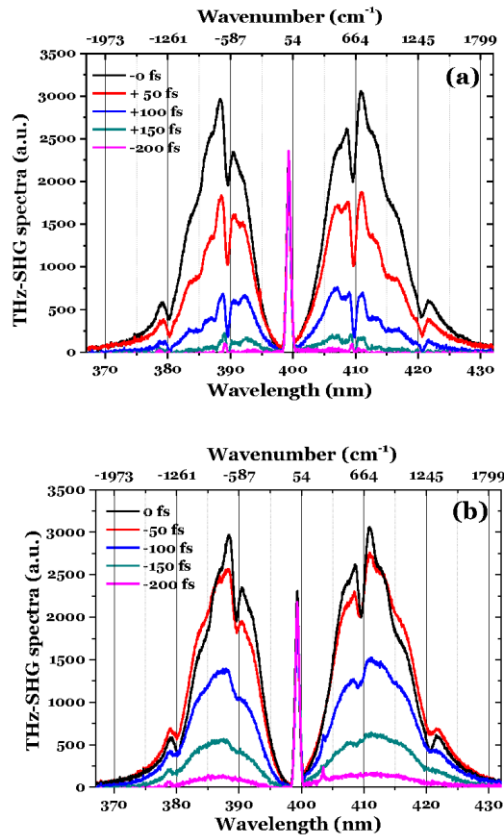
Fig. 1. a) Temporal evolution of the THz-SHG signal. b) Spectra of the THz pulse used to perform the THz-SHG experiment.

Our THz pulse is short (~60 fs) and its spectrum, centered about 5 THz, spans up to 45 THz (Fig. 1a). The latter reveals two IR absorption bands which are induced by silicon wafer used to filter the THz pulse. These bands are centered about 18.5 THz (~610 cm<sup>-1</sup>) and 35.4 THz (~1100 cm<sup>-1</sup>). They are related to Si lattice phonon and the absorption by interstitial oxygen in crystalline silicon[6].

The evolution of the THz induced SHG signal recorded when delaying the THz pulse is displayed in Fig. 1b. Its FWHM is ~520 fs, close to the FWHM of the near IR optical pulse. Silicon being centrosymmetric, it writes  $E_{SHG}(2\omega \pm \Omega) \sim \chi^{(3)}(\omega, \omega, \Omega, 2\omega \pm \Omega) E(\Omega) E^2(\omega)$  where  $E(\Omega)$ ,  $E(\omega)$  and  $\chi^{(3)}(\omega, \omega, \Omega, 2\omega \pm \Omega)$  are the electric field of the THz and optical pulses and the third-order susceptibility of the studied material, respectively. As written, this indicates  $E_{SHG}(2\omega \pm \Omega)$  results from a Hyper-Raman scattering process. The enhancement of the SHG spectrum at a frequency at  $2\omega \pm \Omega_1$  is the signature of resonant mode of the material at  $\Omega_1$ .

The evolution Hyper-Raman (i.e. THz-induced SHG) spectrum reflected by the silicon wafer as we delay the optical pulse with respect to the THz pulse is displayed in Fig. 2. At its maximum, the latter exhibits a sharp and narrow peak centered at ~399 nm and a Stokes and anti-Stokes band shifted

by  $\sim 11$  nm. When the THz pulse is blocked the latter disappears while the peak centered at 399 nm remains. This indicates the central peak results from SHG at the air-silicon wafer interface, while the Stokes and anti-Stokes bands results from difference- and sum-frequency between the THz and second harmonic spectral components. The Stokes and anti-Stokes bands are shifted by  $\sim 610$   $\text{cm}^{-1}$ , indicating they are associated to the resonance of the third order susceptibility for THz spectral component centered around the Si lattice photon frequency.



**Fig. 2.** Evolution of the THz-SHG spectra versus the delay between the near IR optical and THz pulse. a) The THz pulse is ahead of the optical pulse. b) The THz pulse is delayed with respect to the near IR optical pulse.

When the THz pulse is ahead of the optical pulse by  $\sim 150$  fs (Fig. 2a), we will show that we can well reproduce the spectral feature of the Stokes and anti-Stokes bands considering the far-IR spectra of silicon. This indicates that, as we previously mention, the recorded THz-SHG is indeed related to Hyper-Raman scattering and that the number of carriers generated by IR optical pulse do not affect too much the equilibrium lattice phonon. However, when the THz pulse is further delayed with respect to the optical pulse the amplitude of the Stokes and anti-Stokes bands is enhanced and their spectral width is larger (Fig. 2b). This reveals the expected broadening of the phonons and excitation of higher IR active phonon modes induced by electron-phonon scattering. It is worth mentioning that in silicon, the IR absorption are induced by a two-phonons process which is very sensitive to the temperature of the lattice [7], [8]. More details about this ensemble of results will be given during the presentation.

## IV. SUMMARY

In summary, we performed a time-resolved THz induced SHG spectroscopy of a (111) silicon wafer a technic which clearly reveals electro-phonon interaction during generation of photo-carriers in a semiconductor.

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