Exciton absorption in semiconductor superlattices in a strong longitudinal THz field

Jie-Yun Yan\textsuperscript{1}, Ren-Bao Liu\textsuperscript{2} and Bang-fen Zhu\textsuperscript{1,3,4}  
\textsuperscript{1} Department of Physics, Tsinghua University, Beijing 100084, People’s Republic of China  
\textsuperscript{2} Department of Physics, The Chinese University of Hong Kong, Hong Kong, People’s Republic of China  
\textsuperscript{3} Center for Advanced Study, Tsinghua University, Beijing 100084, People’s Republic of China  
E-mail: bfz@tsinghua.edu.cn

Abstract. We theoretically study the excitonic optical absorption in semiconductor superlattices irradiated by an intense terahertz (THz) laser polarized along the growth direction. We calculate the linear excitonic absorption spectra using the finite-difference time-domain method in real space. Our numerical results and qualitative analysis show that when tuning the frequency or intensity of the THz laser, the dynamical localization (DL) and the ac Stark effect (ACSE) are either jointly or individually responsible for the exciton peak shift. Moreover, with the onset of miniband collapse, the excitonic dimensionality crossover and the DL will counteract each other in shifting the exciton peak. The direct experimental verification of the DL through semiconductor optical spectra is still a challenge. Regarding this fact, we propose a method to single out the DL effect from the spectral shift dominated by the ACSE.

\textsuperscript{4} Author to whom any correspondence should be addressed.
1. Introduction

The typical energy scales in semiconductor superlattices (SSLs) or multiple quantum wells (MQWs), such as the miniband width, exciton level spacings, optical phonon energy, Bloch oscillation frequency, etc, are in the terahertz (THz) region. When an SSL is irradiated with radiation from an intense THz laser and probed by a weak near-bandgap laser, a wealth of physical phenomena may be observed, including the dynamical Franz–Keldysh effect (DFKE) [1], the ac Stark effect (ACSE) [2, 3], the dynamical localization (DL) and delocalization [4, 5], the inverse Bloch oscillations [6]–[10], the dynamical Fano effect [11], the exciton stabilization [12] and the high-order THz-sideband generation [13, 14].

In general, strong THz radiation can induce more than one of the effects mentioned above. For example, in a semiconductor MQW driven by an intense THz field polarized in the MQW plane, the excitonic absorption experiments display the interplay of the DFKE and the ACSE [15]. Similarly, in an SSL irradiated with radiation from an intense THz laser polarized along the SSL growth direction, we predict that DL and ACSE will compete and interact with each other in excitonic spectra when changing the THz field intensity or frequency.

The DL refers to the effect [4, 5] that in the presence of a strong ac field $F_{ac} \cos(\omega t)$, an electron in an SSL may return to its initial state periodically if $J_0(eF_{ac}d/\hbar\omega)=0$, where $e$ is the fundamental charge, $d$ is the length of an SSL period and $J_0$ represents the zero-order Bessel function of the first kind. It was revealed that with variation of the field strength, the quasi-miniband width of the SSL would vary and even collapse when the DL occurs [16, 17]. When the Coulomb interaction is taken into account, the exciton discrete levels and miniband continua are formed. Interestingly, except for the collapse of miniband in the condition mentioned above, the discrete excitonic states would also shift accordingly. The DL in the presence of the Coulomb interaction has already been theoretically studied in the linear absorption, THz emission and four-wave-mixing [8, 18, 19].

