

Optical absorption spectra and dynamical fractional Stark ladders in semiconductor superlattices

Koo-Chul Je and Seung-Han Park

Institute of Physics and Applied Physics and Department of Physics, Yonsei University, Seoul 120-749, Korea

Yup Kim

Department of Physics Kyung-Hee University, Seoul 130-701, Korea

and Asia-Pacific Center for Theoretical Physics, Seoul, Korea

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The optical response of semiconductor heterostructure under a combined dc-ac fields, $F_{dc} + F_{ac} \cos \omega_L t$, is studied by calculating absorption spectra based on semiconductor Bloch equations. First, we investigate the periodic motion of the carrier in a band under a ac field or dc field. Under the ac field with the frequency ω_L , we find the periodic motion of ω_L , which is very similar to the Bloch oscillation under the dc field. The ladder spacing of quasienergy states under the ac field is observed to be $\hbar \omega_L$. Under combined ac-dc fields, we discover the absorption peaks corresponding to the so-called dynamical fractional Stark ladders with ladder spacing $m\hbar\omega_B + m'\hbar\omega_L$ where m and m' are integers and ω_B is the Bloch oscillation frequency.

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Recently, coherent effects of applied external fields on the optical properties of the semiconductor quantum wells and superlattices have received considerable interests. The Bloch oscillating charge density induced by the dc (static) electric field generates a tera-hertz field that interacts with the Bloch electrons.¹ The carrier dynamics in presence of the combined dc-ac field, $F(t) = F_{dc} + F_{ac} \cos \omega_L t$, has also drawn much attention. Here, F_{dc} is the strength of the dc field. F_{ac} and ω_L are the strength and the frequency of the ac field, respectively. The superposition of both fields causes new phenomena such as inverse Bloch oscillators,^{2,3} dynamical fractional Stark ladders (DFSLs), and assisted photon transports.⁴⁻⁹

In the presence of the static field, electrons exhibit the Bloch oscillation (BO) with the period of $\tau_B = h/eF_{dc}d$, whose stationary counterparts correspond to the Wannier-Stark ladders (WSLs) with energy spacing of $m\hbar\omega_B$. Here, m is either a positive or negative integer, $\omega_B = eF_{dc}d/\hbar$, and d is the superlattice constant.¹⁰⁻¹²

Under the influence of the ac field, a number of interesting effects have also been predicted, including dynamical localization due to band collapse.^{8,13,14} Carriers also show the periodic motion with the period of $\tau_D = 2\pi/\omega_L$ in k space. The counterparts in the frequency domain of this motion indicate dynamical Stark ladders (DSL's), which are the discrete quasienergy levels with energy spacing of $m'\hbar\omega_L$. Here, m' is an integer. The DSL's are very similar to the occurrences of WSL's.

Under the combined dc-ac fields, a parent band is split into several quasienergy subbands with a fractional ladder structure.^{4-7,15,16} These quasienergy levels, which is the so-called dynamical fractional Stark ladders (DFSL's), are related to a new periodic motion of carriers in k space due to a competition between the static field and the time-dependent field.^{15,16} The dependence of the quasienergy spectra on the ac and dc fields presented as an electric analogue of the well-known Hofstadter spectra under the magnetic field.¹⁸ Recently, these fractional ladder structures have been analyzed when the ratio of ω_B to ω_L is simple fractions and integers.¹⁵⁻¹⁷ These effects have been investigated analytically

from a tight-binding model of a two-band system and from a periodic superlattice potential formed by the δ -function-type barriers.⁴⁻⁷ Especially, Dignam¹⁵ suggested that the optical absorption peaks are shifted by approximately $meF(t_0)d$, where $F(t_0)$ is the strength of the ac field at the time when the optical pulse reaches the sample.

