

Ultrafast carrier generation in $\text{Bi}_{1-x}\text{Sb}_x$ thin films induced by intense monocycle terahertz pulses

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Abstract. Using terahertz-pump and terahertz-probe spectroscopy, we investigated terahertz-induced carrier generation processes in $\text{Bi}_{1-x}\text{Sb}_x$ thin films. The field dependence of the terahertz-induced transmittance change indicates distinct nonlinearity related to the Zener tunneling in narrow band-gap materials.

1 Introduction

Extreme nonlinear processes of materials such as carrier multiplication induced by intense electric field transient hold promises for future ultrafast electro-optic devices. In the terahertz frequency region, impact ionization processes in semiconductor, Zener tunneling in Mott insulator, and high harmonics generation in solid-state materials have been extensively investigated to interrogate the physical origin of the extreme nonlinearities and their possible applications [1-3]. In this context, the tunability in material parameters such as the band-gap and Fermi energy would be of great importance, yet the material system that possesses such controllability remains to be reported. In this work, using terahertz-pump and terahertz-probe spectroscopy, we demonstrate that $\text{Bi}_{1-x}\text{Sb}_x$ single-crystalline thin films deposited on Si (7X7) reconstructed surfaces will offer a promising platform for such systematic investigation, suggesting Zener tunneling as the origin of extreme nonlinearity in the narrow-gap Dirac materials even at room temperature.

2 Experiment

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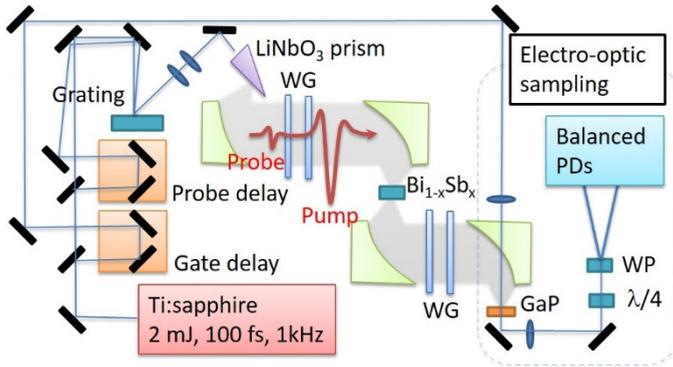


Fig. 1. Experimental setup for terahertz-pump and terahertz-probe spectroscopy. WG: wire-grid polarizer, WP: Wollaston prism, PD: photodiodes.

$\text{Bi}_{1-x}\text{Sb}_x$ single-crystalline thin films ($0 < x < 0.2$) with a thickness of 40 nm were deposited on a Si(111) 7X7 reconstructed surface by molecular beam epitaxy in an ultrahigh vacuum chamber (base pressure below 10^{-9} Pa) [4], and post-annealed at approximately 350 K after the deposition. The crystalline structure was confirmed by reflection high-energy electron diffraction, while the concentration of the antimony was checked by using X-ray fluorescence spectroscopy and X-ray photoemission spectroscopy. With increasing x , the film changes semimetal to semiconductor at $x=0.09$ [5]. The linear terahertz transmission spectra of $\text{Bi}_{1-x}\text{Sb}_x$ thin films were characterized by a homemade broadband terahertz spectrometer [6] and a Fourier-transform infrared spectrometer in the range from 0.3 THz to 50 THz. The carrier density and the damping rate were obtained by the Drude analysis for the transmission spectra.

The experimental setup for our terahertz-pump and terahertz-probe spectroscopy is shown in Fig. 1. We used a Ti:sapphire regenerative amplifier with a pulse duration of 100 fs, a repetition rate of 1 kHz, a pulse energy of 2 mJ, and a center wavelength of 800 nm. Both the pump and probe terahertz pulses were generated at a $\text{MgO}:\text{LiNbO}_3$ prism excited by the laser pulses with a tilted wavefront, and then focused on a film sample [7]. The transmitted terahertz waves were detected by the electro-optic sampling method using GaP (110) with a thickness of 400 μm . The maximum peak electric field at the sample position was 100 kV/cm. We confirmed that any nonlinearities that originate from the substrate and the electro-optic crystal could be neglected in our experimental condition.

