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Nonlinear terahertz response of HgTe/CdTe quantum wells

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Abstract

2015 AIP Publishing LLC. Without breaking the topological order, HgTe/CdTe quantum wells can have two types of bulk band structure: direct gap type (type I) and indirect gap type (type II). We report that the strong nonlinear optical responses exist in both types of bulk states under a moderate electric field in the terahertz regime. Interestingly, for the type II band structure, the third order conductivity changes sign when chemical potentials lies below 10meV due to the significant response of the hole excitation close to the bottom of conduction band. Negative nonlinear conductivities suggest that HgTe/CdTe quantum wells can find application in the gain medium of a laser for terahertz radiation. The thermal influences on nonlinear optical responses of HgTe/CdTe quantum wells are also studied.

Keywords

cdte, wells, nonlinear, quantum, terahertz, response, hgte

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Nonlinear terahertz response of HgTe/CdTe quantum wells

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Without breaking the topological order, HgTe/CdTe quantum wells can have two types of bulk band structure: direct gap type (type I) and indirect gap type (type II). We report that the strong nonlinear optical responses exist in both types of bulk states under a moderate electric field in the terahertz regime. Interestingly, for the type II band structure, the third order conductivity changes sign when chemical potentials lies below 10 meV due to the significant response of the hole excitation close to the bottom of conduction band. Negative nonlinear conductivities suggest that HgTe/CdTe quantum wells can find application in the gain medium of a laser for terahertz radiation. The thermal influences on nonlinear optical responses of HgTe/CdTe quantum wells are also studied. © 2015 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4929754>]

HgTe/CdTe quantum wells (QWs) have been widely studied with interests in semiconducting heterostructures,^{1,2} formation of interface states,^{3,4} and topological quantum Hall edge states.^{5–8} One fascinating property leading to these interests is the inverted band structure, which is due to the enhancement of spin orbit coupling when the well thickness increases up to a critical value (approximately 6.4 nm).^{9,10} The band inversion lifts the first heavy-hole sub-band, H1, up above the first electron sub-band, E1, resulting in a quasizero gap, which is reopened with increasing thickness of the “well” layer.¹¹ HgTe/CdTe QWs have been found to exhibit large Rashba spin-orbit splitting,¹² quantum Andreev effect,¹³ weak anti-localization effect,¹⁰ and so on. In recent years, HgTe/CdTe QWs were first proposed as 2-dimensional (2D) topological insulator materials⁵ and were experimentally realized by König *et al.*⁶ This has led to the first direct observation of the quantum spin Hall phenomena. The topological properties of quantum spin Hall edge states reveal that two independent states propagate in two opposite directions depending on their spin degree of freedom and are protected by time reversal symmetry (TRS).^{11,14} Thus, the edge state transport suffers no back scattering even in the presence of impurities.¹⁴

Optical conductivity of HgTe/CdTe QWs has long been an interesting topic since the tunable band gap can give rise to optical response in the terahertz-far infrared regime, a technologically important frequency window. Optoelectronic devices such as THz detectors and emitters^{15,16} have applications in many areas of science and industry. It has been demonstrated that HgTe/CdTe superlattices have significant non-linear optical properties such as large $\chi^{(3)}$, THz response, and high saturation threshold. In this paper, we will study the nonlinear response of HgTe/CdTe QWs with two type of band structure without breaking the nontrivial topological property.¹⁷

In our model, we consider that the HgTe/CdTe QWs are *n*-type with Fermi energy E_F lying in the conduction band. The effective Hamiltonian of HgTe/CdTe QWs is given by BHZ model⁵