The DL in SSLs, however, has not yet been confirmed experimentally in the semiconductor optical spectroscopy, although a similar effect was observed in optical lattices a decade ago [20]. Recently, Koch et al measured the excitonic absorption spectra in an SSL irradiated with radiation from the UCSB free electron laser (FEL) polarized in the SSL growth direction [21]. When tuning the frequency of the FEL larger than the exciton binding energy of the SSL, they observed a blueshift of the absorption peak associated with the heavy-hole exciton, and attributed this blueshift to the onset of the DL. Such an interpretation, however, is complicated with the ACSE, especially in the parameter regime where $R \equiv eF_{ac}d/\hbar\omega$ is still much smaller than the first root of the Bessel function as in the experiment.
The ACSE in the SSLs arises from the coupling induced by the THz field between the exciton ground state and the excited states with opposite parities. The ACSE leads to energy shift of the exciton levels, which depends sensitively on the ac field strength and frequency \([3, 22]\). Recently, Carter et al. reported the near-infrared (NIR) absorption in MQWs irradiated with radiation from the UCSB FEL polarized along the MQW growth direction with frequency tuned into resonance with the transition between the h1X (the exciton ground state of the first heavy-hole subband) and the h2X (the exciton ground state of the second heavy-hole subband) \([23]\). By comparing with the numerical calculations, they attributed the measured spectral shift to the ACSE. In addition to such observed inter-subband ACSE, the strong THz field along the SSL growth direction (taken as the \(z\)-axis hereafter) should also result in the intra-subband ACSE associated with the coupling between the h1X and its excited state of opposite parity, e.g. \(2\)\(P_z\). This is similar to the ACSE due to the exciton \(1S\)–\(2P\) coupling observed in quantum wells \([15, 24, 25]\) except for the different polarization of the THz field.

So the SSL in the presence of a strong THz laser, in which the DL and ACSE coexist, provides a valuable platform to investigate the interplay of these two effects. Tuning the THz field frequency or strength may influence the DL and ACSE simultaneously. To understand the physical mechanisms behind an experimentally observed excitonic spectrum of an SSL in a strong longitudinal THz field, one needs to identify the role each effect plays and compare the experimental data with numerical results carefully. Thus a complete theory is needed, which should take at least these two effects into account.

In this paper, we intend to understand and evaluate the exciton absorption in SSL under an intense longitudinal THz field, considering both the DL and the ACSE. We will analyze the role played by each effect in exciton absorption spectra and numerically calculate the linear absorption by a real-space finite-difference time-domain method \([12, 26]\), in which these two effects are taken into account on an equal footing. The paper is organized as follows. In section 2, we shall give the qualitative analysis for each effect as well as their mutual interaction. After introducing the theoretical formalism in section 3, we present our numerical results in section 4, and the conclusions are drawn in section 5.

2. Analysis

Before carrying out the numerical calculations, we analyze the ACSE and DL effects qualitatively in this section.

We first consider the ACSE due to the coupling between the 1\(S\) and 2\(P_z\) exciton states by a strong THz field with frequency \(\omega\). Around the \(E_{1S}\) level, two split dressed levels of \(E_L\) and \(E_H\) would be born with energies \([3]\)

\[
E_H = E_{1S} + \frac{1}{2} \left\{ \left( \Delta E \right)^2 + \left( \hbar \omega_R \right)^2 \right\}^{1/2} - \Delta E,
\]

\[
E_L = E_{1S} - \frac{1}{2} \left\{ \left( \Delta E \right)^2 + \left( \hbar \omega_R \right)^2 \right\}^{1/2} + \Delta E,
\]

where the detuning \(\Delta E\) is defined as

\[
\Delta E \equiv \hbar \omega - (E_{2P_z} - E_{1S}) \equiv \hbar \omega - E_{21},
\]

and \(\omega_R\) is the Rabi frequency, the product of the THz field amplitude and the dipole matrix element between the 1\(S\) and 2\(P_z\) states. Usually, for a finite detuning, only one excitonic absorption peak whose component is mainly of the 1\(S\) exciton state can be clearly observed.
(called the bright optical transition) [2], whereas the other peak, mainly of one THz-photon sideband of the 2Pz exciton, is hardly observed experimentally. As shown by equation (2), when increasing ω in the negative detuning regime (ΔE < 0), the bright optical transition, the one with energy EL, shifts toward lower energy. When increasing ω in the positive detuning regime (ΔE > 0), the absorption at EH is the bright one and exhibits a redshift. When the THz frequency ω runs across the resonant point, there will appear an anticrossing of the absorption peak from the EL to the EH associated with a gap of the Rabi frequency. If the dephasing rate is small enough, two optical absorption peaks of the same strength may be discernible when ΔE ≈ 0 [23]. Such a split-peak structure at the anticrossing is one of the characteristics of the ACSE [27]. When this fingerprint appears, the existence of ACSE is clearly identified. Away from the anticrossing regime, increasing the field strength with the THz field frequency fixed will bring about a redshift for negative detuning and a blueshift for positive detuning.

