Relativistic Doppler reflection of terahertz light from a moving plasma front in an optically pumped Si wafer

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Relativistic Doppler reflection is an interesting phenomena introduced by special relativity. When light is reflected by a counter-propagating mirror with a velocity close to the speed of light, a frequency up-shift of the light due to the relativistic Doppler reflection is induced as

$$\nu_r = \nu_i \Gamma = \nu_i\,(1+\beta)/(1-\beta)\,, \tag{1}$$

where ν_r and ν_i are the frequencies of the reflected and incident light, respectively, Γ is the frequency up-shift factor, and $\beta = v_P/v_L$ is the ratio of the velocities of the moving mirror (v_P) and the incident light (v_L) in the medium. A moving plasma mirror has been previously proposed and used to demonstrate relativistic Doppler reflection at the Kansai Photon Science Institute (KPSI).[1] Overdense plasma with a high carrier density can act as a mirror for the light. When the near-infrared (NIR) ultra-intense laser is focused on the plasma, the free electrons in the plasma are accelerated to a velocity close to the speed of light by laser-plasma interaction. As a result, a relativistic moving plasma mirror is generated. Alternatively, Roskos and their colleagues have suggested the occurrence of Doppler reflection with a moving plasma front in a semiconductor excited by a femtosecond laser pulse.[2] A dense plasma layer consisting of free electrons in a conduction band is easily generated by optical pumping with NIR to visible light. In this case, the photo-excited electrons in the semiconductor do not receive a large momentum from the laser field. However, free electrons are successively generated while the optical pump pulse propagates into the semiconductor; subsequently, the ionization front copropagates with the optical pump pulse. Therefore, the moving ionization front can act as a relativistic moving plasma mirror.

The relativistic Doppler reflection of terahertz (THz) light in photo-excited silicon (Si) wafers has been investigated in our previous studies.[3] The THz light oscillates at a low frequency (approximately 10^{12} Hz) as compared to an optical light and therefore, it can be reflected by a plasma with a relatively low carrier density (approximately 10^{17} cm^{-3}). Furthermore, the THz light penetrates the Si wafer without significant absorption and dispersion. Thus, THz light is a suitable probe for relativistic Doppler reflection in Si. Time-resolved THz time-domain spectroscopy (THz-TDS) has been employed to reveal the Doppler reflection mechanism in Si. In THz-TDS, a waveform of the THz light pulse is measured in the time-domain and a complex THz spectrum is obtained by a Fourier transform of the waveform. This provides a significant advantage for understanding the interaction between a moving plasma front and THz light. The THz waveforms and spectra measured as a function of the delay time between the optical pump and THz probe light clearly demonstrated that the evolution of the moving plasma front with time induced the frequency up-shift of the reflected THz light. However, in our previous study, the measured frequency up-shift was considerably smaller than that predicted by Eq. (1). One possible reason for this is an insufficient bandwidth in the THz-TDS to measure a large frequency up-shift.

In the present study, the frequency up-shift in the relativistic Doppler reflection is re-examined by extending the spectral bandwidth in the THz-TDS measurement. The broad bandwidth allows for accurate measurements of the large frequency up-shift to evaluate quantitatively the mechanism for the relativistic Doppler reflection. The high temporal resolution of the THz waveform is also provided by the broadband measurement. The improvement in the measurement allows a highly accurate analysis of the phase and the intensity spectra. The phase shift induced by the Doppler reflection includes detailed information on the moving plasma front. It is demonstrated that the Doppler reflection in the photo-excited Si realizes the spectral broadening and the pulse compression of the THz light, which provides a technique for the manipulation of the THz light.

