Electrical control of terahertz frequency conversion from time-varying surfaces

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Abstract: We investigate the electrical control of frequency conversion from a time-varying interdigitated photo-conductive antenna (IPCA) and time-varying metasurface in the terahertz (THz) frequency range. Ultrafast near-infrared (NIR) optical pulses rapidly modify the conductivities of the IPCA and metasurface; however, external voltages can retard this conductivity transition. Thus, external voltages can be used to control the frequency conversion process based on the interaction between the THz waves and the time-varying surfaces. In the IPCA, both frequency up- and down-conversion processes are suppressed by external voltages. However, in the metasurface, the down-conversion is dramatically suppressed by external voltages, whereas the suppression on the up-conversion is less effective.

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1. Introduction

Manipulating the frequency of light is one of the essential techniques in optics and photonics for diverse applications. In the visible and infrared (IR) frequency ranges, frequency conversion is primarily based on nonlinear processes such as harmonic generation or parametric processes [1]. Such nonlinear processes have been well-developed to mature levels and are easily applicable, offering sufficient conversion efficiencies and frequency shifts [2,3]. To induce nonlinear frequency conversion processes, the intensity of the input light must be sufficiently high and an appropriate nonlinear medium is required. However, especially in the THz frequency range, nonlinear frequency conversion remains in its infancy because of limited intense THz sources and nonlinear media.

An alternative method, proposed instead of using material nonlinearities, is linear frequency conversion employing time-varying media. Based on the theoretical predictions of such frequency conversion [4–8], experiments have been demonstrated in rapidly growing plasmas [9,10] or temporally modulated waveguide structures [11,12]. This type of frequency conversion provides a constant conversion efficiency regarding the input power and therefore, is potentially applicable to achieve comparable or even higher conversion efficiencies compared to nonlinearity-based frequency conversion methods in the THz spectral range [13–17]. A. Nishida et al. first demonstrated such frequency conversion in the THz frequency range from flash ionization in ZnSe [18], and recently, several experimental realizations have been demonstrated from rapid time-varying surfaces [19–21]. In particular, the rapid rate of the time-varying yields a remarkable amount of conversion in this kind of frequency conversion process [20], and it is both theoretically and experimentally proven that the frequency and phase of the converted wave can be controlled to a large degree by engineering the spectro-temporal behavior of the time-varying surface [21]. Furthermore, because a rapid
change in the surface conductivity is the key to this frequency conversion process, controlling the rate of such change can be used to control the frequency conversion process.

In this work, we report the experimental results on the active electrical control of effective surface conductivities on an interdigitated photo-conductive antenna (IPCA) and metasurface. Subsequently, we demonstrate that the frequency conversion based on the change of conductivity is electrically controllable. The effective surface conductivity changes in our IPCA and metasurface fabricated on a gallium arsenide (GaAs) substrate are implemented by photo-carrier injection using ultrafast near-infrared (NIR) laser pulses. Without external voltage, the optical pumping changes the effective surface conductivities rapidly. When an external voltage is applied to the samples, the conductivity change in the region under the direct-current (DC) electric field is delayed by subsequent processes of inter-valley scattering and Coulomb screening. Through such electrical control over the rate of change in the surface conductivities, the frequency conversion process can be controlled. We determined that external voltages applied to the IPCA suppress both up- and down-conversion. However, for the metasurface, the suppression effect is dominant on the down-conversion whereas it is less effective on the up-conversion.