The DL causes the excitonic continuous miniband to collapse and the exciton discrete level to shift, which critically depend on R, the ratio of the THz field strength to its frequency. When increasing R within the region less than 2.405, the first root of J0, the DL would generally lead to a blueshift of the 1S exciton peak. It should be noted here that the DL implies the transformation of the exciton state from three-dimensional (3D) to 2D [28]. As the Coulomb binding energy of the 2D exciton is much enhanced compared with the 3D exciton, the blueshift of the continuum edge is expected to be much larger than that of the 1S bound state of the exciton with R increasing.

In general, when tuning the frequency or strength of the THz field, both the ACSE and the DL will shift the exciton absorption peak. In the positive detuning, both the DL and the ACSE result in a blueshift with increasing the THz field strength, so it is hard to attribute the blueshift of the exciton absorption peak to either effect exclusively. Only in the negative tuning will the DL and the ACSE result in opposite exciton energy shifts with changing the THz field strength. So if a blueshift, by increasing the field strength, is observed in the negative detuning regime, the onset of the DL can be unambiguously identified, which even overcomes the redshift due to the ACSE. To realize this, the ac field strength in the sample should be adjustable, measurable and in the range from several kV cm−1 to tens of kV cm−1, which is still a challenge experimentally.

Moreover, the ACSE and DL affect each other, although the former dominates around the resonance at ΔE = 0, while the latter dominates when R approaches the root of J0(R). In the following, we will discuss the interplay of the ACSE and DL, and show how to single out the DL effect from the spectral shift near the ACSE resonance.

The DL shifts the spectral positions of the 1S and 2Pz states at different rates. On the one hand, the spectral shift of the 1S exciton due to the DL will not simply follow the shift of the continuum edge because of its binding enhancement due to the 3D–2D dimensionality crossover associated with the DL; on the other hand, the 2Pz state of the exciton, which is loosely bound and behaves more like a continuum state, will nearly follow the continuum edge. Thus the energy separation between the 1S excitation peak. It should be noted here that the DL implies the transformation of the exciton state from three-dimensional (3D) to 2D [28]. As the Coulomb binding energy of the 2D exciton is much enhanced compared with the 3D exciton, the blueshift of the continuum edge is expected to be much larger than that of the 1S bound state of the exciton with R increasing.

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To better analyze the spectral shift induced jointly by the ACSE and DL near the resonance at ℏω′ = E21(R0), we assume that E1S(R) and E2Pz(R) can be expanded in terms of R − R0, where R0 = εF′0d/ℏω′ and E21(R0) is the energy separation between 1S and 2Pz.
levels in resonance in the presence of the field $F_e$. After some algebra, we have

$$E_{1S}(R) = E_{1S}(R_0) + \alpha (J_0(R_0) - J_0(R)),$$

where $E_{1S}(R_0)$ is the 1S exciton spectral position in resonance, and the parameter $\alpha$ denotes the variation rate of the 1S position at $R_0$, which is again related to the variation rate of the difference between the miniband edge and the binding energy of the 1S exciton. Of course, such a linear variation assumption for $E_{1S}(R)$ works well only within a small region of $R$ centered at $R_0$, but $R_0$ is not necessarily a small value. We can also write down a similar expression for the 2$P_e$ level position with respect to the ratio $R$. Then the energy separation $E_{21}(R)$ can be expressed as

$$E_{21}(R) = E_{21}(R_0) + \beta [J_0(R_0) - J_0(R)],$$

where $\beta$ is a parameter describing the variation rate of the difference in binding energy between the 1S and 2$P_e$ exciton states due to the DL. As we shall show in section 4, the resonance frequency $\omega'$ can be identified from the numerical results when two split absorption peaks are of the same strength. The above assumption can be checked qualitatively by using the quasi-energy diagram of semiconductor superlattices in the presence of an intense THz field.

