Coherently controlled terahertz source for a time domain spectroscopy system via injection current in bulk ZnSe

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We have observed terahertz generation via injection current induced by harmonically related two-color beams in an unbiased ZnSe bulk at room temperature using a femtosecond Ti:sapphire oscillator. The terahertz intensity is just several times smaller than that obtained via optical rectification and further enhancements are believed possible. Experimental results demonstrate that the terahertz radiation is mainly attributed to the transition from the split-off band. This conclusion provides a novel approach to effectively generate a broadband and coherently controlled terahertz radiation, which leads to practical applications of terahertz radiation via this mechanism.

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In recent years, induced by harmonically related two-color light beams with ω below and 2ω above the bandgap, the so-called "injection current" has been predicted [1–3] and observed [4,5] in semiconductors. It is known as the quantum interference in the one- and two-photon absorption processes (SPA and TPA) [1–5] and has been shown to be a resonance-enhanced third-order nonlinear process [4,6]. As this current occurs on the timescale of the duration of the femtosecond laser pulse, this process can generate electromagnetic radiation at terahertz frequencies. In the case that the influence of space-charge effects are insignificant, the terahertz field can be given by a form of four-wave rectification (supposing optical fields collinearly polarized) [6–8]:

$$E_{\mathrm{THz}} \propto E_{2\omega} E_{\omega}^{2} \sin(2\Phi_{\omega} - \Phi_{2\omega}), \tag{1}$$

where $E_{\omega,2\omega}$ are the optical-field amplitudes and $2\Phi_{\omega} - \Phi_{2\omega}$ is the relative phase parameter of the two beams.

Experimentally, terahertz generation via injection current has been observed in GaAs [7], InP [8], and even indirect bandgap materials, such as Si [6,9] and Ge [9]. It provides a convenient way to research the fast dynamics of carriers in semiconductors and can be used as a coherently controlled broadband terahertz source. However, the current injection efficiency is predicted to sharply decrease with increasing bandgap [3,10]. That is, wide bandgap materials are not suggested for terahertz generation using this concept. Therefore, according to the specific wavelength (~800 nm) used in most terahertz time domain spectroscopy (TDS) systems, this mechanism is not suitable for utilization in terahertz systems except when an optical parameter amplifier is employed.

Here we report observation of terahertz generation via injection current in bulk ZnSe induced by the beam from a Ti:sapphire femtosecond oscillator and the second harmonic. The characteristics of the terahertz emission have been investigated. Experimental evidence shows there is a notable enhancement of terahertz intensity in the case that the split-off band is just excited. Using a reflection scheme, an intensity of terahertz only several times weaker than that

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via optical rectification is achieved, which is adequate for practical application in a terahertz TDS system. Compared with the conventional means (optical recertification and photoconduction), such a terahertz source has some characteristic properties: (1) the intensity and polarity of terahertz can be fully controlled by the relative phase of the pump beams [6,9], and (2) the polarization of terahertz is almost exclusively determined by the pump polarization [6,9] and can be easily tuned.

The optical source for the experiment is a 76 MHz femtosecond Ti:sapphire oscillator, which produces 150 fs pulses with an average power of 1.2 W. As sketched in Fig. 1, the beam from the laser is split into pump and probe beams. The pump is focused on a beta-barium borate (BBO) crystal to generate a few tens of milliwatts of second harmonic. To gain a stable phase-control mechanism with sufficient scan range, a two-color Mach–Zehnder interferometer is employed and, in the 2ω arm, a pair of fused-silica wedges are used to precisely control the relative phase parameter $\Delta\Phi = 2\Phi_{\omega} - \Phi_{2\omega}$. Then the two beams are spatially and temporally overlapped and focused at normal incidence on an 0.5 mm thick (100) oriented ZnSe sample, collinearly polarized along a crystal axis. To achieve the concordance of the wave fronts, in each arm of the interferometer, a collimation lens is used. The fundamental and the second harmonic beams are focused to 300 and 150 μm spots, respectively, with corresponding irradiance of 140 and 12 MW/cm^2. The terahertz radiation is collected with an off-axis parabolic mirror and then focused into a 1 mm thick (110) oriented ZnTe crystal for electro-optic sampling. The terahertz polarization can be determined as the ZnTe electro-optic crystal is rotated about its normal. To reduce the effects of water vapor absorption, the section of terahertz propagation is enclosed in a box, which is purged with dry nitrogen gas. Besides such a transmission scheme, a reflection scheme is also employed to relieve the phase mismatching and the bulk absorption. For the purpose of intensity comparison, an 0.5 mm (110) ZnTe instead of the (100) ZnSe sample is also used for optical rectification induced by the single fundamental beam.