2. Experimental procedure

A conventional 8-F THz time-domain spectroscopy setup [22,23] based on a Ti:sapphire regenerative amplifier with a repetition rate of 1 kHz (Spitfire Ace, Spectra-physics) was used for the experiment. The center wavelength and pulse duration of the laser pulses were 800 nm and 100 fs, respectively. Figure 1(a) schematically illustrates our experimental setup. In this setup, a prism-cut LiNbO3 crystal was used to generate single-cycle THz waves with a tilted wavefront technique [24–27] as displayed in Fig. 1(b). These THz waves were used in our optical-pump/THz-probe (OPTP) measurements. For the frequency conversion experiments, we utilized multi-cycle THz waves modulated through five band-pass filters. Because the multi-cycle THz waves deliver a narrow bandwidth centered at 0.6 THz as indicated in Fig. 1(c), the frequency conversion can be clearly observed. A 90° off-axis parabolic mirror pair with 150-mm and 50-mm focal lengths was used to focus the THz beam at the sample position. The THz beam diameter at the sample position was estimated to be 1 mm. To detect THz waves, we used the well-established electro-optic (EO) sampling technique in a <110>-oriented 2-mm-thick ZnTe crystal. We also use a pair of polarizers to attenuate the amplitude of the THz waves to avoid the nonlinear effects in the samples [28–30] or in the THz detection part [31]. After the attenuation, the peak field amplitudes of the single- and multi-cycle THz waves were approximately 1 kV/cm. To change the effective surface conductivities of the samples, we used the NIR pump pulses with a fluence of ~2 μJ/cm² and a time delay, \( t_d \), to the THz waves. Note that the beam diameter of the NIR pump pulses was approximately 4 mm, sufficiently large to envelop the THz beam at the sample position.
h pulses are injected into the IPCA, and the amount and rate of change vary

3. OPTP measurement and frequency conversion of an IPCA with external voltages

This section presents the effective conductivity changes and frequency conversion results for an IPCA achieved by varying the external voltages. As shown in Figs. 2(a) and 2(b), we fabricated the IPCA in a manner similar to previously reported IPCA structures [32,33]. The electrodes of the IPCA were made of 150-nm-thick gold with a 10-nm-thick chromium adhesion layer on a GaAs substrate; the width of the electrodes was 5 μm and the gap between the electrodes was 6 μm. On the electrodes, we also patterned a block layer that was isolated from the electrodes by a polyimide spacer with a thickness of 1 μm.

The temporal changes of the effective surface conductivity for the IPCA were measured by the OPTP measurements as shown in Figs. 2(c)–2(h). Figures 2(c) and 2(d) display the transmitted spectral change with and without a voltage of 21 V as a function of τd, respectively, and Figs. 2(e)–2(g) are slice views obtained from the measurements. In particular, Fig. 2(e) displays the selected transmission spectra at τd = 2 ps with and without a voltage of 21 V, whereas Fig. 2(f) shows the selected transmission spectra at τd = 10 ps. To illustrate the time-varying behavior of the effective surface conductivity, we plot the transmitted spectral amplitude at 0.6 THz in Fig. 2(g). To compare the conductivity changing rates more clearly, we also show the normalized changes of the amplitudes scaled by their maximum and minimum values in Fig. 2(h). Here, the maximum and minimum values denote the average of the amplitudes before optical pumping and the average of the changed amplitudes after optical pumping, respectively. As indicated in the results, the effective surface conductivity of the IPCA increases and the transmission of the THz waves decreases after the NIR pulses are injected into the IPCA, and the amount and rate of the change vary
with the external voltage. Specifically, the rate of change in conductivity decreases as the voltage is increased.

Inter-valley scattering and Coulomb screening can explain such a delay in conductivity change with the external voltage. First, when the photo-excited carriers are formed under a strong DC electric field, the carriers are accelerated by the DC field, and some of the electrons immediately fall into the satellite valleys by the inter-valley scattering process [34–36]. Because the effective mass of the electrons in the satellite valleys (X- or L-valley) is considerably greater than that in the main valley (Γ-valley), the electrons in the satellite valleys contribute minimally to the increase in the overall surface conductivity [28–30,34–36]. However, the carriers accelerated by the DC field also move closer to the electrodes to establish Coulomb screening regions [33,37,38]. After the Coulomb screening regions are established, the majority of the electrons are no longer under the DC field and the electrons in the satellite valleys return to the Γ-valley within picoseconds. Consequently, the external voltage retards the temporal changing process of the surface conductivity as indicated in Fig. 2(h). Note that the external voltages also cause the depletion of carriers and consequently increase the changed transmission amplitudes as displayed in Fig. 2(g).