3. Theoretical formalism

To fully understand and evaluate the spectral shift of the SSL exciton in a strong longitudinal THz field, besides the qualitative picture mentioned above, we should carry out numerical calculations for the linear absorption spectra. The model and method are the same as in [12] and are briefly described below.

In the framework of the effective mass approximation, the Hamiltonian of the system under investigation is [6, 9, 13]

$$H_0 = -\frac{\hbar^2}{2m_e} \frac{\partial^2}{\partial r^2} + V_e(z_e) + V_h(z_h) + eFz_e - eFz_h + V_c(r_e - r_h),$$

where $m_e$ ($m_h$) denotes the effective mass of an electron (hole), $V_e$ ($V_h$) stands for the periodic superlattice potential of the electron (hole) along the $z$-direction, $V_c$ is the Coulomb potential between an electron and a hole and $F = F_{ac} \cos(\omega t)$ is the electric field of the THz laser polarized along the $z$-direction. When the SSL is excited by an NIR laser field $E(t)$, the system evolution is governed by the following equation:

$$i(h \partial_t + \gamma_2) \psi(r_e, r_h, t) = H_0 \psi(r_e, r_h, t) - d_{cv} E(t) \delta(r_e - r_h),$$

with the initial condition

$$\psi(r_e, r_h, -\infty) = 0,$$

where $d_{cv}$ is the interband dipole matrix element and $\gamma_2$ is the interband dephasing rate. For the sake of simplicity, in the following calculations we consider only one electron and one hole miniband in the tight binding formalism with the nearest-neighbor coupling considered, and neglect the center-of-mass motion of the electron–hole pair. Then, equation (7) becomes [12]

$$i(h \partial_t + \gamma_2) \psi(\rho, \ell, t) = -\frac{\hbar^2}{2\mu} \nabla^2 \psi(\rho, \ell, t) + [E_e + eF\ell d] \psi(\rho, \ell, t) - \frac{\Delta}{4} \psi(\rho, \ell + 1, t)$$

$$+ \psi(\rho, \ell - 1, t) - d_{cv} E(t) \frac{\delta_{\ell,0}\delta(\rho)}{2\pi \rho} - \frac{e^2}{4\pi \epsilon \epsilon_0 \sqrt{\rho^2 + (\ell d)^2}} \psi(\rho, \ell, t),$$

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where $\mu$ is the reduced mass of the electron–hole pair, $\rho$ is the relative coordinate of the electron and hole in the $x$–$y$ plane, $\ell$ labels the index of the Wannier state and $\Delta$ represents the joint electron–hole miniband width.

The transient interband polarization is

$$
P(t) = \frac{1}{V} \int d^2 \rho \, d_{e}^{*} \delta(\rho) \psi(\rho, \ell = 0, t),
$$

and the optical absorption spectrum is

$$
\alpha(\omega) \propto \mathcal{I} \left[ \tilde{P}(\omega) \tilde{E}(\omega) \right],
$$

where $\tilde{P}(\omega)$ and $\tilde{E}(\omega)$ are the Fourier transformations of $P(t)$ and $E(t)$, respectively.

We solve the equation with the finite-difference time-domain method in real space \cite{12, 26}, and obtain the optical absorption by fast Fourier transformation.

4. Numerical results

We calculate the excitonic absorption in a sample similarly to the experiment in \cite{21}. Namely, the SSL is made up of 40 periods of 3.8 nm GaAs wells sandwiched by 3.8 nm Al$_{0.3}$Ga$_{0.7}$As barriers. The energy gaps in the well and barrier materials are 1.517 and 1.897 eV, respectively. The conduction band offset between GaAs and Al$_{0.3}$Ga$_{0.7}$As is taken to be 240 meV. As the electron is mainly confined to the well, we neglect the effective mass difference between GaAs and Al$_{0.3}$Ga$_{0.7}$As, and take the GaAs effective mass as $0.067 m_0$ for the electron and $0.34 m_0$ for the heavy hole ($m_0$ is the free electron mass). The static dielectric constant $\epsilon$ is chosen as 12.5. By the Kronig–Penney model, we estimate the miniband width to be 35 and 2 meV for the lowest electron and the lowest heavy hole, respectively. So the combined miniband width is 37 meV. Thus the calculated binding energy of the $1S$ exciton is 8.8 meV and the spectral position $E_{1S}(0)$ is 1647.08 meV in the absence of THz field.