In this paper, we report on the periodic motion of the carriers in k space under the applied dc and/or ac fields. First, we demonstrate that DSL's correspond to the discrete quasienergy levels with energy difference $\hbar\omega_L$ and, under the combined ac-dc fields, the quasi-energy levels have ladder structure with energy spacing $m\hbar\omega_B + m'\hbar\omega_L$. Then, we show that such ladder structure between WSL's under the combined fields, which is the so-called DFSL, can exist by studying the optical absorption peaks. Next, we analyze the optical-absorption spectra in a real three-dimensional (3D) semiconductor superlattice under the combined ac and dc fields. In order to analyze the absorption peaks of 3D superlattices, we compare the structure of the peaks in 3D superlattice to DFSL with the energy spacing $m\hbar\omega_B + m'\hbar\omega_L$, which is nearly the exact results for 1D superlattice. Finally, we find that we can interpret the physical origins of the variety of the peaks in the optical-absorption spectra in the anisotropic semiconductor superlattices under the influence of the combined dc-ac fields.

In order to consider the motion of the carriers in the energy band by the applied field, we calculate the single-particle energy, $\epsilon(k)$, which is obtained from the Kronig-Penney equation for the superlattices. When the external field $F(t)$ is applied along the growth axis of the superlattice, the motion of a carrier in an energy band can be described by the acceleration theorem, $\hbar\dot{k}(t) = qF(t)$, where q is the charge of the carrier. Therefore, under the combined field, the electron that is initially in the energy state of $\epsilon(k_0)$ will be in the state with the energy of $\epsilon(k_0 - eF_{dc}t/\hbar - eF_{ac}\sin\omega_L t/\hbar\omega_L)$ at a later time t .¹⁹ The motion of the carrier in the energy band results in a modulation of the amplitude of the optical polarization related to the entire photoexcited wave packet.

Optical spectra can be calculated by the semiconductor Bloch equations with an external field of $F(t)$ as:²⁰

$$\begin{aligned} \left[\frac{\partial}{\partial t} + \frac{e\vec{F}(t)}{\hbar} \nabla_{\mathbf{k}} \right] P_{\mathbf{k}} &= \frac{1}{i\hbar} (\mathcal{E}_{\mathbf{k}}^e + \mathcal{E}_{-\mathbf{k}}^h) P_{\mathbf{k}} + \frac{1}{i\hbar} \mathcal{U}_{\mathbf{k}} (1 - f_{\mathbf{k}}^e - f_{-\mathbf{k}}^h) + \frac{\partial P_{\mathbf{k}}}{\partial t} \Big|_{scat}, \\ \left[\frac{\partial}{\partial t} + \frac{e\vec{F}(t)}{\hbar} \nabla_{\mathbf{k}} \right] f_{\mathbf{k}}^e &= \frac{1}{i\hbar} (\mathcal{U}_{\mathbf{k}} P_{\mathbf{k}}^* - \mathcal{U}_{\mathbf{k}}^* P_{\mathbf{k}}) + \frac{\partial f_{\mathbf{k}}^e}{\partial t} \Big|_{scat}. \end{aligned}$$

