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Photomixing in topological insulator HgTe/CdTe quantum wells in terahertz regime

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We reveal that topological insulators (TI) HgTe/CdTe quantum well have a strong nonlinear optical property in the three-photon mixing. While the gapless surface state in TI can exhibit strong nonlinear effect due to the linear energy dispersion, the nonparabolic energy dispersion of the bulk state is responsible for the photo mixing effect reported here. To produce response at terahertz frequency regime from femtosecond electrical fields, the mixing efficiency is around 10^{-4} comparable to that of nonlinear semiconductor crystals. The optimal temperature for this nonlinear effect is around 100 K. The results suggest a potential application of TI in terahertz photonics. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4768781>]

Topological insulators (TIs) are new state of materials with the bulk insulating states surrounded by the non-trivial topological gapless band structures on the edge or surface for 2- or 3-dimension (2D, 3D), respectively.¹⁻⁷ These unique properties arise from the strong spin orbit coupling (SSOC) and the protection of time reversal symmetry (TRS). Under SSOC, energy gaps of TI materials collapse due to Kramers degenerate, and form an odd number pairs of helical cones, on which the spin is coupled to the momentum.⁶⁻¹⁰ This spin-momentum helical locking phenomenon is strictly protected by TRS, and thereby keeps the edge or surface states insensitive to disorder.¹¹ HgTe/CdTe quantum well (QW) was first discovered to be a 2D TI when the width of the well is thicker than 6.3 nm.¹² Recently, materials including Bi₂Te₃, Bi₂Se₃, etc., were experimentally proved to be 3D TIs by using surface sensitive techniques such as the angle-resolved photoemission spectroscopy (ARPES)^{4,13,14} and scanning tunnelling microscopy (STM).^{15,16} The TI materials exhibit intrinsic quantum spin Hall effect,^{2,12} dissipationless transport property,¹¹ anomalous magneto-electric coupling,¹⁷ and giant magnetoresistance.¹⁸ Intensive research activities in seeking for new TI materials are ongoing. Meanwhile, thanks to the well developed technique for fabricating TI materials in nanoscale, hot carrier density of the bulk can be lowered to an acceptable level,^{19,20} which paves the ways for the more detail study of properties of TIs.

Nonlinear photomixing process is a common way to obtain low frequency electromagnetic wave from 0.3 THz to 10 THz.²¹ The nonlinear response of graphene has been predicted and observed.²¹⁻²⁴ Graphene has failed to be a TI because of its weak intrinsic SOC. However, the low energy states of graphene and the surface states of TIs share a common feature, i.e., the quasiparticle obeys linear energy dispersion. Therefore, exotic phenomena in graphene such as minimum optical conductivity and quantized quantum Hall effect are also observable in TI.^{12,25,26} On the other hand, a finite bandgap can be engineered in TI and is tunable via sample thickness, suggesting greater practicality of TI-based electronic devices.²⁷ So far, TIs have shown great potential

application for optical devices, especially in terahertz regime.²⁷⁻³⁰ Zhang *et al.*²⁷ studied the linear optical conductivity of TIs, and proposed a possible application of TI from terahertz to infrared regime devices, showing that the enhanced performance of photodetection when the thickness was reduced to several quintuple layers.

The nonlinear optical response (NOR) of the surface state of HgTe/CdTe QW is similar to that of graphene (except that they have slightly different Fermi velocities)²² and hence the TI surface state is also expected to be strongly optically nonlinear. In this letter, we demonstrate that bulk state with nonparabolic energy dispersion of a TI in HgTe/CdTe QW can lead to a strong photomixing effect in the terahertz frequency regime under femtosecond irradiation. It is shown that at a moderate electrical field of around 10⁴ V/cm, generation of terahertz current from femtosecond fields with an efficiency of 10^{-4} can be achieved. Furthermore, in this class of TI structures, the nonlinear effect remains strong for temperatures up to 150 K. Unlike the gapless surface state in a TI whose existence is critically dependent on the thickness of the sample, the bulk state is much more robust. Therefore, our result should lead to wider application of TIs in electronics and photonics.

