Red-shift law of intense laser-induced electro-absorption in solids*

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A theoretical study on the red-shift of laser-induced electro-absorption is presented. It is found that laser-induced red-shift scales with the cube root of the pump laser intensity in the optical tunneling regime and has an obvious deviation from this scale in the multi-photon regime. Our results show that in the optical tunneling regime, the laser-induced red shift has the same law as that in the direct current (DC) approximation. Though the scales are the same in the optical tunneling regime, the physical pictures in the two cases are quite different. The electro-absorption in the DC case is a tunneling-assisted transition process, while the laser-induced electro-absorption is a mixed multi-photon process.

Keywords: laser-induced electro-absorption, red shift of absorption edge in solids, multi-photon process, pump–probe technology

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1. Introduction

The interaction between solids and intense lasers can cause dramatic modification of the energy band of the solid including band gap oscillation, dynamic location, and the appearance of new gaps.[1,2] Many interesting phenomena have been observed such as laser-induced transparency,[2] harmonic generation,[3,4] sideband generation,[5] and ultrafast electro-absorption.[6,7] The electro-absorption of near band-gap radiation in the presence of a strong pump laser has been investigated both theoretically[8,9] and experimentally.[2,6] Previous work focused on the multi-photon regime and dynamic regime. The intensity of the pump laser is up to the case $U_{\mathrm{P}}^{*} \sim \hbar\omega_0$,[2] where U_{P}^{*} is the ponderomotive potential of the electrons, and $\hbar\omega_0$ the photon energy of the pump laser. Due to the laser-induced damage to sample, relatively speaking, the optical tunneling regime remains unexplored. Recently, Ghimire *et al.*[7] made an experimental study about this new regime, and the phenomena of high order harmonics and substantial red-shift of absorption edges has been observed. Though the red-shift of absorption edges, which is induced by electrostatic field, is known very well (usual Franz–Keldysh effect), the theoretical study about laser-induced red-shift of absorption edges has not been performed before. In this work, we present a theoretical study about laser-induced red-shift from the multi-photon regime to the optical tunneling regime.

2. Theory and model

The setup of the laser-induced electro-absorption is illustrated in Fig. 1. In Fig. 1, the pump laser is used to modulate the transmission or absorption spectrum of the probe laser. In the presence of the pump laser, the optical absorption process of the probe laser can happen even though the photon energy of the probe laser is smaller than the band gap of the solid and the absorption increases with the intensity of the pump laser increasing. This effect can be interpreted as a shift of the absorption edge toward lower photon energies (red-shift of the absorption edge).

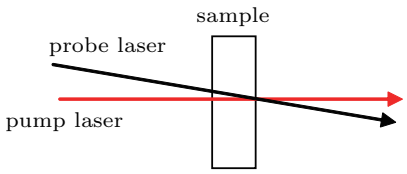


Fig. 1. (color online) Setup of laser-induced electro-absorption.

An intense laser is usually used as the pump laser and the Keldysh parameter $\gamma = \sqrt{E_{\mathrm{g}}/4U_{\mathrm{P}}^{*}}$ is employed to distinguish the different regimes of the pump laser,[2,10] where E_{g} is the band gap of the solids and $U_{\mathrm{P}}^{*} = e^2A_0^2/4m_{\mathrm{r}}$ is the ponderomotive potential, with $m_{\mathrm{r}} = 1/m_{\mathrm{c}} - 1/m_{\mathrm{v}}$ being the reduced mass of electron and hole, and $\boldsymbol{A}_0$ the vector potential of the pump laser. In the case of $\gamma \gg 1$, the intensity of the pump laser is in the multi-photon regime. In the case of $\gamma \sim 1$, the intensity

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is in the dynamic regime. Finally, in the case of $\gamma \ll 1$, it is in the tunneling regime.

For the intense laser, it is necessary to treat the interaction between laser and electron as being non-perturbative.[11–15] The Volkov wave is usually employed to describe the wave function of a free electron in an electromagnetic field. In solids, the Bloch wave function of an electron, accelerated by intense pump laser inside each of the bands can be written in the analogous form of the Volkov wave as[11]