Here, $P_{\mathbf{k}}$ is the interband polarization and $f_{\mathbf{k}}^e (= f_{\mathbf{k}}^h)$ is a distribution function of electron (hole). The renormalized electron (hole) energy is $\mathcal{E}_{\mathbf{k}}^e = \epsilon_{\mathbf{k}}^e - \sum_{\mathbf{k}'} V^{ee}(\mathbf{k}, \mathbf{k}') f_{\mathbf{k}'}^e$, [$\mathcal{E}_{\mathbf{k}}^h = \epsilon_{\mathbf{k}}^h - \sum_{\mathbf{k}'} V^{hh}(\mathbf{k}, \mathbf{k}') f_{\mathbf{k}'}^h$]. $\epsilon_{\mathbf{k}}^{e(h)} \equiv \epsilon^{e(h)}(\mathbf{k}) = \hbar^2 k_{\parallel}^2 / 2m^* + \epsilon(k_z)$ is the single-particle energy, where $\epsilon(k_z)$ is obtained from the Kronig-Penney equation for the superlattice, and $V(\mathbf{k}, \mathbf{k}')$ denotes the Coulomb matrix elements.²¹ The renormalized Rabi-frequency is $\mathcal{U}_{\mathbf{k}} = \mu_{\mathbf{k}} E(t) - \sum_{\mathbf{k}'} V^{eh}(\mathbf{k}, \mathbf{k}') P_{\mathbf{k}'}$, where $E(t)$ is the incident optical laser field and $\mu_{\mathbf{k}}$ is the optical dipole matrix element. Beyond the Hartree-Fock approximation $\partial/\partial t|_{scat}$ represents the scattering terms, such as interactions of carrier with other quasiparticles, contributing to a dephasing of the optical absorption spectra. We consider electron-LO-phonon coupling up to second order with the well-known Markov-approximation in the semiconductor Bloch equations. The coupling contributes to dephasing of both intra- and interband polarizations as well as energy-relaxation processes.²² For our theoretical approach, we introduce a moving coordinate frame $\tilde{t} = t$ and $\tilde{\mathbf{k}}(t) = \mathbf{k} - eF_{dc}t/\hbar - eF_{ac} \sin \omega_L t / \hbar \omega_L$.

In order to investigate one dimensional (1D) superlattice, we assume the superlattice of 111 Å-GaAs well layers and 17 Å-AlGaAs barrier layers. The width of combined miniband is set as $\Delta = \Delta_c + \Delta_v = 14$ meV, where $\Delta_c (\Delta_v)$ is the width of the first conduction (valence) miniband. The band-gap (ϵ_0) between these bands is $\epsilon_0 = \hbar \omega_0 = 1550$ meV. All the calculations are performed at $T = 10$ K. The incident laser pulse $E(t)$ is assumed as $E(t) = E_0(t) \exp(i\omega_0 t)$, where $E_0(t)$ has the Gaussian whose full width at half maximum is assigned to be 50 fs. In the analysis of the 1D superlattice, we do not consider the effects of Coulomb interactions. The results are shown in Figs. 1, 2, and 3.

Figure 1(a) displays the temporal variation of an energy state for the various applied dc fields, where $eF_{dc}d = 5, 10, \text{ or } 15$ meV. The dc field induces an oscillating motion of the carrier in k space. The temporal variation of the state has the same period of Bloch oscillation (BO), $\tau_B = \hbar / eF_{dc}d$. Figure 1(b) shows the linear absorption spectra of the superlattice under the same dc fields as those in Fig. 1(a). The peak at 1.550 eV of the linear absorption spectra for the various strengths of the applied dc field corresponds to the band gap of ϵ_0 . A series of equally spaced peaks, which correspond to the so-