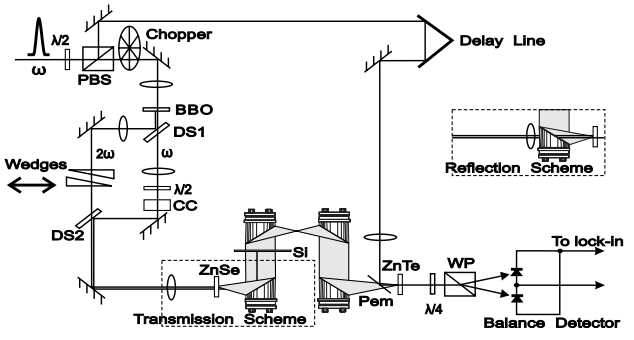

Fig. 1. Experimental setup for coherently controlled terahertz generation and detection. DS, dichroic splitter; CC, compensation crystal; Pem, pellicle mirror; WP, Wollaston prism. In the reflection scheme, the two-color beams are focused through a hole in the collecting parabolic mirror.

The ZnSe we used here is a zinc-blend-type semiconductor with bandgap $E_g = 2.67$ eV and split-off energy $\Delta = 0.43$ eV at room temperature [11]. According to the SPA (400 nm) and TPA (800 nm) coefficients [12,13], near-surface carrier densities of $N_{2\omega} = 4 \times 10^{17}$ cm^{-3} and $N_{\omega} = 3 \times 10^{13}$ cm^{-3} are excited, respectively. The plasma frequency is out of the terahertz region and the influence on terahertz intensity is not important.

Figure 2 shows the temporal dependence of the electro-optical signal using the transmission scheme. For comparison, the signal of optical rectification in ZnTe is also shown, and is about 25 times larger. Here the wavelength of the femtosecond oscillator is tuned as 770 nm for a clear presentation with high signal-to-noise ratio. The terahertz emission via injection current is linear polarized and the polarization direction is the same as the pump. The measurements rotating 2π around the ZnSe sample normal indicate that the terahertz intensity and polarization are almost exclusively determined by the excitation field, and the orientation of the crystal is of only minor importance.

As predicted, the terahertz field amplitude varies with the relative phase $\Delta\Phi = 2\Phi_{\omega} - \Phi_{2\omega}$. The traces for values of $\Delta\Phi$ separated by π are presented and a more detailed dependence is shown in the contour graph in the upper inset. This indicates that the terahertz intensity can be controlled, but the shape of the temporal waveform is not changeless as the relative phase is varying, which suggests considerable phase mismatching for the 0.5 mm thick sample. According to Spasenović *et al.*'s analysis [9], the terahertz intensity will be enhanced remarkably if a sample with proper thickness is used.

The lower inset in Fig. 2 shows the normalized Fourier contents of the terahertz fields. The spectrum

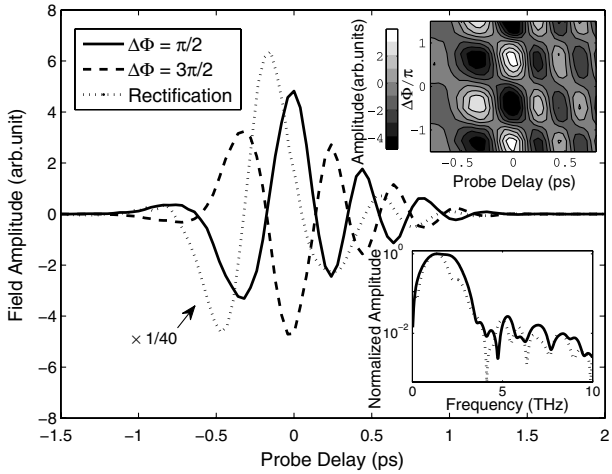

Fig. 2. Time-dependent electro-optic signals for different values of phase delay employing the transmission scheme, compared with that of the second-order rectification. A more detailed dependence on the relative phase is shown in the contour graph in the upper inset and the normalized spectra are shown in the lower inset.

reveals a broad bandwidth around 3 THz that is cut off at about 4 THz. As the bandwidth of terahertz radiation mainly lies on the pulse duration and the current relaxation time for low carrier densities [7], higher frequency may be achieved. Here the spectrum is confined by the absorption of crystals and the limited bandwidth of the electro-optic detection scheme, which is predominantly determined by the 1 mm thick electro-optic crystal used for terahertz detection [14]. Even so, higher frequencies and broader bandwidth are achieved compared with the spectrum of optical rectification.