Because the frequency conversion from a time-varying surface is strongly related to the temporal changes of the surface conductivity [20,21], the frequency conversion method using time-varying IPCA is also electrically controllable. Experimental results of the frequency conversion from the IPCA with different external voltages are illustrated in Fig. 3. Figure 3(a) displays the measured waveforms through the time-varying IPCA and Fig. 3(b) indicates their subtracted waveforms from the waveform measured through the IPCA without pumping. Here, we used multi-cycle THz waves (Fig. 1(c)) for precise observation of the frequency conversion and chose the time delay placing the pump beam at the maximum envelope of the input THz wave. Note that this time delay ensures the maximum conversion efficiency [21]. As indicated in Fig. 3(b), the external voltage reduces both the rate and amount of the temporal variation, and subsequently, a reduction of the frequency conversion is observed from such electrical modulation in spectra as shown in Figs. 3(c) and 3(d). To verify the frequency conversion, we also plot the reference spectrum measured through a GaAs substrate as shown with the dashed line in Figs. 3(c) and 3(d). Here, we display both the
logarithmic and linear plot to indicate clearly the spectra in a full view and the up- and down-converted frequency components in an extended view, respectively. As depicted in Figs. 3(c) and 3(d), frequency up- and down-conversion well occur without external voltages, and these converted spectral components reduce when applying the external voltage. The frequency conversion occurs continuously owing to the trade-off relation between the conversion efficiency and relative amount of frequency change [21]. Thus, we can evaluate the conversion efficiency by $|\hat{E}_i(\omega)| / |\hat{E}_c(\omega)|$ where $\hat{E}_i(\omega)$ is the spectral amplitude of the input wave at the input frequency, and $\hat{E}_c(\omega)$ is the spectral amplitude of the converted wave at a frequency, $\omega$. For a frequency change of 0.3 THz, corresponding to half of the fundamental frequency, both to lower and higher frequencies, we achieved an up-conversion efficiency of $1.4 \times 10^{-4}$ at 0.9 THz and a down-conversion efficiency of $6 \times 10^{-5}$ at 0.3 THz without voltages. With a voltage of 21 V, the up- and down-conversion efficiencies are reduced to approximately $2 \times 10^{-7}$.

Fig. 3. Experimental results of frequency conversion through IPCA. (a) Transmitted THz waveforms through time-varying IPCA with (red) and without voltage of 21 V (blue) under optical pumping. Black curve displays transmitted THz waveform without optical pumping. (b) Subtracted waveforms from waveforms measured through IPCA without optical pumping. Red and blue dashed lines indicate the field profiles with and without voltage, respectively; red and blue solid lines indicate their envelopes. (c) Logarithmic plot of experimental results of frequency conversion from IPCA with different external voltages. Dashed curve indicates reference spectrum measured through GaAs substrate. (d) Linear plot of experimental results of frequency conversion from IPCA with different electric voltages.

The presence of the external voltage changes the amount and rate of the conductivity change, both of which contribute to the frequency conversion process. However, the change in the rate is the main origin of the change in the frequency conversion. To understand the relationship between the rate of change in conductivity and the frequency conversion, we established a simple model considering only the changing rate and calculated the frequency conversion. In this calculation, we assume that the surface of the IPCA was homogeneous and
infinitesimally thin. With this assumption, we are able to set the transmitted field through the surface, \( E_t(t) \), as a function of the input field, \( E_i(t) \), and the surface current, \( J(t) \), as follows,

\[
E_t(t) = \frac{2}{1+n}[E_i(t) - \frac{Z_0}{2} J(t)],
\]

where \( n \) is the reflective index of GaAs and \( Z_0 \) is the impedance of free space [21,23]. Because \( E_i(t) \) does not contain a frequency component at the converted frequency, the converted frequency component originates purely from \( J(t) \). For simplicity, we assume that the surface conductivity of the photo-excited layer, \( \sigma \), is wholly non-dispersive yet time-varying as \( \sigma(\omega,t) = \sigma(t) \). Note that the structure of the IPCA is similar to wire-grid structures, which are non-dispersive [39,40], and the measured dispersion as indicated in Figs. 2(e) and 2(f) is not strong. With this assumption, \( J(t) \) would have a linear relationship with \( E_t(t) \) as follows:

\[
J(t) = \int \sigma_e(t) \delta(t-\tau) E_i(\tau) d\tau = \sigma_e(t) E_t(t)
\]