The linear excitonic absorption spectra in a strong THz field for four different frequencies are shown in figure 1(a). We set the dephasing rate $\gamma_2$ as 1 meV in the calculation. With such a big dephasing rate, the split peaks at the anticrossing point are not resolved. The absorption peak is associated with the bright exciton transition. A blueshift is observed for the two larger frequencies ($\omega = 10.4$ and 6.33 meV), whereas a redshift is observed for the two lower THz field frequencies. Such opposite spectral shifts for the THz frequency tuned higher or lower than the $1S$–$2P_z$ transition can be well understood with the ACSE. To compare with the experimental results in \cite{21}, we plot the negative difference absorption spectra in figure 1(b). Good agreement is found between our calculation and the experimental data.

To quantitatively confirm the existence of the ACSE, we set the dephasing rate to be a smaller value of 0.4 meV and calculate the absorption peak positions as functions of the THz frequency as plotted in figure 2(a). A clear $E_{1S} - E_{21}$ anticrossing appears in figure 2(b), which proves the existence of ACSE unambiguously. When $\Delta E = 0$, the two absorption peaks are of the same strength, whereby an accurate value of $E_{21}(R_0)$ can be obtained. In our case, $E_{21}(R_0)$ is deduced to be 8.32 meV, corresponding to $R_0 = 0.274$ for $F_c$ taken to be 3 kV cm$^{-1}$.

The joint effects of the DL and ACSE of a strong THz field on the absorption peak position of the $1S$ exciton can be analyzed by using equations (1)–(5), in which $E_{1S}$ and $E_{21}$ depend on the THz field strength and frequency through the DL ratio $R$. By fitting numerical results...
Figure 1. The calculated excitonic (a) linear and (b) negative differential absorption spectra in (GaAs)$_{38}\AA$($Al_{0.3}Ga_{0.7}As$)$_{38}\AA$ in the presence of a THz field of 3 kV cm$^{-1}$ with four frequencies at 2.72, 6.33, 10.4 and 14.0 meV. For clarity, a magnified view of the circled part in the figure is shown in the inset.

Figure 2. (a) The calculated absorption spectra of the 1S exciton as a function of THz frequencies, in which the dephasing rate is taken to be 0.4 meV and the THz field is fixed at 3 kV cm$^{-1}$. (b) The calculated spectral position of two split levels of the 1S exciton as functions of the THz frequency. The circles stand for the numerical results, while the solid lines are fitted by using equations (1)–(5).

with equations (1)–(5), we obtain two fitting parameters $\alpha$ and $\beta$ in the analytical formulae as 1 and 20 meV, respectively. As seen in figure 2(b), the analytical expressions with these two parameters agree with the numerical results excellently.

The fact that $\alpha$ is small and $\beta$ is about half the combined miniband width means that the 1S exciton position changes little with $R$, and the 2P$_z$ state nearly follows the continuum edge as the miniband shrinks due to the DL. This is more explicitly shown in figure 3, in which for a fixed field strength the numerically calculated $E_{1S}$ and 2P$_z$ are plotted versus the frequency $\omega$. The 1S excitonic position shifts little with $\omega$ except near the zero point of $J_0(R)$ (not shown in the figure), whereas the 2P$_z$ excitonic position varies more significantly. This trend can be understood from the fact that when the minibands shrink due to the DL, the binding energy of the ground-state exciton is enhanced because of the excitonic dimensionality crossover from 3D...
Figure 3. The excitonic spectral positions as functions of the THz frequency $\omega$ for a fixed strength of 3 kV cm$^{-1}$. The solid line stands for the miniband edge and the dashed (dotted) line represents the 1$S$ ($2P_z$) excitonic spectral position.

to 2D accompanying the DL, whereas the excited state of the exciton still sticks to the miniband edge to some extent. The effect of the excitonic dimensionality crossover on the ACSE is a special phenomenon in SSLs as compared with the cases in MQWs [23] and QDs [29]. In SSLs the 1$S$–$2P_z$ resonance frequency may be changed with the ac field strength, whereas in the latter two experiments the resonance frequencies are almost fixed.