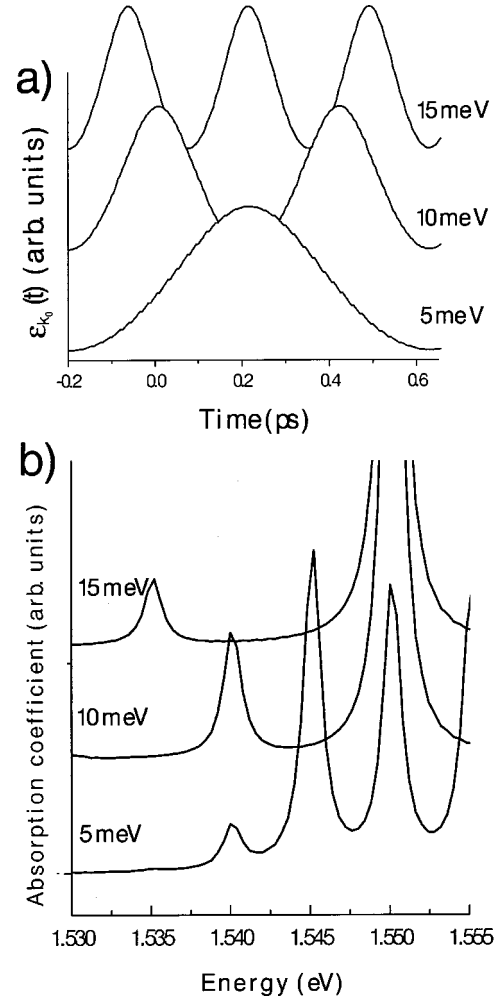


FIG. 1. (a) Temporal variations $\epsilon_{k_o}(t)$ of an energy state due to the periodic motion of the carrier with the initial Bloch momentum k_o under the several applied dc fields. Here we show the case for the conduction electron with $k_o = 0$. The variations are shown for the conduction electron with $k_o = 0$. The variations are shown for the conduction electron with $k_o = 0$, where the periods of the variations are exactly the same as the values of $\tau_B = \hbar / eF_{dc}d$. (b) The absorption spectra under the same strengths of dc fields as in (a). The main peak at 1.550 eV corresponds to the band-gap energy. The other peaks correspond to WSL's with the energy spacing $eF_{dc}d$.

called Wannier Stark Ladders (WSL's) with $\epsilon_0 + m\hbar\omega_B = \epsilon_0 + meF_{dc}d$, can be explicitly seen in Fig. 1(b). The location of the peaks corresponds to the frequency of the periodic motion or BO of the carrier in an energy band. The peaks with only negative indices of m are shown in Fig. 1(b) for convenience. We have also observed that the same series of the absorption peaks corresponding to the state with positive indices of m clearly exist. The oscillator strengths for the peaks become smaller as $|m|$ increases, because the magnitude of the overlap integral between electron and hole wave function gets smaller. This fact explains why the peaks corresponding to the WSL's with $|m| > 2$ cannot be seen clearly in Fig. 1(b).

Figure 2 shows the optical responses under two different ac fields with frequency $\hbar\omega_L = 15$ meV. As can be seen in Fig. 2(a), the periodic variation of the energy state due to the

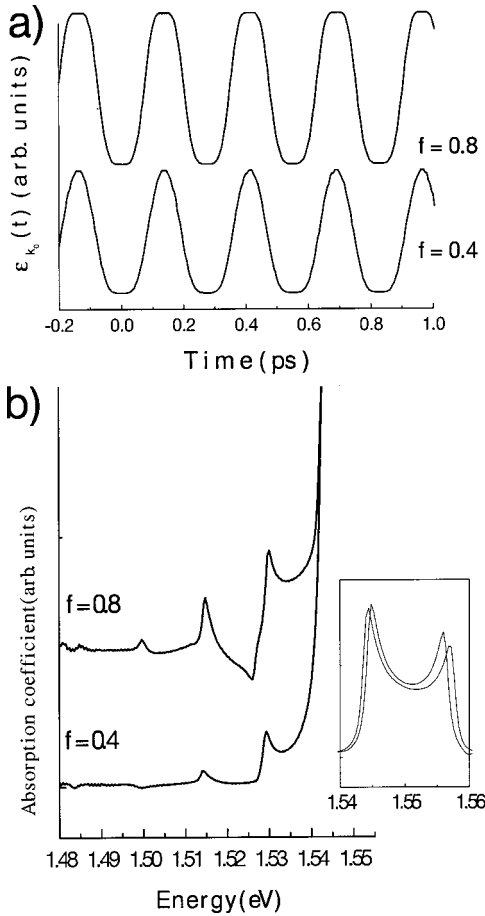


FIG. 2. (a) Temporal variations $\epsilon_{k_0}(t)(k_0=0)$ of an energy state due to the periodic motion of the carrier under the several ac fields with the fixed frequency ($\hbar\omega_L=15$ meV). The variations are shown for $f=0.4$ and 0.8 , where the periods of the variations are exactly the same as $\tau_L=2\pi/\omega_L$ regardless of values of f . (b) The corresponding absorption spectra. Inset shows the splitting of the peak at the band-gap energy 1.550 eV into two peaks at $\epsilon'_o=1.545$ eV and $\epsilon''_o=1.555$ eV. The other peaks corresponds to DSL's at $\epsilon'_o+m'\hbar\omega_L(m'<0)$.