$$\begin{aligned}\psi_{\rm C}(\boldsymbol{P}) &= \phi_{\rm C}(r)\exp\left\{-\frac{\rm i}{\hbar}\left[E_{\rm g}t+\frac{P^2}{2m_{\rm C}^*}t+\frac{e\boldsymbol{A}_0\cdot\boldsymbol{P}\sin(\omega_0 t)}{\omega_0 m_{\rm C}^*}\right.\right.\\ &\left.\left.+\frac{e^2A_0^2}{8m_{\rm C}^*\omega_0}\sin(2\omega_0 t)+\frac{e^2A_0^2 t}{4m_{\rm C}^*}\right]+\frac{\rm i}{\hbar}\boldsymbol{P}\cdot\boldsymbol{r}\right\},\\ \psi_{\rm V}(\boldsymbol{P}) &= \phi_{\rm V}(r)\exp\left\{-\frac{\rm i}{\hbar}\left[\frac{P^2}{2m_{\rm V}^*}t+\frac{e\boldsymbol{A}_0\cdot\boldsymbol{P}\sin(\omega_0 t)}{\omega_0 m_{\rm V}^*}\right.\right.\\ &\left.\left.+\frac{e^2A_0^2}{8m_{\rm V}^*\omega_0}\sin(2\omega_0 t)+\frac{e^2A_0^2 t}{4m_{\rm V}^*}\right]+\frac{\rm i}{\hbar}\boldsymbol{P}\cdot\boldsymbol{r}\right\},\end{aligned}\tag{1}$$

where $\psi_{\rm C}$ and $\psi_{\rm V}$ are the wave functions of the conduction band and valance band, respectively; $\boldsymbol{P}$ and $\boldsymbol{A}_0$ are the quasi-momentum of electron and the laser vector potential of the pump laser, respectively; $m_{\rm C}^*$ and $m_{\rm V}^*(<0)$ are the effective mass of electron in the conduction band and valence band, respectively; $\phi_{\rm C}(r)$ and $\phi_{\rm V}(r)$ are the functions that both have the translational symmetry of the lattice; $E_{\rm g}$ is the band gap of the solid and

$$\frac{P^2}{2m^*}=\frac{P_x^2+P_y^2+P_z^2}{2m^*}$$

is the the kinetic energy of the electron.

The transition probability amplitude $C_{\rm f}$ from valance band to conduction band, caused by the probe laser, is in the form of

$$\begin{aligned}C_{\rm f} &= \frac{1}{{\rm i}\hbar}\int_0^t\int\psi_{\rm C}H_{\rm el}\psi_{\rm V}\,{\rm d}\boldsymbol{r}\,{\rm d}t'\\ &= \frac{1}{{\rm i}\hbar}\int_0^t{\rm d}t'H_{\rm fi}\exp\left\{\frac{\rm i}{\hbar}\left(\frac{P^2}{2m_{\rm C}^*}-\frac{P^2}{2m_{\rm V}^*}+E_{\rm g}+U_{\rm P}^*-\hbar\omega\right)t'\right.\\ &\left.+\frac{\rm i}{\hbar}\frac{e\boldsymbol{A}_0\cdot\boldsymbol{P}\sin(\omega_0 t')}{\omega_0}\frac{1}{m_{\rm r}}+\frac{\rm i}{\hbar}\frac{e^2A_0^2}{8m_{\rm r}\omega_0}\sin(2\omega_0 t')\right\},\end{aligned}\tag{2}$$

where $H_{\rm el}=\boldsymbol{A}\cos(\omega t)\cdot\boldsymbol{P}/m_{\rm e}$ is the Hamiltonian of electron–probe laser interaction, with $m_{\rm e}$ being the mass of free electron, and $H_{\rm fi}=\int\psi_{\rm c}^*H_{\rm el}\psi_{\rm V}|_{t=0}\,{\rm d}\boldsymbol{r}$. In a way similar to that in Refs. [10]– [12], we now expand the term on the left-hand side in Eq. (2)

$$\begin{aligned}L(t') &= H_{\rm fi}\exp\left\{\frac{\rm i}{\hbar}\left(\frac{P^2}{2m_{\rm C}^*}-\frac{P^2}{2m_{\rm V}^*}+E_{\rm g}+U_{\rm P}^*-\hbar\omega\right)t'\right.\\ &+\frac{\rm i}{\hbar}\frac{e\boldsymbol{A}_0\cdot\boldsymbol{P}\sin(\omega_0 t')}{\omega_0}\frac{1}{m_{\rm r}}\\ &\left.+\frac{\rm i}{\hbar}\frac{e^2A_0^2}{8m_{\rm r}\omega_0}\sin(2\omega_0 t')\right\}\end{aligned}\tag{3}$$