The Hamiltonian of a TI is given as⁷

$$H(\mathbf{k}) = \begin{pmatrix} h(\mathbf{k}) & 0 \\ 0 & h^*(-\mathbf{k}) \end{pmatrix}, \quad (1)$$

$$h(\mathbf{k}) = \varepsilon(\mathbf{k})\mathbf{I}_{2 \times 2} + d_a(\mathbf{k})\sigma^a,$$

Where $\mathbf{I}_{2 \times 2}$ is the 2 identity matrix, σ^a is the Pauli matrix, and $\varepsilon(\mathbf{k}) = C - D(k_x^2 + k_y^2)$, $d_a(\mathbf{k}) = [Ak_x, -Ak_y, M(\mathbf{k})]$, $M(\mathbf{k}) = M - B(k_x^2 + k_y^2)$. A, B, C, D, M are material parameters determined by QW geometry (values of those parameter of a quantum with thickness 70 nm can be found in Ref. 31).

The energy spectrum of bulk state is given as³¹

$$\varepsilon_k = C - D(k_x^2 + k_y^2) + s\sqrt{A^2(k_x^2 + k_y^2) + [M - B(k_x^2 + k_y^2)]^2}. \quad (2)$$

$s = \pm 1$ indicates valence and conduction band, respectively. The square-root term in Eq. (2) has rendered the energy spectrum highly non-parabolic. In what follows, we shall show that this non-parabolic energy dispersion leads to a strong photomixing in TI bulk state.

We consider a TI subjected to an external field in the form of $\mathbf{E}(\mathbf{r}, t) = \sum \mathbf{E}_\mu \exp[i(\mathbf{q}_\mu \cdot \mathbf{r} - \omega_\mu t)]$, where \mathbf{E}_μ , \mathbf{q}_μ , and ω_μ are the amplitude, wavevector, and frequency and μ -th wave of the electric field. The velocity of electrons in the bulk states along the direction of the electric field is given as $v_x = \partial \varepsilon_k / \partial k_x$. Without loss of general properties, we assume the applied field is along the x-direction. Under minimum coupling scheme, $k_x \rightarrow k_x - eA_x(\mathbf{r}, t)$, where $E_x(\mathbf{r}, t) = -\partial A_x(\mathbf{r}, t) / \partial t$, denote $u_x = -eA_x(\mathbf{r}, t)$,

$$\frac{\partial E}{\partial k_x} = -2D(k_x + u_x) + \frac{A^2(k_x + u_x) - 2B(k_x + u_x) \left[M - B \left((k_x + u_x)^2 + k_y^2 \right) \right]}{\sqrt{A^2 \left((k_x + u_x)^2 + k_y^2 \right) + \left[M - B \left((k_x + u_x)^2 + k_y^2 \right) \right]^2}} \quad (3)$$

To study the nonlinear effect, we expand the group velocity in successive order of the applied electric field. Up to third order of the electric field, it can be shown that the velocity components are given as

$$v_x^{(0)} = -2Dk_x + \frac{Rk_x}{\sqrt{F}}, \quad (4)$$

$$v_x^{(1)} = -2Du_x + \frac{HF - R^2k_x^2}{F\sqrt{F}}u_x, \quad (5)$$

$$v_x^{(2)} = \frac{\frac{3}{2}R^3k_x^3 + 6B^2F^3k_x - \frac{3}{2}HFRk_x}{F^2\sqrt{F}}u_x^2, \quad (6)$$

$$v_x^{(3)} = G(k_x)u_x^3, \quad (7)$$

$$G(k) = \frac{1}{F^3\sqrt{F}} \left(-\frac{5}{2}R^4k_x^4 + 3HFR^2k_x^2 - 8B^2F^2Rk_x^2 + 2B^2F^3 - \frac{1}{2}H^2F^2 \right).$$