In this way, we can simulate and analyze how the DL and ACSE affect each other. More specifically, we can estimate the effect of the DL on the ACSE around the $R_0$ where the anti-crossing occurs. For instance, with the fitting parameters appropriate to the experiment [21], we have calculated the excitonic absorption peaks as a function of the detuning with and without the DL correction, respectively, and found that for the parameters relevant to the experiment, the 1$S$ exciton shift attributable to the DL is negligibly small.

As we stated previously, for a fixed negative detuning, when increasing the field strength, the ACSE will make the excitonic peak redshift, whereas the DL will do the opposite. This is the only configuration that may be used to distinguish the DL from the ACSE unambiguously provided that the blueshift due to the DL overcomes the counteraction due to the 3D–2D crossover of the exciton by the field strength. Near the ACSE resonance, the ACSE splitting is a linear function of $R$ (if $\omega_R$ is small but larger than $|\Delta E|$), while the miniband width shrinking is a quadratic function of $R$, so the dominating effect is always the ACSE for a small $R$. At large detuning with lower THz frequency ($|\Delta E| > \omega_R$), the ACSE splitting is also a quadratic function of $R$. Figure 4 shows the redshift of the exciton absorption peaks in the field of $\omega = 3$ meV at several field strengths, which vary with $R^2$ approximately. Figure 4 also demonstrates that quite similarly to figure 3, the 1$S$ exciton peak shifts rather slightly with respect to the field strength. As for the complex structure of the absorption at $R = 0.9$, the THz-photon sideband effect may play a role. As the intensity of the THz field gets stronger, the higher-order THz-photon sidebands of the miniband become more and more important and overlap with each other, which will result in complicated variation of the exciton level versus the field strength. That means that,
Figure 4. The excitonic linear absorption in an SSL under intense THz radiation with the frequency fixed as 3 meV. The THz field strength is denoted by $R$ as shown in the figure, and $F = 0$ means the absence of the THz field. The dephasing rate $\gamma_2$ here is set to a small value of 0.01 meV in order to clearly observe the peak shift. The parameters for the SSL structure are described in the text.

In this situation, it becomes very difficult to verify the DL by solely observing the $1S$ exciton absorption.

5. Conclusion

In summary, we have theoretically investigated the influence of a strong longitudinal THz field on the excitonic absorption spectra in SSLs, with an emphasis on the exciton spectral shift by tuning the THz frequency and field strength. We point out that three effects, the ACSE, the DL effect and the exciton dimensionality crossover effect, may jointly or individually contribute to the excitonic peak shift in SSLs in a strong longitudinal THz field. The DL and the exciton dimensionality effect always counteract each other and are responsible for the insensitiveness of the $1S$ exciton position to the THz frequency and strength. The DL dominates when the ratio $R$ of the THz field strength over its frequency approaches the root of the Bessel function, while the ACSE dominates when the THz photon is nearly resonant with the $1S–2P_z$ level separation of the exciton. Tuning the THz frequency and field strength in most cases cannot directly tell the DL from the ACSE on the basis of the spectral shift. Increasing the field strength at the negative detuning is the only configuration in which the exciton peak shift driven by the DL differs from that by the ACSE. Even in this configuration, we must keep the exciton dimensionality crossover effect in mind, which makes the direct recognition of the DL more complicated. The $2P_z$ state position follows the miniband edge closely, while the $1S$ state position is insensitive to the DL effect, so we propose to measure the enhancement of the $1S–2P_z$ splitting to identify the DL effect. Measurement of the $1S–2P_z$ splitting could be made by seeing two split peaks in the absorption spectrum when the THz frequency is in resonance with the splitting.
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