Here, \( \delta(\tau) \) is the Dirac delta function. From Eqs. (1) and (2), \( E_t(t) \) becomes a function of \( E_i(t) \), as follows:

\[
E_t(t) = \frac{2}{1+n + Z_0 \sigma_e(t)} E_i(t)
\]

To fit the OPTP measurements in Fig. 2, we assume that \( \sigma(t) \) is zero for \( t_d < 0 \) and \( \sigma(t) \) is \( (1+n)/4Z_0 \) for \( t_d >> 1 \) ps. Note that such values set the amplitude transmissions normalized by the reference to 1 and 0.8 for \( t_d < 0 \) and \( t_d >> 1 \) ps, respectively. The conductivity changing behavior with external voltages relates to the inter-valley scattering [34–36], and the conductivity increment may follow two routes. First, the optically doped carriers that remain in the \( \Gamma \)-valley immediately contribute to the increment of the surface conductivity. Secondly, the doped carriers that are scattered to the satellite valleys do not contribute on the surface conductivity increment for \( t_d = 0 \); however, these carriers relax into \( \Gamma \)-valley after the Coulomb screening region is established and then contribute to the increment of the surface conductivity [38]. Considering these two processes, we can set a model equation for \( \sigma_e(t) \) as follows:

\[
\sigma_e(t) = (1-x) \frac{1+n}{4Z_0} u(t-t_d) + x \frac{1+n}{4Z_0} u(t-t_d)[1-e^{-\frac{t_d}{\tau}}]
\]

where \( x, \tau, t_d \) and \( u(t) \) are the ratio of the inter-valley scattering, relaxation time from satellite valleys to \( \Gamma \)-valley, pump beam arrival time, and unit step function, respectively. Here, the establishment of the Coulomb screening region is assumed to be a rapid process for simplicity. In the expression of \( \sigma_e(t) \), \( x \) would conclusively determine the conductivity transition time, which increases with the external voltages in the experiments. For a small \( x \), a rapid change from \( E_i(t) \) is dominant in \( E_t(t) \), and the Fourier transform of \( E_t(t) \) has numerous converted frequency components in the frequency domain. For a large \( x \), the slow change from \( E_i(t) \) becomes dominant in \( E_t(t) \), implying a reduction of the converted frequency components.
Figure 4(a) represents the calculated amplitude transmission based on the modeled $\sigma_{t}(t)$ with different inter-valley scattering ratios. Here, we set $\tau_{r}$ as 2 ps from the previous study [29]. Note that the calculated amplitude transmission in Fig. 4(a) appears to be different from the corresponding experimental measurements in Fig. 2(h). This inconsistency could derive from the limited temporal resolution of the OPTP measurement considering that the conductivity change of GaAs with ultrafast laser pumping is considerably faster than a picosecond level [33,41]. With an assumption of the rapid conductivity change without voltages, the point spread function for the temporal measurement can be obtained by differentiating the OPTP result with 0 V, as indicated in Fig. 4(b), and we are able to reproduce the OPTP measurement by convoluting it to the calculated amplitude transmission. Such a reproduced amplitude transmission is displayed in Fig. 4(c), which indicates good agreement with the experimental result in Fig. 2(h). Figure 4(d) presents the calculated results of the frequency conversion based on the proposed model with different inter-valley scattering ratios. We used the measured reference waveform as $2E_{t}(t)/\left(1 + n\right)$ for calculation. As shown in Fig. 4(d), weak frequency conversion is observed for longer transition times with a large $x$. As the calculation results clearly demonstrate, the decreased rate of change in the conductivity with an external voltage might be the main origin of the electrical modulation in the frequency conversion from the IPCA.

4. OPTP measurement and frequency conversion of a metasurface with external voltages

We also performed OPTP measurements and frequency conversion experiments with a metasurface with an external voltage. The metasurface structure was fabricated similar to the
IPCA case; however, the electrodes were designed to form split-ring resonators (SRR) as displayed in Fig. 5(a) (schematic illustration) and Fig. 5(b) (microscopic image). The lattice constant, electrode width, and gap width of the metasurface were 81 μm, 5 μm, and 4 μm, respectively, and the thickness of the metallic structure was identical to that of the IPCA. In this structure, the DC electric field was spatially concentrated in the gap area by an external voltage, whereas it was relatively weak in the remaining region. Thus, owing to the inter-valley scattering and Coulomb screening, as in the IPCA case, the conductivity transition in the gap region could be delayed by external voltage, as indicated in Fig. 5(c). Conversely, the conductivity in the remaining region would undergo a sudden change due to optical pumping regardless of whether the external voltage is applied.