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

Here, $h(k) = \varepsilon(k)\mathbf{I}_{2 \times 2} + d_a(k)\sigma^a$, where $\mathbf{I}_{2 \times 2}$ is the 2×2 identity matrix, σ^a is the Pauli matrix, $\varepsilon(k) = C - D(k_x^2 + k_y^2)$, and $d_a(k) = [Ak_x, -Ak_y, M(k)]$ with a mass term $M(k) = M - B(k_x^2 + k_y^2)$. A , B , C , D , and M are material parameters determined by the QW geometry. The diagonal blocks in the Hamiltonian (Eq. (1)) are mutually time reversal, so the eigenvalues remain degenerate as long as the time reversal symmetry is conserved. Because of this, we can restrict our interest in the upper block. The energy eigenvalues are given as $\varepsilon(k) = Dk^2 + s\sqrt{(M - Bk^2)^2 + A^2k^2}$. $s = \pm 1$ denotes the two solutions for the H1 and E1 levels.⁵ We have omitted the parameter C , since it only shifts the energy reference of the band structure and does not affect the wave function.¹¹ B and D are the effective mass parameters, and M defines the gap between the E1 and H1 levels near the Γ point.⁵ The sign of parameter M changes with respect to the thickness of HgTe well layer. Meanwhile, the well state of a normal band structure (at $M > 0$) closes its gap at $M = 0$, and then at $M < 0$ forms an inverted band structure, where H1 is lifted as conduction band and E1 as valence band. It is well known that the sign of M/B is used to identify the topological order of the surface state.^{17–19} Generally, B is negative, so the nontrivial surface topological order only exists in the inverted band structure when $M < 0$.⁶ It has also been pointed out that the topologically nontrivial insulator cannot be adiabatically tuned to a trivial insulator as long as $M/2B > 0$.¹¹ Therefore, we fix the values of M and B , and slowly vary the values of A and D from that presented in Ref. 11 to some smaller values. This process induces an evolution in the bulk band structure from a direct energy gap type (denoted as type I) to an indirect energy gap type (denoted as type II). We present the two types of band structures in Fig. 1, for example, with the corresponding values of the parameters (A , B , D , and M) in Table I. As seen in the figure, the energy gap is about 20 meV for type I (Fig. 1(a)), and around 10 meV for type II (Fig. 1(b)).

In the formalism, we assume that *n*-type HgTe/CdTe QWs is set under a time dependent electric field

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FIG. 1. Two types of band structures in the small momentum regime: (a) direct-gap band structure denoted as type I. The gap is 20 meV at $k=0$. (b) Mexican hat band structure, denoted as type II. The minimum gap about 10 meV is not at $k=0$.

$E(t) = E_0 e^{-i\omega t}$ along the x direction. In a homogenous system, the electron distribution function, in the presence of scattering, can be derived from the Boltzmann equation

$$\frac{\partial f}{\partial t} - e\mathbf{E} \cdot \nabla_{\mathbf{k}} f = -\frac{f - f_0}{\tau}, \quad (2)$$

τ is the momentum relaxation time of electron and hole caused by impurities and phonon scattering. f_0 and f are the equilibrium and non-equilibrium Fermi-Dirac distribution function, respectively. f_0 has the form: $f_0 = \frac{1}{1 + \exp[(\epsilon_k - \mu)/k_B T]}$, where μ is the chemical potential, k_B is the Boltzmann constant; ϵ_k is the energy dispersion of HgTe/CdTe QWs. Defining $F = f - f_0$ and expanding F in successive orders of the applied field. The n th order term $F_n \propto \exp(-i\omega t)$ and $\frac{\partial F_n}{\partial t} = -i\omega F_n$. We obtain a recursion relation

$$F_n = \frac{e\tau E_0}{1 - i\omega\tau} \frac{\partial F_{n-1}}{\partial k_x} \quad (n \geq 1), \quad (3)$$

with $F_0 = f_0$. Therefore, the current density is written as

$$J^{(n)} = \frac{1}{2\pi^2} \int -e v_x F_n d\mathbf{k}. \quad (4)$$

Here, $v_x = v_F \cos \theta$ with the Fermi velocity v_F is about 5.5×10^5 m/s.²⁰ The n th order optical conductivity is related to the current density by $\sigma^{(n)} = J^{(n)}/E_0$. As a result, the n th order optical conductivity along x direction is

$$\sigma^{(n)} = -\frac{e^2}{2\pi^2 \hbar} \frac{v_F \tau}{1 - i\omega\tau} \int_0^{2\pi} \cos \theta \int_0^\infty \frac{\partial F_{n-1}}{\partial k_x} k dk d\theta. \quad (5)$$

The second order conductivity is always zero as long as the TRS is preserved. Using the fact $\frac{\partial f_0}{\partial k_x} = \frac{\partial f_0}{\cos \theta \partial k} - \frac{1}{k \sin \theta} \frac{\partial f_0}{\partial \theta}$,

TABLE I. The value of parameters A , B , D , and M .