motion of the carrier can be identified as clearly as BO, as displayed in Fig. 1(a). The period τ_L is the same as that of the applied ac field (or equal to $\tau_L=2\pi/\omega_L$), regardless of the strength of the ac field or the numerical values of the parameter, $f=eF_{ac}d/\hbar\omega_L$. The periodic motion under the ac field may have subperiodic motion. In other words, the quasienergy states can be deformed to have additional period as the field strength f is increased. We have observed the subperiodic motion clearly when $f>1.0$. This might be an indication of band-collapse effects. The variation of band width (Δ) due to band collapse induced by the ac field can be estimated from the function of $\Delta=\Delta_o J_0(f)$,⁸ where Δ_o is the width without any external field and J_0 is the zeroth-order Bessel function. Figure 2(b) shows the linear absorption spectra of the superlattice under the same ac fields.

In the presence of the dc field [or in Fig. 1(b)], the peaks of the absorption spectra corresponding to the splitting of the energy state into the ladder states of $\hbar\omega_L$, which we call the dynamical Stark ladders (DSL's), can be identified. The

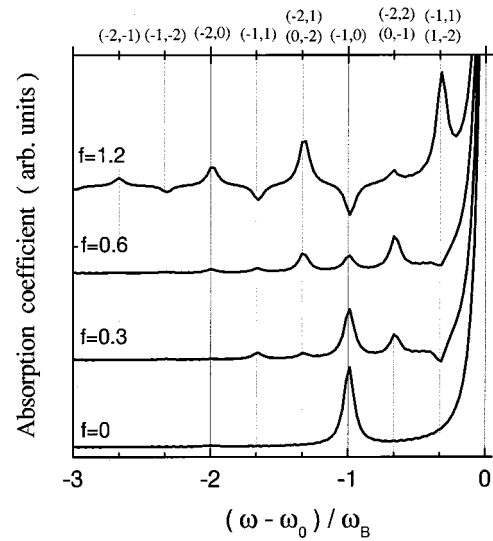


FIG. 3. Absorption spectra under the combined dc-ac fields when $\hbar\omega_B=eF_{dc}d=15$ meV and $\hbar\omega_L=10$ meV. The spectra are shown for the different strengths of the ac fields or $f=0, 0.3, 0.6$, and 1.2 . ω_o is the center frequency of the incident laser pulse and $\epsilon_o=\hbar\omega_o$. Indices such as $(-1,1)$, $(-2,2)$, and etc. in the upper part of the figure are for the pointing out the spectral locations corresponding to the fractional dynamical Stark ladders with $\epsilon(m,m')=\epsilon_o+m\hbar\omega_B+m'\hbar\omega_L$. Here, absorption spectra only for $\epsilon(m,m')<\epsilon_o$ are shown for the convenience.