into a Fourier series in t' as given below

$$\begin{aligned}L(t') &= \sum_{n=-\infty}^{\infty}\exp\left\{\frac{\rm i}{\hbar}\left(\frac{P^2}{2m_{\rm C}^*}-\frac{P^2}{2m_{\rm V}^*}+E_{\rm g}+U_{\rm P}^*-\hbar\omega\right)t'\right\}\\ &\times\exp(-{\rm i}n\omega_0 t')L_n,\end{aligned}\tag{4}$$

where

$$\begin{aligned}L_n &= H_{\rm fi}\frac{1}{2\pi}\int_{-\pi}^{\pi}\exp\left[{\rm i}\frac{e\boldsymbol{A}_0\cdot\boldsymbol{P}}{\hbar\omega_0 m_{\rm r}}\sin x+{\rm i}\frac{e^2A_0^2}{8\hbar m_{\rm r}\omega_0}\sin 2x\right]\\ &\times\exp({\rm i}nx)\,{\rm d}x\\ &= H_{\rm fi}{\rm J}_{-n}\left(\frac{e\boldsymbol{A}_0\cdot\boldsymbol{k}}{\omega_0 m_{\rm r}},\frac{e^2A_0^2}{8\hbar m_{\rm r}\omega_0}\right),\end{aligned}$$

with $\hbar\boldsymbol{k}=\boldsymbol{P}$, ${\rm J}_n(u,\upsilon)=\sum_{k=-\infty}^{\infty}{\rm J}_{n-2k}(u){\rm J}_k(\upsilon)$ being the generalized Bessel function which can be described by the sum of first-kind Bessel function, $x=\omega t'$. Substituting Eq. (4) into Eq. (2) and then using the same calculation procedure as that in the usual perturbation theory,[16] the transition rate for the absorption of the probe laser is in the form (the spin state of electron is considered):

$$\begin{aligned}W &= \sum_n\frac{2\pi}{\hbar}\int|L_n|^2\,\delta\left(\frac{P'^2}{2m_{\rm C}^*}+\frac{P^2}{2m_{\rm v}}+E_{\rm g}\right.\\ &\left.+U_{\rm P}^*-n\hbar\omega_0-\hbar\omega\right)\frac{2}{(2\pi)^3}\,{\rm d}\boldsymbol{k}.\end{aligned}\tag{5}$$

From electrodynamics,[17] the absorption coefficient of the probe laser, which is caused by this band–band transition, can be written as

$$\alpha=\frac{\omega}{n_{\rm r}c\omega\varepsilon_0}\frac{2W\hbar\omega}{E^2}=\frac{2W\hbar\omega}{n_{\rm r}c\varepsilon_0E^2},\tag{6}$$

where ε_0 is the permittivity of a vacuum, c the light speed in a vacuum, $n_{\rm r}$ the refractivity of solids, and E the amplitude of the electrical field of the probe laser. We now discuss Eq. (5). In the case of no pump laser radiation, $\boldsymbol{A}_0=0$, we have ${\rm J}_{-n}(0,0)=0$ $(n\neq 0)$, ${\rm J}_0(0,0)=1$. It can be found that equation (5) describes the usual band–band transition of the probe laser. In the presence of the pump laser,

$${\rm J}_{-n}\left(\frac{e\boldsymbol{A}_0\cdot\boldsymbol{k}}{\omega_0 m_{\rm r}},\frac{e^2A_0^2}{8\hbar m_{\rm r}\omega_0}\right)\neq 0,$$

the absorption process of probe light is modulated by the pump laser.

3. Results and discussion

The Monte Carlo method is employed to carry out the numerical calculation of Eq. (5), which is an effective approach for multiple integrals. In Fig. 2, the absorption coefficients of the probe laser at different wavelengths are presented. Here the pump laser intensity is 0.05 TW/cm^2; the wavelength of the pump laser is 3250 nm; the band gap of solid is 3.2 eV, and the reduced mass is $0.33m_0$ (the numerical parameters are from ZnO). To make a comparison, the case of the no pump

laser is also presented in Fig. 2. From Fig. 2, it can be found that in the presence of the pump laser, the absorption process can happen even though the photon energy of the probe laser is smaller than the band gap. This effect is called laser-induced electro-absorption. Similar to the case of electrostatic field-induced electro-absorption, laser-induced electro-absorption can be interpreted as a shift of absorption edges of solids toward lower photon energies (red-shift of absorption edge).