Figure 1 shows a schematic diagram of the counter-propagation geometry of the moving plasma front and the THz pulse in Si.[3, 4] (1) First, the surface of a Si wafer is irradiated with a THz pulse that propagates through the Si. (2) When the THz pulse arrives at the rear surface, the pulse splits into two parts: the transmission to the air (Pulse *A*) and the internal reflection (Pulse *B*). (3) Prior to the arrival of the reflected THz Pulse *B* at the input surface, the optical pump light is shone on the Si wafer to generate a plasma layer. (4) The THz Pulse *B* is affected by the plasma near the input surface and is then subsequently reflected. The THz Pulse *B* then counter-propagates with respect to the pump light in the Si; subsequently, the plasma front approaches the THz light with a velocity close to that of the pump light. This induces relativistic Doppler reflection and causes a frequency up-shift for the THz light. In this scheme, the THz and optical light are input collinearly to the front surface of the Si, and the reflected THz pulse is taken from the rear surface. The two pulses (Pulse *A* and *B*) from the rear surface are separable by their arrival times at the detector. Thus, the THz pulse reflected by the moving plasma front is easily measured using this scheme.

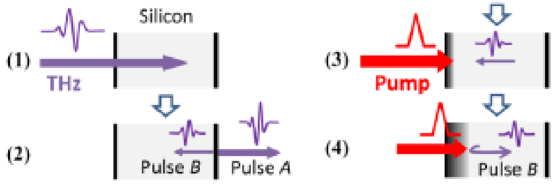

Fig. 1. (a) Diagram of the relativistic Doppler reflection. The time sequence of the THz probe and optical pump pulses and their propagation in the Si wafer are shown from (1) to (4).

The experimental apparatus is based on the optical pump-THz probe time-resolved measurement with THz-TDS as shown in [5]. Ti:Sapphire regenerative and multi-pass amplifiers were employed to generate pulses for THz light generation, THz pulse detection, and optical pumping of the Si wafer to generate the plasma layer. The THz pulse was generated by optical rectification in a $LiNbO_3$ prism with pulse front control. The optical pump and THz probe light collinearly propagated into a 1-mm thick high-resistivity (2 kΩcm) Si wafer. The waveform of the transmitted THz pulse from the Si was measured by electro-optic (EO) sampling. The bandwidth of the THz-TDS measurement was limited by the pulse width of the EO sampling light and the absorption of the THz light in the birefringent EO crystal. The pulse width of the EO sampling light (50 fs) was sufficiently short to measure the frequency up-shift to 10 THz. In the previous study, a 1-mm thick ZnTe crystal was used for the EO sampling.[3] However, due to the strong phonon absorption, the bandwidth was limited to under 2.5 THz. In the present study, to obtain the broadband measurements, including those in the higher frequency range, a 0.4-mm thick GaP crystal was employed whose edge of the phonon absorption band was approximately 5 THz.

Figure 2(a) shows the THz waveforms transmitted through the Si wafer as a function of the pump-probe delay time. Only Pulse *B* is displayed in the figure. ZnTe (thin black lines) and GaP (thick red lines) crystals were used for EO sampling. The energy density of the optical pump pulse penetrating into the Si wafer was $3.3\ \mu\mathrm{J/mm}^2$. When the THz pulse arrives at the front surface prior to the pump pulse ($\Delta t << 0$), the THz Pulse *B* has just completed a round trip between the back and front surfaces in the Si wafer after separating from Pulse *A* and reaching the detector. At Δt = -2 ps, the waveforms measured by the GaP crystal are identical to those from the ZnTe crystal. When the pump pulse excites the front surface no later than the THz reflection, the waveform of the reflected THz pulse changes. Around $\Delta t = 0$, when the THz light interacts with the moving plasma front near the input surface, Doppler reflection is expected. The results using the GaP crystal clearly reveal the pulse compression of the THz light pulse due to the Doppler reflection, indicating the frequency up-shift in the spectrum. By contrast, the ZnTe crystal cannot monitor the Doppler reflection accurately due to the lack in its time resolution. At a delay time of 2 ps, the reflected THz waveform phase changes by π, with a temporal advance of 0.67 ps as shown by the thick arrow in Figure 2(a). This is clear evidence of the reflection from the static plasma front in the Si close to the input surface.