peaks of the absorption spectra can be explained by the ladder structure with energy of $\epsilon'_o+m'\hbar\omega_L$. ϵ'_o is the shifted energy level of ϵ_o itself by the ac field through the band collapse.^{8,13,14} This shift of ϵ_o reflects the band-collapse effects as mentioned earlier.⁸ Then the peaks at the energies less than $\epsilon'_o=1.545$ eV in Fig. 2(b) can be identified by $\epsilon'_o+m'\hbar\omega_L$, where $\hbar\omega_L=15$ meV. Corresponding peaks are those at 1530 meV ($m'=-1$), 1515 eV ($m'=-2$), 15 eV ($m'=-3$), etc. Although we do not show the absorption spectra for the energy larger than $\epsilon''_o=1.555$ eV, we have also confirmed that the peaks corresponding to $\epsilon''_o+m'\hbar\omega_L(m'>0)$ exist. The peaks with the ladder spacing exist at the same spectral energies, regardless of values of F_{ac} or $f=(eF_{ac}d)/(\hbar\omega_L)$. The peaks due to DSL's at $\epsilon'_o+m'\hbar\omega_L(m'<0)$ and $\epsilon''_o+m'\hbar\omega_L(m'>0)$ correspond to the periodic frequency of the electrons under the ac field. When $f>1.0$, additional peaks or valleys are appeared in the absorption spectra. These might be originated from the correlation between the periodic motion of the carriers in a band and the band-collapse effects with $\Delta=\Delta_o J_0(f)$.⁸

Based on these results, we have also studied the linear absorption spectra when the ac field and dc field are simultaneously applied. The frequency of the applied ac field is fixed as $\hbar\omega_L=10$ meV, but the strength of the ac field (or the value of f) is varied. The strength of the dc field is fixed as $eF_{dc}d=15$ meV. The peak corresponding to the band gap (ϵ_o) is located at 1.550 eV and is not shifted. As can be seen in Fig. 2(b), the band-collapse effects due to the ac field, shift the band-gap peak to ϵ'_o and ϵ''_o . However, under the combined dc-ac field, we find that the peak is not shifted. In

contrast to large values of f or large strength of the ac field, the band-collapse effects would make the spectra much complex.

The other peaks are spectrally located at $\epsilon(m, m') = \epsilon_0 + m\hbar\omega_B + m'\hbar\omega_L$, where m and m' are indices of WSL and DSL. In Fig. 3, we specify the locations of all the spectral frequencies corresponding to every $\epsilon(m, m') (< \epsilon_0)$ with $|m| \leq 2$ and $|m'| \leq 2$. It can be identified that the peak exists at every frequency. We have confirmed that the peaks corresponding to $\epsilon(m, m') > \epsilon_0$ explicitly exist in the absorption spectra. Peaks with indices $(m, m') = (-1, 0)$ and $(m, m') = (-2, 0)$ correspond to WSL's with index $m = -1$ and $m = -2$, respectively. Peaks with indices such as $(0, -1)$ and $(0, -2)$ are DSL's. The peaks with indices $(m \neq 0, m' \neq 0)$ such as $(-2, 1)$ and $(-1, 1)$ between WSL's are the dynamical fractional Stark ladders (DFSL's). DFSL's, which are the quasienergy states under the combined dc-ac fields, can be well interpreted by the energy states with $\epsilon(m, m') = \epsilon_0 + m\hbar\omega_B + m'\hbar\omega_L$. We have also observed that the quasienergy spectra under the combined dc-ac fields with $\omega_B = p\omega_L$ for an integer p are well assigned by the formula $\epsilon_0 + m'\hbar\omega_L$. The oscillator strength of the peaks for the given F_{ac} and F_{dc} gets smaller as the indices m or m' increases. These effects can be understood from the fact that the magnitude of the overlap integral between the electron wave function at the corresponding ladder state and the hole wave function gets smaller when index m or m' increases. As f increases, the oscillator strength for the peak for given indices (m, m') gets larger. (See the curve for $f = 1.2$.) In addition, some peaks for lower f become valleys for the higher f . [Compare the peak at $(-1, 0)$ for $f = 0.6$ to the valley at $(-1, 0)$ for $f = 1.2$.] This might be come from the correlation effects between the band collapse and the motion of the carrier in a band due to the acceleration theorem. However, such anomalous behavior for the relatively large strength of the ac field (or large f) is very difficult and complex to analyze at the present stage. The physical origin of such behavior is now under investigation.