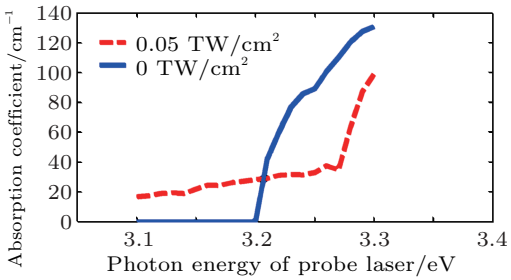


Fig. 2. (color online) Absorption coefficients of the probe laser at pump intensity of 0 TW/cm^2 and 0.05 TW/cm^2.

The coefficients of the probe laser at different pump intensities are shown in Fig. 3, where the wavelength of the pump laser is 3250 nm. Figure 3(a) shows the coefficients of the probe laser at intensities of 0.0001 TW/cm^2, 0.01 TW/cm^2, 0.05 TW/cm^2, and 0.1 TW/cm^2 (the corresponding Keldysh parameters γ are 72, 7.2, 3.2, and 2.7, respectively); figure 3(b) shows the coefficients at intensities of 0.1 TW/cm^2, 1 TW/cm^2, 5 TW/cm^2, and 10 TW/cm^2, (the values of corresponding Keldysh parameter γ are 2.7, 0.72, 0.32, and 0.23, respectively).

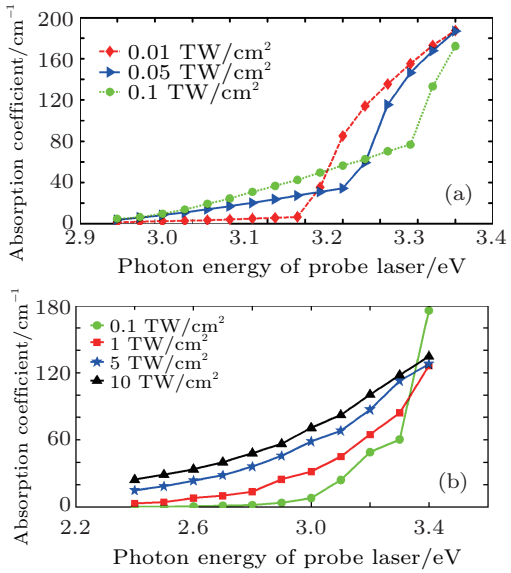


Fig. 3. (color online) Absorption coefficients of the probe laser at different pump intensities.

If we take the red-shift of absorption edge at pump laser intensity I_s as $\Delta E_g(I_s)$, the red shift $\Delta E_g(I_0)$ at intensity I_0 can be calculated by

$$\frac{\alpha(E_g-\Delta E_g(I_0),I_0)}{\alpha(E_g,I_0)}=\frac{\alpha(E_g-\Delta E_g(I_s),I_s)}{\alpha(E_g,I_s)}, \tag{7}$$

where $\alpha(E_g-\Delta E_g(I_0),I_0)$ is the absorption coefficient of the probe laser at photon energy $E_g-\Delta E_g(I_0)$, with I_0 being the intensity of the pump laser; $\alpha(E_g,I_0)$ is the absorption coefficient of the probe laser at photon energy E_g; I_0 is the intensity of the pump laser; $\alpha(E_g-\Delta E_g(I_s),I_s)$ is the absorption coefficient of the probe laser at photon energy $E_g-\Delta E_g(I_s)$; I_s is the pump laser intensity; $\alpha(E_g,I_s)$ is the absorption coefficient of the probe laser at photon energy E_g; I_s is the pump laser intensity. $\Delta E_g(I_s)$ can be determined in experiment.

In Fig. 4, the laser-induced red-shifts at different intensities are presented, where $I_s=1$ TW/cm^2 and $\Delta E_g(I_s)=0.38$ eV (this value is close to the experimental result of ZnO[7]). The red-shift from direct current (DC) approximation,[7,18] in which the laser is regarded as an electrostatic field, is also presented (dashed line). In the presence of an electrostatic field (usual Franz–Keldysh effect), the transition rate of electron is in the form of[7,18]

$$W=C\exp\left\{-\frac{4\sqrt{2m^*}}{3e\hbar E_0}(E_g-\hbar\omega)^{3/2}\right\},\quad \hbar\omega<E_g, \tag{8}$$

where the pre-factor $C(E_0,\omega)$ is a relatively slowly varying function of E_0 and ω compared with the exponential part in the above formula, with E_0 being the field amplitude of the pump laser. Using the same definition as that in Eq. (7), we have

$$\begin{aligned}&\exp\left\{-\frac{4\sqrt{2m^*}}{3e\hbar E_0}\Delta E_g(I_0)^{3/2}\right\}\\ &=\exp\left\{-\frac{4\sqrt{2m^*}}{3e\hbar E_s}\Delta E_g(I_s)^{3/2}\right\}\\ &=C_0,\end{aligned} \tag{9}$$

where C_0 is a constant and E_s the field amplitude of the pump laser with intensity I_s. Thus, the electrostatic field induced red-shift ΔE_g can be described by the law $\Delta E_g\propto E_0^{2/3}\propto I_0^{1/3}$.[7,18]