Fig. 5. Metasurface and the related OPTP results. (a) Schematic illustration and (b) microscopic image of metasurface. Arrows in (a) indicate polarization of input THz wave. (c) Schematic diagram of temporal conductivity change on metasurface with and without external voltage. (d) Measured transmission spectra through metasurface without external voltage as function of time delay, t₀ and (e) measured transmission spectra with external voltage of 32 V. (f) Selected transmission spectra at t₀ = 2 ps with (red) and without voltage (blue). Black line indicates transmission spectrum through metasurface without optical pumping. (g) Selected transmission spectra at t₀ = 10 ps with and without voltage. (h) Numerical simulation of transmission spectra through metasurface with conducting layer for entire exposed region (blue), conducting layer of region except for gap (red) and without conducting layer (black).

This electrical modulation of the spatiotemporal conductivity distribution leads to dispersive changes in the time-varying behavior of the metasurface. The time-varying behavior of the metasurface and its electrical modulation were investigated with OPTP measurements as shown in Figs. 5(d)–5(g). Owing to the SRR structures, the metasurface originally exhibits deep resonance at 0.6 THz. Without external voltage, the resonance is suddenly dulled and red-shifted by optical pumping, and the dulled resonance is maintained for more than ten picoseconds as shown in Fig. 5(d). Conversely, the transmission spectra of
the metasurface with an external voltage of 32 V indicate blue-shifted resonance immediately after the optical pumping as presented in Figs. 5(e) and 5(f). This type of resonance blue-shift originates from the low conductivity of the gap region due to inter-valley scattering. However, the blue-shifted resonance gradually moves to a lower frequency as the time delay increases, eventually resembling the resonance measured without external voltage as shown in Fig. 5(g). We attribute this resonance shift to the conductivity recovery of the gap region due to Coulomb screening.

To verify that the blue-shift in resonance originated from the low conductivity in the gap region, we also performed a finite difference time domain (FDTD) simulation as displayed in Fig. 5(h). In this plot, the black, blue, and red lines indicate the results without a photoconducting layer, with a photo-conducting layer on the entire photo-exposed region, and with a conducting layer everywhere apart from the gap region, respectively. In the FDTD simulation, we used a Drude model for the top 0.74 μm of the GaAs substrate, which corresponds to the penetration depth of the NIR pump beam [42], to incorporate the photoconductivity of the substrate. The plasma frequency and collision frequency of photo-doped GaAs were set to be $6.2 \times 10^{13}$ rad/s and $2.5 \times 10^{15}$ rad/s, respectively. Because the calculation demonstrates results consistent with the transmission result for $t_d = 2$ ps in Fig. 5(f), the resonance blue-shift caused by the external voltage must be a result of the conductivity reduction in the gap region due to inter-valley scattering.

![Fig. 6. Frequency conversion results through metasurface with external voltages. (a) Converted spectral amplitudes with an external voltage of 32 V (red) and without (blue) at (a) 0.3 THz and (b) 0.9 THz as function of time delay, $t_d$. Amplitudes are normalized by maximum amplitude of reference spectrum. (c) Logarithmic and (d) linear plots of frequency conversion from metasurface at $t_d = -6$ ps with different external voltages. Dashed line indicates reference spectrum as measured through GaAs substrate.](image)