3 Results and discussion

The terahertz-induced transmittance change of $\text{Bi}_{1-x}\text{Sb}_x$ thin films ($x=0, 0.02$ and 0.16) under the intense terahertz electric field of 80 kV/cm is shown in Fig. 2a. An increase of the transmittance signal is clearly observed near the time origin for the samples with small antimony concentrations, which is attributed to the effective relativistic acceleration of electrons along the Dirac band dispersion [8]. After a few picoseconds, the decrease of the transmittance is observed subsequently, which is observed in the samples even with large antimony concentrations.

To unveil the origin of the observed terahertz-induced transmittance changes, we measured the terahertz field strength dependence of the transmittance changes at 4 ps, as shown in Fig. 2b. The induced transmittance (ΔT) shows a nonlinear characteristic as a function of the pump field strength, suggesting that the carrier generation via strong terahertz pump pulses, such as Zener tunneling and impact ionization processes, takes place. The energy momentum conservation rule for the impact ionization process, however,

cannot be satisfied in materials with Dirac dispersion. On the other hand, the Zener tunneling process can occur in any systems under high electric fields and might be more favorable for the origin of the observed terahertz nonlinearity in $\text{Bi}_{1-x}\text{Sb}_x$. To elucidate this speculation, we calculated the field strength dependence of the transmittance change based on the Zener tunneling process [9]. As shown by solid curves in Fig. 2b, the calculated results agree very well with the experimental ones, demonstrating that the Zener tunneling is the fundamental process for the observed terahertz nonlinearity not only in semimetal samples ($x=0$ and 0.02) but also in the narrow gap semiconductor samples ($x=0.16$) even at room temperature.

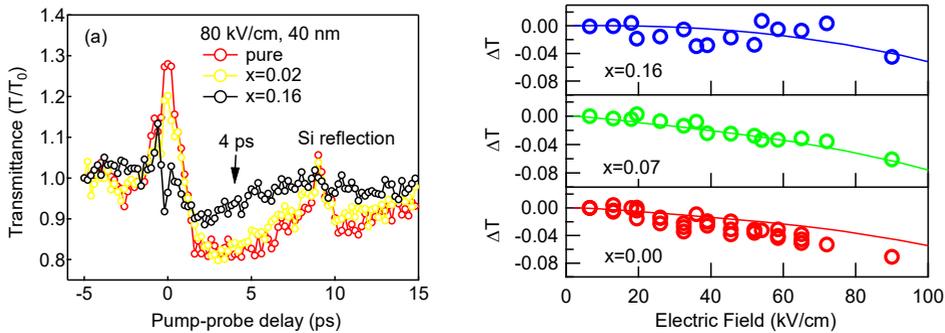


Fig. 2. (a) Normalized transient transmittance change of $\text{Bi}_{1-x}\text{Sb}_x$ thin films as a function of the pump-probe delay time. The transmittance is evaluated by the ratio of the peak electric field strength for the sample with that for the reference Si substrate. (b) The transmittance change at the pump-probe delay time of 4 ps as a function of the pump electric field strength for samples with different antimony concentrations. Solid lines indicate the calculation using the probability of the Zener tunneling and the band structure of $\text{Bi}_{1-x}\text{Sb}_x$ reported for bulk crystals [5].

3 Results and discussion

We have investigated the nonlinear carrier generation processes in $\text{Bi}_{1-x}\text{Sb}_x$ thin films with both semiconducting and semimetallic properties using terahertz-pump and terahertz-probe spectroscopy. We observed the transmission increase in the semimetal samples, and the induced transmittance decrease in both the semiconducting and semimetal samples. The former can be assigned to the effective relativistic acceleration of the carriers, and the latter is attributed to the carrier generation through the Zener tunneling process induced by the strong pump terahertz electric field.

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