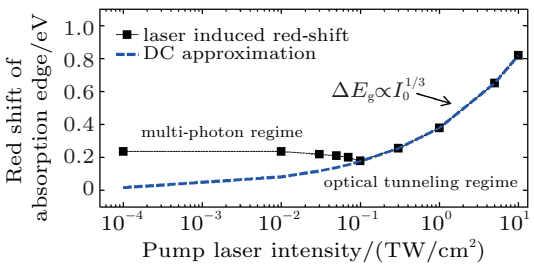


Fig. 4. (color online) Red-shifts of absorption edge at different intensities.

From Fig. 4, it can be found that laser-induced red-shift (solid squares) scales with the cube root of the pump laser intensity in optical tunneling regime and has an obvious deviation from this scale in the multi-photon regime. It can also be

found that in the optical tunneling regime, the laser-induced red shift follows the same law as that in the direct current (DC) approximation. Though it is inconsistent with the usual expectation that in the strong field limit the laser can be considered as an electrostatic field, the physical pictures of the two cases are quite different.

In the presence of an electrostatic field, the energy bands of electrons will be tilted due to the additional potential energy which is caused by the electrostatic field. The electron in a valence band can transit to a conduction band by the tunneling effect even though the photon energy of the probe laser is smaller than the band gap of solid, where the width of the electrostatic field-induced barrier is $d = (E_g - \hbar\omega_0)/eE_0$. For the case of laser-induced electro-absorption, the pump laser photon takes part in the transition process of electron. With the help of the pump photon, the transition process can happen even though the photon energy of the probe laser is lower than the band gap of solids (mixed multi-photon process). The band–band transition process of laser-induced electro-absorption and DC approximation are illustrated in Fig. 5(a) and 5(b), respectively.

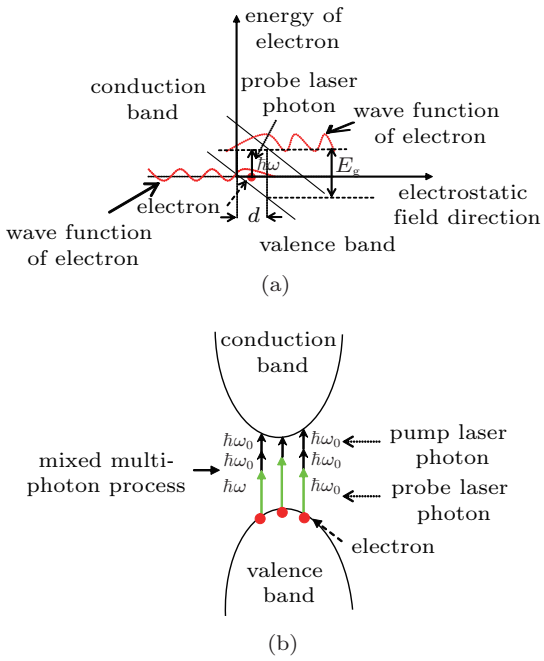


Fig. 5. (color online) Band–band transition process of electron for (a) DC approximation (electrostatic field-induced electro-absorption), and (b) laser-induced electro-absorption.

It should be noted that if the intensity of the pump laser is extremely high, e.g., the laser electric field becomes comparable to the ratio of band gap to the lattice space,[7] the Bragg scattering from the Brillouin zone boundaries and non-parabolic band structure effect should be considered. The deviation from the cube root scale in the case of extremely high intensity has been observed.[7] The influences of Bragg scattering and non-parabolic structures on laser-induced electro-absorption will be considered in the further work.

4. Summary

A theoretical study on the red-shift of laser-induced electro-absorption from the multi-photon regime to the optical tunneling regime is presented. It is found that the red-shift scales with the cube root of laser intensity in optical tunneling regime and has an obvious deviation from this scale in the multi-photon regime. Our work shows that in the optical tunneling regime, the laser-induced red-shift has the same law as that in the DC approximation. The different physical pictures of the two cases are discussed.

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