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# Nonlinear Carrier Dynamics in Semi-Metal Bismuth Induced by Intense Terahertz Field

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**Abstract:** We investigated nonlinear carrier response of semi-metal bismuth under intense terahertz pulse illumination. By applying the intense terahertz field, the transmittance increases more than 10 %, indicating an increase of the effective mass.
**OCIS codes:** (300.6495) Spectroscopy, terahertz; (310.6860) Thin films; (320.7110) Ultrafast nonlinear optics

## 1. Introduction

Semi-metal bismuth (Bi) has been investigated over the years, and many interesting properties such as very small effective mass, high Hall coefficient, and non-parabolic band dispersion have been observed [1]. In addition, semimetal-to-semiconductor transition (SMSC) and structural phase transformation have been reported for the ultrathin Bi films below 30 nm and 1-2 nm, respectively [2, 3]. Therefore, the optical properties of Bi ultrathin film have been attracting attention. Recently, we have experimentally revealed the optical response of the surface state of Bi ultra-thin films by using broadband terahertz time domain spectroscopy (THz-TDS) [4]. In the present work, we have found an anomalous transmittance increase in the transmission spectrum in Bi film induced by intense THz pulses, revealing the nonlinear carrier response coming from the non-parabolic band structure of Bi.

## 2. Experiment

Single-crystalline Bi (001) film was deposited at room temperature on Si (111)-7×7 surface after the cleaning by DC resistive heating at 1500 K in an ultrahigh vacuum chamber. After the deposition, the sample was annealed at 350 K to obtain atomically flat film. The thickness of Bi film was 40 nm. Details of the method were previously described elsewhere [5].

The experimental setup of THz-TDS is shown in Figure 1(a). The light source is a Ti-Sapphire regenerative amplifier with center wavelength of 800 nm, pulse duration of 130 fs, repetition of 1 kHz, and pulse energy of 1.7 mJ. The wavefront-tilted laser pulse satisfying the non-collinear phase matching condition of optical rectification in MgO:LiNbO3 generates an intense single cycle terahertz pulse [6]. The maximum field strength of the terahertz pulse is more than 160 kV/cm. The terahertz pulse is focused onto Bi thin film by off-axis parabolic mirrors, and then the transmitted terahertz pulse is detected by electro-optic (EO) sampling method using a 0.4-mm-thick GaP (110) crystal. All the measurements are performed at room temperature and under a nitrogen atmosphere to remove the influence of water vapor. Wire grid polarizers are used to adjust the intensity of the generated terahertz wave.

## 3. Result and Discussion

Figure 1(b) shows the maximum terahertz field dependence of the transmittance of Bi. When increasing the terahertz field up to 160 kV/cm, transmittance increased by 10 % or more compared with the low-field limit. The transmittance increases by 10 % or more with increasing the terahertz field up to 160 kV/cm. This result shows the anomalous non-linearity of the carrier response in Bi. In order to explain this behavior, we analyzed the transmitted spectra using a Drude model. In this model, the complex dielectric constant is described as

$$\tilde{\varepsilon}(\omega) = \varepsilon_\infty - \frac{{\omega_p}^2}{\omega(\omega + i\gamma)}, \qquad (1)$$

where $\varepsilon_\infty$ is dielectric constant at high frequency limit, $\omega_p$ is the plasma frequency, and $\gamma$ is the damping constant, respectively. Here, the plasma frequency is expressed as

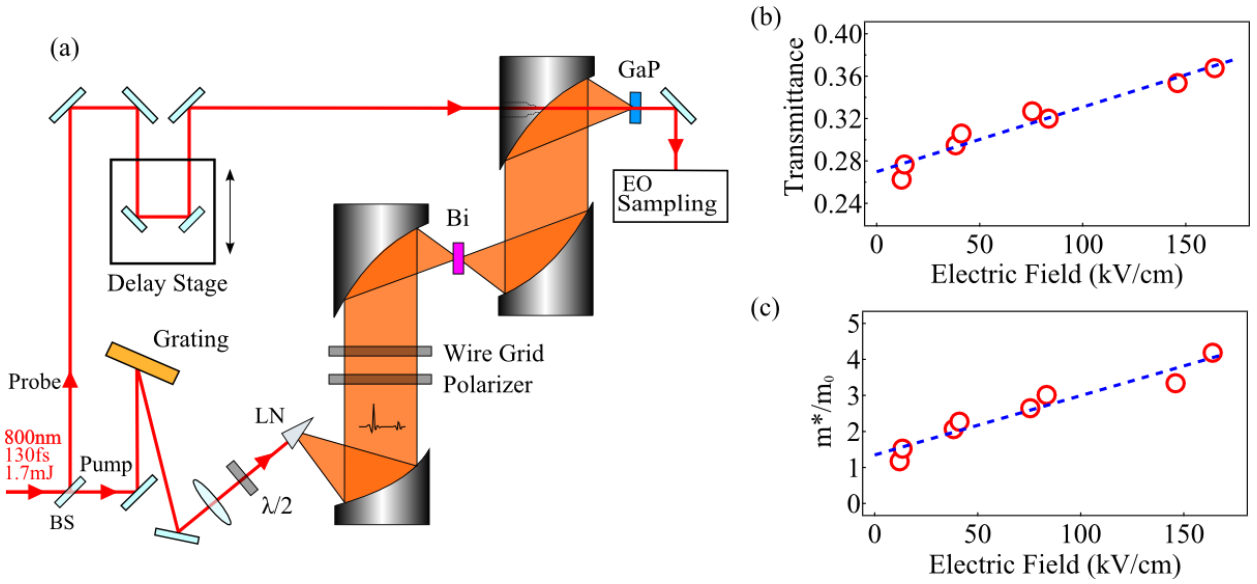


Fig. 1 (a) Schematic view of the experimental setup. Intense terahertz pulses are generated by the tilted pulse front excitation for MgO:$LiNbO_3$ (LN) and detected by EO sampling using 0.4-mm-thick GaP crystal. (b) Transmittance intensity as a function of the maximum electric field intensity. Open circle is experimental data, and a dashed line is the guide for eyes. (c) Maximum electric field intensity dependence of the effective mass of carriers normalized by the effective mass at the low-field limit (< 1 kV/cm).

$$\omega_p = \sqrt{\frac{ne^2}{\varepsilon_0 m^*}}, \qquad (2)$$

where $n$ is the carrier density, $m^*$ is the effective mass of the carriers, $e$ is the elementary charge of electrons, and $\varepsilon_0$ is the dielectric constant in a vacuum. Assuming that the carrier density is constant, the effective mass can be estimated from eqs. (1) and (2). Figure 1(c) shows the field intensity dependence of the effective mass normalized by that at low electric field limit. As shown in Figure 1(c), the effective mass dramatically increases more than 300 % under the intense terahertz field illumination. This result indicates that the electrons at L-point of Bi are intensively moved to a nonlinear region. Because the band structure of electrons at L-point is similar to that of Dirac electrons, the electrons can easily be accelerated along the linear dispersion, leading to the anomalous nonlinear response.

## 4. Conclusion

We have investigated the electric field strength dependence of the terahertz transmittance of Bi film. Based on the Drude model analysis, we found the anomalous non-linear response of electrons whose effective mass dramatically increases. The result comes from characteristics of electrons with non-parabolic band dispersion in Bi, which is analogous to that of Dirac electrons.

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