Next we calculate the absorption spectra for a 3D semiconductor superlattice based on 1D results as described in Figs. 1, 2, and 3. We use an anisotropic superlattice with 75 Å-GaAs well layers and 17 Å-AlGaAs barrier layers. The absorption spectra are calculated with the full consideration of Coulomb interactions between carriers in the Bloch equation. The results are presented in Figs. 4 and 5 for 3D superlattices.

Figure 4 shows the absorption spectra under a dc field ($eF_{dc}d = 20$ meV) without applied ac field and those under an ac field ($f = 3.83$ and $\hbar\omega_L = 10$ meV) without applied dc field. The dc field has sufficient strength to provide WSL's as the frequency-domain counterpart of Bloch oscillation. The data have four dominant peaks. The peaks at 1.577, 1.559, and 1.542 eV correspond to WSL with $m = 0$, $m = -1$, and $m = -2$, respectively. Another peak around 1.60 eV is the WSL with $m = 1$. The ladder spacing between WSL's is around 18 meV, which is smaller than the value ($eF_{dc}d = \hbar\omega_B = 20$ meV). This discrepancy of about 2 meV comes from the Coulomb interactions. The positive WSL states are

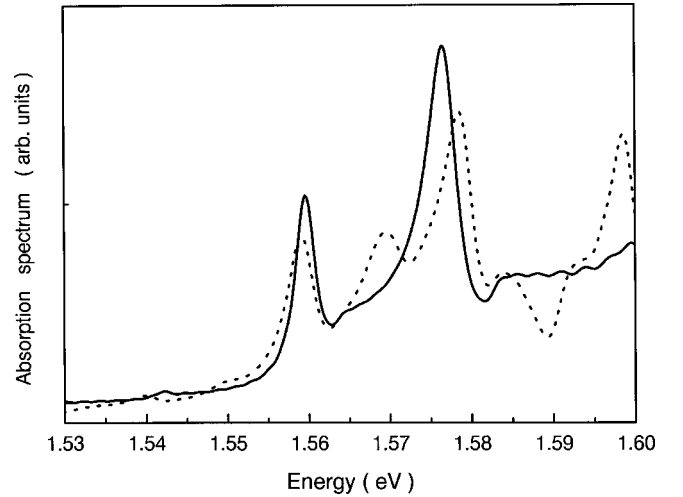


FIG. 4. The absorption spectra for $eF_{dc}d = 20$ meV and $f = 0$ (solid line) and $F_{dc} = 0$ and $f = 2.83$ with $\hbar\omega_L = 10$ meV (dotted line) in anisotropic three-dimensional structure.

largely broadened and the WSL peak with $m = 1$ is much weaker than the WSL peak with $m = -1$, since they exist in the ionization-continuum of negative WSL states.²¹ Figure 4 shows the case with ac field only with $f = 3.83$. The absorption spectra have an exciton peak at 1.578 eV ($m' = 0$) and its second eigen state at 1.569 eV ($m' = -1$). Another peaks at 1.559 eV ($m' = -2$), 1.55 eV ($m' = -3$), and 1.54 eV ($m' = -4$) correspond to dynamical Stark ladders (DSL's) caused by the ac field. The ladder spacing estimated from these DSL's is almost $\hbar\omega_L (= 10$ meV) as expected from 1D results (See Fig. 2). In practice, the scattering and screening effects make many possible peaks broadened. The absence of WSL's with positive indices should be due to the fact that two-dimensional density of states in the conduction band