As sketched in Fig. 3, the field amplitude is enhanced about 4 times if the reflection scheme is used. It is just about 6 times smaller than that from the rectification and the signal-to-noise ratio is up to 1200, which is strong enough for some applications, such as terahertz spectroscopic analysis and injection current gratings.

Fixing $\Delta\Phi = \pi/2$, Fig. 4 shows the dependence of terahertz field on the average pump power, I_ω, $I_{2\omega}$. Measurements have been done using both the transmission scheme and the reflection scheme. Within the experimental accuracy, the dependence of the terahertz field on the pump fits the relation of $I_\omega(I_{2\omega})^{1/2}$ very well, which proves it is a third-order process, as Eq. (1) described.

Although the divergence angle is found to be larger than the collecting angle (19° here), which is restricted by our experimental condition, an approximate measurement is made as the ZnSe sample is rotated about one of its crystal axes. According to the mechanism of injection current generation, only the pump irradiance and facula have been charged in terms of the reference frame of sample. The measured divergence angle is about 50°, which is much larger than that of the terahertz emission via optical rectification (about 4° in our experiment). That is, much more intense terahertz radiation could be expected if a collecting parabolic mirror with larger aperture and shorter focus is used.

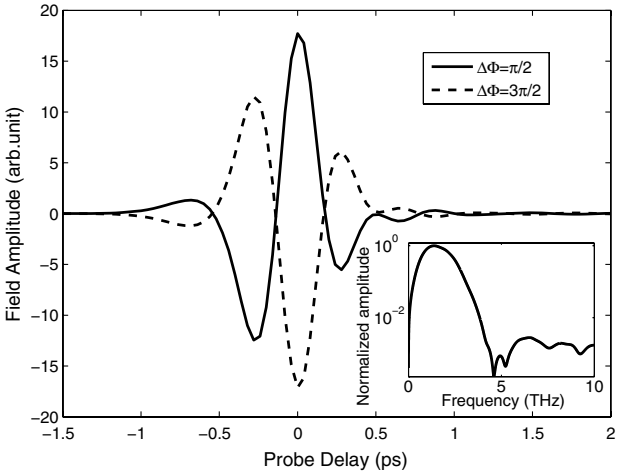

Fig. 3. Time-dependent electro-optic signals for different values of phase delay employing the reflection scheme. The normalized spectrum is shown in the lower inset.

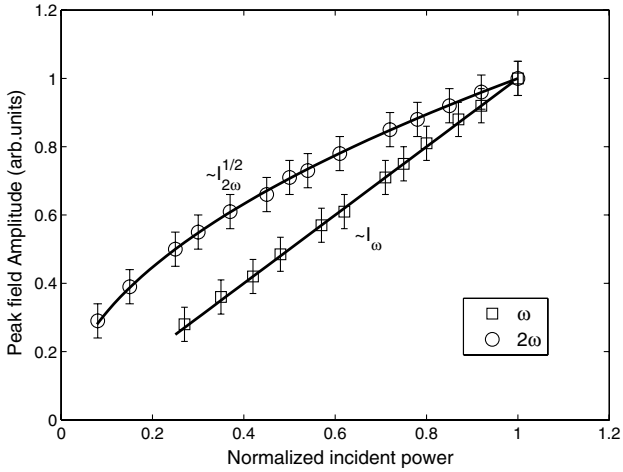

Fig. 4. Peak terahertz field strengths of injection current in ZnSe bulk for various excitation powers of the fundamental and the second harmonic pulses. The solid curves indicate linear and square-root power laws. The error bars indicate the deviation of the terahertz signal strength, which is induced by relative phase control.

While terahertz generation using this conception in several kinds of narrow bandgap materials has been reported [6–9], to the best of our knowledge, this is the first terahertz observation via injection current in such a wide gap semiconductor and using a commercial femtosecond Ti:sapphire oscillator. According to the materials and photon energy in the previous experiments, transitions from the heavy-hole (HH) band and light-hole (LH) band are dominant. As we here used large photon energy, $2\hbar\omega > E_g + \Delta$, mention should also be made of the split-off band. To reveal its contribution, the wavelength of the fundamental beam is tuned in the range of 760–850 nm. The peak terahertz field strength normalized by $I_\omega(I_{2\omega})^{1/2}$ is shown in Fig. 5. The experimental data for smaller and larger photon energy are not presented for the consideration of the group-velocity mismatching in the electro-optic sampling process. Within our experimental sensitivity, terahertz emission can be detected only in the case that the wavelength is shorter than 810 nm ($2\hbar\omega = 3.06$ eV). Then the terahertz field strength rises rapidly with the increasing of the photon energy and tends to be steady as $2\hbar\omega >$ 3.25 eV. Although it suggests that more intense terahertz emission could be obtained using laser beams with higher photon energy, theoretically it has not been predicted. As a complete theoretical description is beyond the scope of this work, here we only suggest how the variation of terahertz intensity occurs.