Parameters	A (meV·nm)	B (meV·nm ²)	D (meV·nm ²)	M (meV)
Type I ¹¹	364.5	-686	-512	-10
Type II	36.45	-686	-51.2	-10

the real part of the conductivity for first order and third order can be written as

$$Re\sigma^{(1)} = -\frac{e^2}{2\hbar} \frac{\tau}{1 + \omega^2 \tau^2} \frac{v_F}{\pi} \int_0^\infty \frac{\partial f_0}{\partial k} k dk, \quad (6)$$

$$Re\sigma^{(3)} = -\frac{e^2}{2\hbar} \frac{\tau^3 (1 - 11\omega^2 \tau^2)}{(1 - 11\omega^2 \tau^2)^2 + 36(\omega^2 \tau^2 - \omega^4 \tau^4)^2} \times \frac{3e^2 v_F E_0^2}{4\pi \hbar} \int_0^\infty \left(\frac{1}{k} \frac{\partial f_0}{\partial k} - \frac{\partial^2 f_0}{\partial k^2} - k \frac{\partial^3 f_0}{\partial k^3} \right) dk. \quad (7)$$

At zero temperature, $\partial f_0 / \partial k$ is simply a Dirac delta function $-\delta(k - k_F)$. The first order and third order conductivity, in the Eqs. (6) and (7), respectively, are simplified to

$$Re\sigma_{T=0}^{(1)} = -\frac{e^2}{2\hbar} \frac{\tau}{1 + \omega^2 \tau^2} \frac{v_F (k_{F1} - k_{F2})}{\pi}, \quad (8)$$

$$Re\sigma_{T=0}^{(3)} = -\frac{e^2}{2\hbar} \frac{\tau^3 (1 - 11\omega^2 \tau^2)}{(1 - 11\omega^2 \tau^2)^2 + 36(\omega^2 \tau^2 - \omega^4 \tau^4)^2} \times \frac{3e^2 v_F E_0^2}{4\pi \hbar} \left[\frac{1}{k_{F1}} - \frac{1}{k_{F2}} \right]. \quad (9)$$

There are two solutions of Fermi wavevectors when Fermi energy lies between E_A and E_B indicated in Fig. 1(b). The scattering time is $\tau = \mu^* m^* / e = 0.57$ ps with the bulk mobility $\mu^* \sim 10^5$ cm²/(V s) and effective mass at the bottom of the conduction band $m^* \sim 0.01 m_e$.²⁰ The electric field is tuned at 5000 V/cm. In our numerical results, we plot the ratio of the real part of the third order conductivity to the first order conductivity ($\sigma^{(3)}/\sigma^{(1)}$) as a function of frequency and chemical potential under different temperatures. Since the gap of the HgTe/CdTe QWs changes rapidly due to the lattice expansion with increasing temperature, the inverted band structure survives below 60 K only. It changes to semi-metal at temperatures between 60 and 150 K, and finally becomes a normal semiconductor at higher temperatures.²¹ We will limit our analysis of the thermal effect for temperatures below 60 K.

Fig. 2 shows that the frequency dependency of $\sigma^{(3)}/\sigma^{(1)}$ for both type I and type II band structures at five different temperatures: 0 K, 4 K, 10 K, 20 K, and 60 K. Within the frequency regime considered, the nonlinear response dominates the first order linear conductivity, and has a maximum responses located at the frequency around 0.65 THz, which is independent of temperature and chemical potential. The nonlinear responses exhibit a dramatic drop toward higher frequency after the peak due to the enhancement of the linear response and the decrease in the third order nonlinear response. The nonlinear response of the two types band structures differ from each other only when the chemical potential in type II band structure lying between the E_A (energy at point A) and E_B (energy at point B), e.g., $\mu = 8$ meV, as shown in Fig. 2(c). There are clear sign changes of the ratios at low temperature. Since the linear response is always positive, the opposite sign of $\sigma^{(3)}/\sigma^{(1)}$ at low temperatures must originate from the third order nonlinear response. This anomalous response happens only when

FIG. 2. $\sigma^{(3)}/\sigma^{(1)}$ as a function of frequency. (a) and (b) are from the type I band structure with μ equalling 12 meV and 15 meV, respectively. (c) and (d) are from the type II band structure with values of μ equalling 8 meV and 12 meV, respectively.

the chemical potential lies well below energy E_B (10 meV) due to the significant response of the hole excitation close to point B, where the density of hole states are much more condensed. As shown in Fig. 1(b), the conduction band of the type II band structure is deformed to a “Mexican hat” shape, whose minimum shifts to E_A level. When the chemical potential is in the region $E_B > \mu > E_A$ at low temperature, the electrons distribute those states well below E_B , leaving highly concentrated hole states near E_B . The linear conductivity is unaffected by the field, and the 3rd order conductivity is proportional to E^2 . An electric field of 5 kV/cm is high enough that the 3rd order nonlinear effect. The ratio plotted in Fig. 2 increases with E^2 . This effect remains valid as long as the fifth order effect does not exceed the third order effect in this frequency regime.