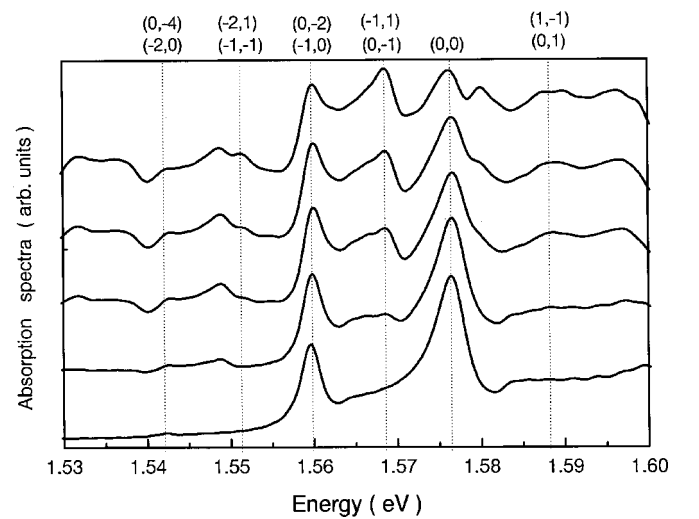


FIG. 5. Absorption spectra under the combined dc-ac fields in three-dimensional structure when $eF_{dc}d = 20$ meV and $\hbar\omega_L = 10$ meV. The spectra are shown for the different strengths of the ac fields or $f = 0, 0.2, 0.4, 0.6, 0.8,$ and 1 from the bottom. Indices in the upper part of the figure have the same meaning as in Fig. 3.

plays an important role.²² Thus, we believe that we only observe a few DSL's with $m' > 0$ in 3D anisotropic superlattice.

Figure 5 displays the absorption spectra under the ac fields with different strengths f and the dc field of a fixed strength $eF_{dc}d = 20$ meV. A peak at 1.577 eV corresponds the WSL with the index $m = 0$. As the strength of the ac field increases, the DSL's appear with the same spacing energy $\hbar\omega_L$. We observe also the DFSL's between WSL's positions. Nearly all of the peaks, corresponding to the spectral positions at the formula $\epsilon_o + m\hbar\omega_B + m'\hbar\omega_L$, exist. As explained in Fig. 3, almost all the peaks can be identified by (m, m') . Here, ϵ_o is the energy position of 1S excitonic absorption peak. However, all the spectral positions of the peaks do not exactly coincide with the positions calculated from the formula of $\epsilon_o + m\hbar\omega_B + m'\hbar\omega_L$. The energy spacing between WSL peaks are slightly different from the value estimated from the 1D results due to Coulomb interactions. The spacing between DSL's are almost the same as the value from the 1D results. These facts explain why the spectral positions of peaks induced by the combined fields are slightly different from the positions estimated from the formula, which explains 1D results relatively well. The oscillator strengths of DFSL's are modulated for different strength of ac fields, as such the one-dimensional structure. In the three-dimensional structure, the exciton peak can be bleached if the strength of the ac field in the combined applied fields is increased. It means that under the combined fields, the excitons can be delocalized and again localized,

repeatedly. These phenomena depend on the parameters of the combined fields. Thus, we conclude that the ac-field dependence of the optical spectra in the anisotropic superlattice for a given dc field is almost consistent with the theory of the 1D tight-binding model that the ac field modifies the combined band width as $\Delta = \Delta_o J_n(f)$.

In summary, we have analyzed the ladder structure of the quasienergy states of semiconductor superlattices under the combined dc-ac field. The most important correlation effects observed by a competition between dc-field- and ac-field-induced localizations are the quasi-energy states of the so-called dynamical fractional Stark ladders with $\epsilon_o + m\hbar\omega_B + m'\hbar\omega_L$, where m and m' are integers. We have also demonstrated that those states are the counterparts in the frequency domain of the periodic motion of the carriers in a band due to the acceleration theorem. However, the anomalous behaviors of the absorption spectra for large values of f (or F_{ac}) occurs, such as the enhancement of the peak at the given (m, m') and the change from the peak at some (m, m') for lower f to the valley, as f increases. This might be originated from the correlation effects between the band collapse and the motion of the carrier in a band. These anomalous behaviors remain to be investigated.

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