In ZnSe, both electron and hole currents occur; because of their different effective mass, the electrons dominate the charge displacement. Therefore, ignoring the influence of the angular distribution (which has been discussed by Sheik-Bahae [3] and van Driel [10]), in the process of injection current generation, the states of the electrons are important and it does not matter which valence band is excited. This assertion could be proved by Atanasov's calculation for

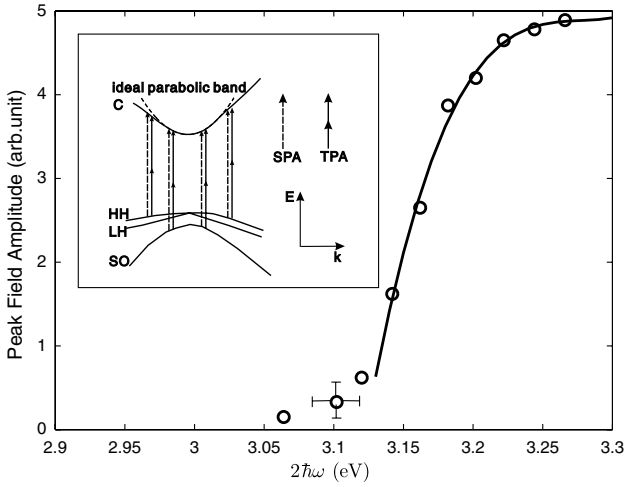

Fig. 5. Peak terahertz field strengths as a function of photon energy $2\hbar\omega$, with the solid curve indicating a trend. The field strengths are normalized by $I_\omega(I_{2\omega})^{1/2}$ with the variation in pump power. The horizontal and vertical error bars indicate the energy bandwidth of the excitation pulses and the deviation of the terahertz signal strength that is induced by the relative phase control. The inset indicates the schematic diagram of single- and two-photon transitions in ZnSe from the HH band to the conduction as $2\hbar\omega \gg E_g + \Delta$, compared with that from the split-off band. The transition from the LH band is not plotted for figure clarity.

GaAs, which shows that the efficiency of the injection current rises distinctly as the split-off band is excited.

In our experiment, for photon energy $E_g < \hbar\omega < E_g + \Delta$, the SPA and TPA processes just couple the HH and LH bands with the conduction band, injection current surely has been generated. However, as the photon energy is much larger than the bandgap, the electron current and its instantaneous rate are strongly influenced by the variation of the electron mobility. Within our detection sensitivity, no terahertz emission has been detected for the photon energy ($2\hbar\omega$) 2.9 eV ~ 3.0 eV. As the photon energy increases to $E_g + \Delta$, the top of the split-off band can be coupled with the bottom of the conduction band. The good mobility of the electrons near the conduction band edge leads an efficient generation injection current and rapid instantaneous current rate. The terahertz emission is enhanced. As the photon energy increases, the terahertz intensity rapidly rises with the increasing state density in the split-off band. On the other hand, within an independent-particle approximation, the carrier velocity, the current density, and, thus, the terahertz intensity will also increase with more excessive energy. If too high photon energy is used, however, the poorer electron mobility has influence again. The tendency of terahertz intensity becomes flat and maybe decreases if higher photon energy is employed. It should also be noted that the absorption coefficients and the phase matching also have notable influence; as the pump wavelength does not change very much, the mobility of the electron is more important.

As the band structure and electron mobility are complex and distinct for different materials, a full band structure calculation should be more reliable. From a perspective based on Atanasov's calculation and our experimental results, we can conclude, as a universal rule for most direct-bandgap semiconductors, that the current injection efficiency can be enhanced in the case the split-off band is just excited.

In summary, we provide a new terahertz source for a terahertz TDS system via injection current in an unbiased ZnSe bulk at room temperature using a Ti:sapphire femtosecond oscillator. The intensity, polarity, and polarization of the terahertz emission can be easily tuned by the pump beams. The terahertz intensity is strong enough for practical applications and can be further enhanced. The physics mechanism is discussed and it provides a method to relax the restrictions of pump wavelength in injection current generation.

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