The broad conductivity peak in Fig. 2 is due to the resonant absorption of field in the third order process. Since our model is based on the Boltzmann transport equation under relaxation time approximation, the peak position is only dependent on the relaxation time but is independent of temperature and the chemical potential. However, in real systems, the peak position will be dependent on both temperature and the chemical potential due to that the scattering is dependent on both the temperature and carrier density (or chemical potential).^{22,23} High temperature will lead to a shorter

electron-phonon scattering time. In turn, the peak position will move towards high frequency. To the first approximation, the dependence of scattering on the chemical potential is through the screening effect. In this case, higher chemical potential leads to a weak scattering. As a result, the peak in the conductivity moves towards the low frequency.

Negative conductivity is a reflection of the amplification of the electromagnetic field, while positive conductivity is related to the absorption. The sign changes of the third order nonlinear response suggests that by modifying the band structure or gating the chemical potential, the HgTe/CdTe QW can be designed as the gain medium for terahertz radiation. It has been experimentally demonstrated that hole states can co-exist in valence band and conduction band with the “well” widths larger than 6.3 nm, forming a so called 2D electron-hole system, which indicates no spatial separation between the 2D electrons and 2D holes.²⁴ As a result, the self-consistent oscillations of electron and hole densities could occur under an appropriate external electric field, and gives rise to the terahertz emission.^{25,26}

Fig. 3 presents the nonlinear response as a function of chemical potential corresponding to the peak frequencies in Fig. 2. For the type I band structure in Fig. 3(a), the ratio decreases with increasing chemical potential, which is ascribed to the reduced vector phase at higher energy levels,

FIG. 3. $\sigma^{(3)}/\sigma^{(1)}$ as a function of chemical potential for (a) type I and (b) type II band structure with frequency 0.65 THz chosen from the peaks in Fig. 2.

while in the type II band structure in Fig. 3(b), the nonlinear response is divided into two regimes regarding of the position of μ . The ratio dramatically increases in the regime where μ lies below energy E_B , after which the ratio turns sharply to positive before slowly decreasing with the chemical potential. It is clear that at low temperature and small chemical potential, the third order conductivity dominates the first order conductivity.

The thermal effect on the nonlinear response can be seen in Fig. 3. The thermal effect causes a significant suppression of the third order conductivity. At low temperature, only a small number of electrons are thermally excited in the conduction band, and thus, the weak thermal excitation does not destroy too many conduction band electrons, which is of benefit for the third order nonlinear conductivity. At higher temperature, the linear response increases rapidly, and other scattering processes, such as electron-phonon scattering, are involved due to the strong thermal excitation. Therefore, the ratio $\sigma^{(3)}/\sigma^{(1)}$ decreases dramatically. Finally, we note that our result is based a sample of 7 nm thickness. While our result does not explicitly depend on the sample thickness, changing thickness can lead to a different band structure; thus, the conductivity can change. The calculated conductivity remains valid under the change of thickness as long as the band structure is not qualitatively altered. The threshold thickness at which the required band structure becomes invalid can be determined through a first principle band structure calculation.

In conclusion, we calculated the third order $\sigma^{(3)}$ in two types of bulk states of HgTe/CdTe QWs. The third order conductivity dominates in the low temperature regime under an appropriate electric field. In the type II band structure, the negative conductivity is observed when μ is located below E_A , which is ascribed to hole excitations from the states near E_B . The negative conductivity is a reflection of the amplification of the electromagnetic field. Our studies suggest a potential application of HgTe/CdTe in nonlinear photonics and optoelectronics devices in both light emission and amplification.