Here we have used the shorthand notations, $R = A^2 - 2MB + 2B^2(k_x^2 + k_y^2)$, $H = A^2 - 2MB + 2B^2(3k_x^2 + k_y^2)$, and $F = (B(k_x^2 + k_y^2) - M)^2 + A^2(k_x^2 + k_y^2)$. The n-th-order current density is given by

$$\mathbf{J}_x^{(n)} = e \int_0^{2\pi} \int_0^\infty v_x^{(n)} [f(\varepsilon_k) - f(\varepsilon_k + \varepsilon_{ph}^n)] d^2k, \quad (8)$$

where $f(\varepsilon)$ is the Fermi-Dirac distribution function, ε_{ph}^n is total energy of the incoming photons in the n-th order process, and k_B is the Boltzmann constant. The upper integration limit is equal to the Fermi energy μ for $T = 0$ K. At finite temperature, the upper limit is chosen as 2μ . Due to TRS in TI, the second order velocity does not contribute to the current density. The mixing of three different fields ($\omega_1, \omega_2, \omega_3$) will result in the third order current oscillations at frequencies correspond to all

possible combinations of ($\omega_1, \omega_2, \omega_3$). Here, we are only interested in an important case where the three ultrafast fields (of the order of femtosecond) incident on the TI and search for the response of the order of terahertz. For $\omega_1 = \omega + \delta_1$, $\omega_2 = \omega + \delta_2$, and $\omega_3 = 2\omega$, the third order current at $\omega_1 + \omega_2 - \omega_3 = \delta_1 + \delta_2 = \delta$ is given as

$$\mathbf{J}_x^{(3)} = i \frac{e^4 \mathbf{E}_1 \mathbf{E}_2 \mathbf{E}_3}{\omega_1 \omega_2 \omega_3} \int_0^{2\pi} \int_0^{2\mu} G(k_x) [f(\varepsilon_k) - f(\varepsilon_k + \delta)] k dk d\theta. \quad (9)$$

Fig. 1 shows the temperature dependence of the third order current $J^{(3)}(\delta)$. The frequencies of three incident photons are chosen to be $\omega = 100$ THz and $\delta = 1$ THz. Since the reported carrier density in HgTe/CdTe QWs varies over a wide range from $2-40 \times 10^{11} \text{ cm}^{-2}$,³² we choose a range of chemical potentials, from 0.04 eV to 0.06 eV, to show the influence of carrier concentration on the third order current. We set the amplitude of three fields to be the same. There exists an optimal temperature around 50 K at which the mixing efficiency is maximum. At low temperature, only those carriers in a small region of δ below the Fermi surface contribute to $J^{(3)}(\delta)$. As temperature increases, more carriers contribute to $J^{(3)}(\delta)$. Further increasing temperature leads to more low energy carriers to conduct. Due to the unique band structure of TI, the third order velocity given in Eq. (7) is positive at high energy and negative at low energy. As a result, at high temperatures, $J^{(3)}(\delta)$ decreases with temperature.

In Fig. 2, we plot the ratio of the third order current to the linear current as a function of frequency difference δ at zero temperature. In response to three fields, there are three linear currents of equal amplitude oscillating at ω_1, ω_2 , and ω_3 . In calculating $J^{(3)}(\delta)/J^{(1)}$, we choose the one oscillating at ω_1 as $J^{(1)}$. Under an electric field of 10^4 V/cm, $J^{(3)}(\delta)$ is around 10^{-4} of that of the linear current, suggesting that terahertz generation efficiency of TI from the femtosecond lasers is comparable or slightly better than that from nonlinear semiconductor crystal commonly used in terahertz generation. The nonlinear current increases approximately as δ^2 at low δ . This can be understood from the phase space analysis. At zero temperature, the available carrier number contributing to the

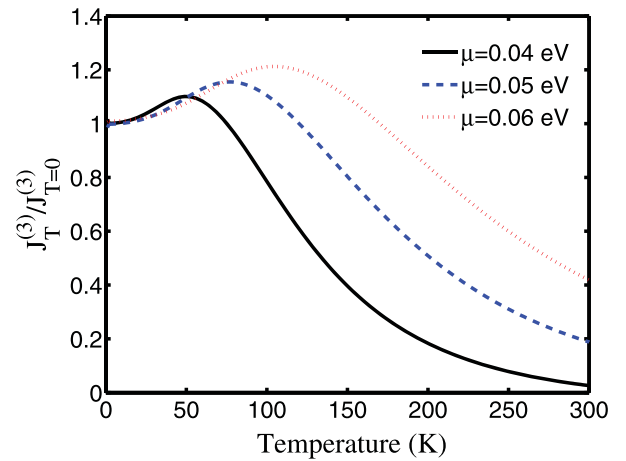


FIG. 1. Temperature dependent third order current at various chemical potentials. The electric field is 10^4 V/cm, and the frequency photons are $\omega = 100$ THz, $\omega_3 = 200$ THz.