The frequency conversion from the metasurface can also be electrically modulated by electrical controlling of the time-varying behavior. Figure 6 illustrates the experimental results of the frequency conversion from the metasurface with an external voltage. Figures 6(a) and 6(b) illustrate the converted frequency components with and without an external voltage of 32 V as a function of the time delay at 0.3 THz and 0.9 THz, respectively. We used
the multi-cycle THz waves again, as indicated in Fig. 1(c), for these measurements, and we plotted the results with the time delay as defined in the OPTP measurement. In our measured data, both up- and down-conversion occurs when the multi-cycle THz waves and NIR pump beams overlap. The external voltage suppresses both the up- and down-conversion processes in general; however, the down-conversion components are suppressed more dominantly compared to the up-conversion components. We attribute this asymmetry of electrical modulation for up- and down-conversion to the blue-shifted resonance induced by the external voltage. Because the frequency conversion is related to the dispersion of the time-varied surface [21], the blue-shifted resonance causes the re-radiation from the metasurface with higher frequencies. Note that that the theoretical model introduced above is not valid for this type of electrical modulation, as the metasurface is strongly dispersive. We also note that the fringes at the converted frequencies originate from the interference between two complex components of the frequency conversion for single-band inputs [21].

At a particular time delay position, the asymmetry of the electrical modulation for up- and down-conversion becomes more apparent. Figures 6(c) and 6(d) display the experimental results of the frequency conversion at $t_d = -6$ ps with different external voltages in a logarithmic and linear plot, respectively. At this time delay, the down-conversion is substantially reduced by the voltage, whereas the up-conversion indicates only a marginal reduction. Using the formula, $|\hat{E}(\omega)|^2 / |\hat{E}(\omega_0)|^2$, for the shift amount of 0.3 THz, we obtain the up-conversion efficiency of $1.7 \times 10^{-4}$ and the down-conversion efficiency of $1.9 \times 10^{-4}$ without voltages. At a voltage level of 32 V, the up-conversion efficiency is reduced to $1.5 \times 10^{-4}$, whereas the voltage reduces the down-conversion efficiency to the level of the input spectrum. Consequently, the spectral broadening (up- and down-conversion)-type frequency conversion is altered to the up-conversion type by the external voltage. Without voltages, we obtain up-conversion efficiencies of $1.4 \times 10^{-4}$ for the IPCA and $1.7 \times 10^{-4}$ for the metasurface at 0.9 THz. Note that these values are less than the previously reported value of $6 \times 10^{-4}$ [21], because the time-varying surface used in the previous study was designed for the narrowband case and optimized for the frequency conversion from 0.6 to 0.9 THz, whereas the ICPA and metasurface used in the present work are designed for both up- and down-conversion processes.

Although the frequency conversion demonstrated indicates a similar behavior comparable with the nonlinear spectral broadening induced by self-phase modulation, the physical origin of the frequency conversion process from the time-varying surfaces is completely different from the nonlinear effect. The temporal change of the media is independent of the input wave, and hence, the degree of frequency manipulation and the corresponding conversion efficiency are invariant to the input power. Furthermore, noticeable frequency manipulation is even possible despite an extremely short interaction length in the thin film geometry because the external control by optical pumping can change the property of the media dramatically. Finally, an abrupt change contributes primarily to the frequency conversion process, and thus the conversion is controllable electrically by changing the speed of the conductivity change as demonstrated in this work.

5. Conclusion

We experimentally investigated the electrical control of surface conductivity changing processes and the related frequency conversion processes on a time-varying IPCA and a metasurface in the THz frequency range. When we apply an external voltage to the GaAs-based samples where photo-carriers are injected, the DC electric field induces inter-valley scattering for photo-induced electrons and prevents the increase in the surface conductivity. However, Coulomb screening subsequently occurs near the electrodes, and the scattered electrons return to the main conduction valley within a few picoseconds, resulting in a surface conductivity increment. Owing to this sequential process, the external voltage can delay the
change in the surface conductivity. Because frequency conversion based on the time-varying surface is strongly related to its rate of change, the external voltage can also control the frequency conversion process. In the case of an IPCA, the external voltage suppressed both up- and down-conversion, whereas the external voltage suppressed only the down-conversion substantially for the metasurface. This asymmetrical suppression is mainly attributed to the temporally resonant blue-shift induced by the conductivity reduction in the gap of the SRR. As indicated in the two demonstrated examples, electrical control could be widely implemented in a frequency conversion scheme based on a time-varying surface platform. Such an implementation could diversify frequency conversion techniques in the THz frequency range.

Funding
National Research Foundation of Korea (2019R1A2C3003504, 2016R1A6A3A03009053, 2017R1A4A1015426).

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