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- ¹Zh. I. Alferov, *Semiconductors* **32**, 1 (1998).
- ²K. Ortner, X. C. Zhang, A. Pfeuffer-Jeschke, C. R. Becker, G. Landwehr, and L. W. Molenkamp, *Phys. Rev. B* **66**, 075322 (2002).
- ³Y. R. Lin-Liu and L. J. Sham, *Phys. Rev. B* **32**, 5561 (1985).
- ⁴Y.-C. Chang, J. N. Schulman, G. Bastard, Y. Guldner, and M. Voos, *Phys. Rev. B* **31**, 2557 (1985).
- ⁵B. A. Bernevig, T. L. Hughes, and S. C. Zhang, *Science* **314**, 1757 (2006).
- ⁶M. König, S. Wiedmann, C. Brüne, A. Roth, H. Buhmann, L. W. Molenkamp, X. L. Qi, and S. C. Zhang, *Science* **318**, 766 (2007).
- ⁷G. M. Gusev, E. B. Olshanetsky, Z. D. Kvon, N. N. Mikhailov, S. A. Dvoretzky, and J. C. Portal, *Phys. Rev. Lett.* **104**, 166401 (2010).
- ⁸K. C. Nowack, E. M. Spanton, M. Baeninger, M. König, J. R. Kirtley, B. Kalisky, C. Ames, P. Leubner, C. Brüne, H. Buhmann, L. W. Molenkamp, D. Goldhaber-Gordon, and K. A. Moler, *Nat. Mater.* **12**, 787 (2013).
- ⁹M. Schultz, F. Heinrichs, U. Merkt, T. Colin, T. Skauli, and S. Løvold, *Semicond. Sci. Technol.* **11**, 1168 (1996).
- ¹⁰G. M. Minkov, A. V. Germanenko, O. E. Rut, A. A. Sherstobitov, S. A. Dvoretzky, and N. N. Mikhailov, *Phys. Rev. B* **88**, 045323 (2013).
- ¹¹M. König, H. Buhmann, L. W. Molenkamp, T. Hughes, C.-X. Liu, X. L. Qi, and S. C. Zhang, *J. Phys. Soc. Jpn.* **77**, 031007 (2008).
- ¹²Q. F. Sun, Y. X. Li, W. Long, and J. Wang, *Phys. Rev. B* **83**, 115315 (2011).
- ¹³X. L. Qi and S. C. Zhang, *Phys. Today* **63**(1), 33 (2010).
- ¹⁴X. C. Zhang, A. Pfeuffer-Jeschke, K. Ortner, V. Hock, H. Buhmann, C. R. Becker, and G. Landwehr, *Phys. Rev. B* **63**, 245305 (2001).
- ¹⁵S. Dvoretzky, N. Mikhailov, Y. Sidorov, V. Shvets, S. Danilov, B. Wittman, and S. Ganichev, *J. Electron. Mater.* **39**, 918 (2010).
- ¹⁶J. N. Schulman and T. C. McGill, *Appl. Phys. Lett.* **34**, 663 (1979).
- ¹⁷E. R. Youngdale, C. A. Hoffman, J. R. Meyer, F. J. Bartoli, M. A. Engelhardt, and H. Höchst, *Semicond. Sci. Technol.* **5**, S253 (1990).
- ¹⁸B. Zhou, H. Z. Lu, R. L. Chu, S. Q. Shen, and Q. Niu, *Phys. Rev. Lett.* **101**, 246807 (2008).
- ¹⁹E. G. Novik, P. Recher, E. M. Hankiewicz, and B. Trauzettel, *Phys. Rev. B* **81**, 241303(R) (2010).
- ²⁰J. Maciejko, X.-L. Qi, and S.-C. Zhang, *Phys. Rev. B* **82**, 155310 (2010).
- ²¹M. von Truchsess, V. Latussek, C. R. Becker, and E. Batke, *J. Cryst. Growth* **159**, 1128 (1996).
- ²²N. Tzoar and C. Zhang, *Phys. Rev. B* **32**, 1146–1151 (1985).
- ²³C. Zhang and Y. Takahashi, *J. Phys.: Condens. Matter* **5**, 5009 (1993).
- ²⁴Z. D. Kvon, E. B. Olshanetsky, D. A. Kozlov, N. N. Mikhailov, and S. A. Dvoretzky, *JETP Lett.* **87**, 502 (2008).
- ²⁵M. Ryzhii and V. Ryzhii, *Jpn. J. Appl. Phys., Part 2* **46**, L151 (2007).
- ²⁶X. G. Xu, S. Sultan, C. Zhang, and J. C. Cao, *Appl. Phys. Lett.* **97**, 011907 (2010).