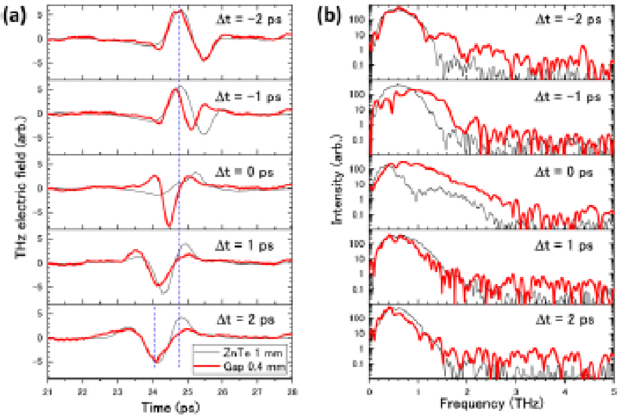

Fig. 2. (a) Waveforms and (b) Fourier transformed spectra of the reflected THz light as a function of the pump-probe delay Δt. The thin black and thick red lines show the results measured by the 1-mm thick ZnTe and 0.4-mm thick GaP EO crystals, respectively.

Figure 2(b) shows the time-dependent intensity spectra obtained by the Fourier transform of the THz waveforms shown in Figure 2(a). Before and after the pump light irradiation, the up-shift is not observed in the spectra since the reflection planes for the THz pulse are the static Si–air interface and the static plasma front, respectively. In contrast, the frequency up-shift induced by the relativistic Doppler reflection is clearly observed around Δt = 0 when the plasma front is moving. Due to the larger bandwidth, the larger frequency up-shift can be obtained (approximately 3 THz) by the GaP crystal rather than by the ZnTe crystal.

The THz-TDS provides not only the intensity, but also the phase spectra. Therefore, the phase shift, which is the difference between the phases of the reflected THz light with and without the irradiation of the optical pump pulse, is easily obtained. The improvement in the time resolution in the THz-TDS measurement enables the accurate analysis of the spectral phase. Figure 3 shows the phase shift spectra of the reflected THz light as a function of the pump-probe delay time. At the negative delay time, the THz light is reflected at the air–Si boundary without influence from the optical pump light and no phase shift was observed. At Δt = 0, the THz light interacted with the moving plasma front close to the surface and a π phase jump of approximately 0.7 THz was found, which is one of the signatures of Doppler reflection. After the pump light is completely absorbed by the Si and the static plasma layer generated ($\Delta t > 0$), the phase shift monotonically decreases as the frequency increases. The observed phase shifts are compared to the one-dimensional finite-difference time-domain (1D-FDTD) simulation in Figure 3. The Fresnel reflection at the negative delays and the static plasma reflections at the positive delays are well reproduced by the 1D-FDTD simulation. However, the 1D-FDTD simulation poorly reproduces the observed result during the Doppler reflection around Δt = 0. One reason for this is that the simple Drude model adopted in the 1D-FDTD simulation is not suitable for describing the interaction of the instantaneously generated dense plasma with the THz light.

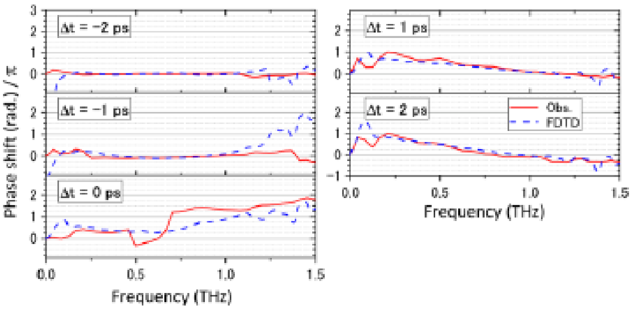

Fig. 3. Phase shift spectra of the reflected THz light as a function of the pump-probe delay time Δt. The solid red and blue dashed lines show the observed and calculated spectra, respectively.

Relativistic Doppler reflection has a potential application in pulse compression and spectral broadening of THz light for THz-TDS. Solid materials with optical functions (e.g., semi-conductors and perovskite) have been investigated by optical-pump THz-probe time-resolved spectroscopy to reveal free-carrier and exciton dynamics in optically excited states. The frequency up-shift investigated in this study was realized by the conventional THz generation scheme and easily coupled with the optical-pump THz-probe experiment. The up-shift will be applied to reveal the phonon and exciton dynamics in optically excited materials.

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