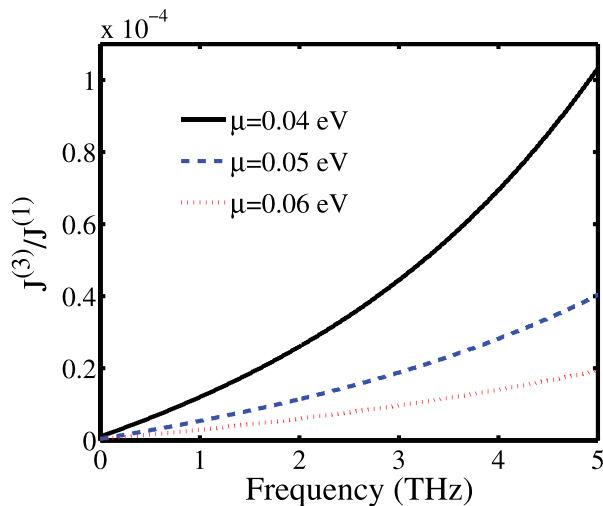


FIG. 2. The ratio of the third order nonlinear optical response to the linear optical response versus tuning frequency δ at various chemical potentials and at zero temperature. The electric field is 10^4 V/cm, and the frequency photons are $\omega = 100$ THz, $\omega_3 = 200$ THz.

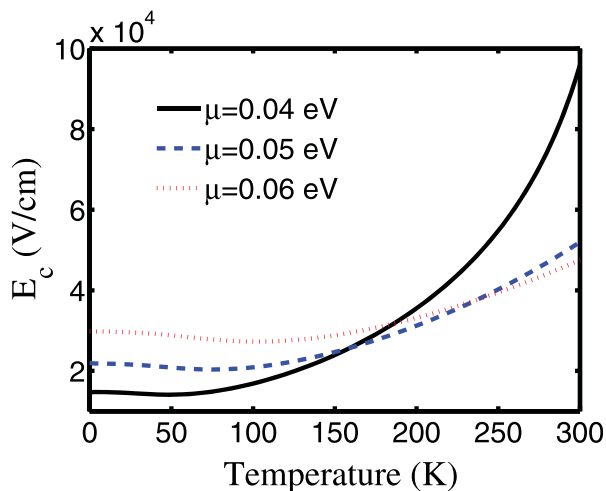


FIG. 3. Temperature dependent critical electric field at various chemical potential for $J^{(3)}(\delta)/J^{(1)} = 10^{-5}$, $\omega = 100$ THz, $\omega_3 = 200$ THz, and $\delta = 1$ THz.

linear current is nearly constant since ω is much larger than μ and nearly all electrons in the conduction band can conduct. The available carrier number contributing to the nonlinear current is directly proportional to δ . The average energy of each photoexcited carrier in the final state is also δ . As a result the nonlinear current increases approximately as δ^2 .

A critical electric field can be defined to characterize the electric field strength required for the third order NOR to achieve a given conversion efficient. Here we consider an efficiency of 10^{-5} and plot the critical field as a function of temperature in Fig. 3. It can be seen that the critical field decreases very slowly for the temperature below 100 K. This is due to the slow increase of carrier numbers contributing to the nonlinear current. At high temperature, the nonlinear current decreases rapidly with temperature, leading to a rapid increase of the critical field. In the present system where the nonlinear velocity is inversely proportional to the energy of the carrier, the nonlinear term is generally stronger for low

concentration sample at low temperature and vice versa at high temperatures. This results in a crossover of the critical field from low temperature to high temperature.

In summary, we have shown that bulk states of TI can also exhibit a strong photon mixing effect in terahertz regime under an electric field around 10^4 V/cm and frequency of the order of femtosecond. The conversion efficiency can be increased by a factor of 4 if the radiation frequencies are reduced to 50, 50, and 100 THz. Our results suggest that HgTe/CdTe QWs can be of potential use in device application for nonlinear photon mixing. Since the gapless surface state in TI is critically dependent on the thickness of the sample and the bulk state is much more robust, our finding is of particular interest in potential application of TI in photonics and optics.

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