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LETTER TO THE EDITOR

Dynamic Fano resonance of Floquet-state excitons in superlattices

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Abstract. The dynamic Fano resonance between discrete quasienergy excitons and sidebands of their ionization continua is predicted and investigated in dc- and ac-driven semiconductor superlattices. This dynamic Fano resonance, well controlled by the ac field, opens another ionization channel for the otherwise bounded excitons.

The Fano resonance (FR) results from quantum coupling between a discrete state and a degenerate continuum of states and manifests itself in optical spectra as an asymmetric lineshape [1]. FR was observed in a variety of atomic and molecular systems. Recently, FR of excitonic transitions has been reported in semiconductor quantum wells [2, 3], in biased semiconductor superlattices [4], and in bulk GaAs in the presence of a magnetic field [5,6] where discrete excitons couple to continua of the lower transitions through a Coulomb interaction [7].

An intense ac field, e.g. by the free electron laser, can induce the dynamical localization [8] of electrons in superlattices, which is directly related to the collapse of the quasienergy band of Floquet states [9–11], the temporal analogue to the Bloch states in spatially periodic potential [12]. Research, taking into account the Coulomb interaction, has shown that the dynamical localization may cause the dimension cross-over of excitons [13] and the enhancement of exciton binding and optical strength in linear [13, 14] and nonlinear spectra [15]. Meanwhile the dynamical delocalization effect of an ac field is also investigated on localized Wannier–Stark (WS) states [16] in biased superlattices [10, 11, 15], which is hindered by Coulomb binding [15].

In this letter, we predict the dynamic Fano resonance (DFR) between quasienergy exciton states in biased superlattices driven by an intense THz field. While the conventional FR in semiconductors roots in static coupling, like the Coulomb interaction between different subbands [2–6, 17, 18] or interface scattering between Γ- and X-valleys [19], this DFR results from the coupling between a discrete quasienergy exciton and the continuum of quasienergy excitons associated with a *neighbour* sideband, which is in substance a nonlinear optical process with respect to the THz field. This DFR will also lead to the dynamical ionization of bound excitons in superlattices, which itself is a new effect of the ac field.

The one-dimensional tight-binding model [15, 20] is adopted in the present investigation. By excluding the realistic three-dimensional excitonic motion, the results cannot be compared to experimental data quantitatively, but we believe this simple model does present qualitatively correct results, in view of the fact that the ground WS exciton-state usually possesses much

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larger oscillator strength than its ionization continuum of in-plane motion, and the coupling induced by the ac field between the in-plane motion associated with different WS states is negligible because of their approximate orthogonality.

Within the linear response regime, the interband polarization $p_k \equiv \langle a_{\mathrm{h}-k} a_{\mathrm{e}k} \rangle$ satisfies the inhomogeneous equation [20]

$$\partial_t p_k = -\mathrm{i}\,[E_0 + \varepsilon_k]\, p_k + [\omega_{\mathrm{BO}} + \lambda \cos(\omega t)]\, \partial_k p_k + \mathrm{i} \sum_{\mathrm{k}'} \mathrm{V}_{\mathrm{k,k}'} \mathrm{p}_{\mathrm{k}'} + \mathrm{i}\chi(\mathrm{t}) - \gamma_2 \mathrm{p}_{\mathrm{k}} \qquad (1)$$

where $a_{\mathrm{e}k}$ ($a_{\mathrm{h}k}$) is the electron (hole) annihilation operator for the quasi-momentum state $|\mathrm{e(h)}, k\rangle$, E_0 is the separation between centres of the conduction and the valence minibands, $\varepsilon_k = -\frac{\Delta}{2}\cos k$ is the combined e–h miniband dispersion, ω_{BO} is the Bloch oscillation frequency due to the dc-field [21], λ is the strength of the THz field with frequency ω, $V_{k,k'}$ is the matrix element of the Coulomb potential, $\chi(t)$ is the strength of a near infrared optical excitation (here the dipole element is assumed k-independent), and γ_2 is the dephasing rate phenomenologically introduced to account for the scattering effect.

The Schrödinger equation for the exciton wave function is just the homogeneous part of (1) with the dephasing term removed. The quasienergy is obtained by numerically diagonalizing the propagator

$$U(T + t, t) \equiv \hat{T} \exp\left(-\mathrm{i} \int_t^{T+t} H(t)\mathrm{d}t\right)$$

($H(t)$ is the Hamiltonian, $T \equiv 2\pi/\omega$), according to the secular equation

$$U(T + t, t) u_q(k, t) = \exp\left(-i\varepsilon_q T\right) u_q(k, t)$$

where $u_q(k, t)$ is the eigenstate with quasienergy ε_q. Obviously, for any integer number m, $u_{q,m} \equiv u_q \exp(\mathrm{i}m\omega t)$ is also a solution with quasienergy $\varepsilon_q + m\omega$, and, in analogy to the conventional Brillouin zone in quasi-momentum space [12], so is defined the Brillouin zone of quasienergy $\{[m\omega, (m + 1)\omega]\}$.

In the ac-driven system, the linear absorption spectrum, depending on the excitation profile $\chi(\Omega)$, can be calculated from the formula [22,23]

$$\alpha(\Omega) \propto \Im\,[P(\Omega)/\chi(\Omega)]$$

where $P(\Omega)$ is the Fourier transformation of the optical response $P(t) \equiv \sum_k p_k$. In this work, only the continuous wave absorption is considered, whereby the interband susceptibility and oscillator strength are of the same form as in the static case, except that the transition matrix element is replaced by its time-average and the transition energy by the quasienergy [23].

For the sake of simplicity, let us focus on a specific case. The ac field is resonant with the Bloch oscillation, and the Coulomb potential is of on-site type, i.e. $V_{k,k'} = V_0/N$ (N is the size of the basis set $\{|k\rangle\}$). Parameters for our calculations which correspond to typical superlattice structures and free electron lasers, are taken as follows. $V_0 = 10$ meV, $\Delta = 4V_0$, $\omega_{\mathrm{BO}} = \omega = 1.5V_0$, and $\gamma_2 = 0.1V_0$. The continuous excitation takes the form $\chi(t) = \chi \exp(-\mathrm{i}\Omega t)$ with χ taken as unity. N is chosen to be 40 for calculating quasienergy, and 80 for optical spectra. Our calculation shows that an extended basis set produces no significant difference.

As shown in figure 1, the quasienergy spectrum consists of continuous minibands† and a few well-separated discrete states below‡. As the ac field strengthens, owing to the dynamical

† The discretization of the realistic continuous miniband is only an artifact of the discrete model.

‡ It is well known that the contact Coulomb potential model contains single bounded state in vanishing electric field [24]. But in the present case, more than one discrete Floquet states can exist, because there are more than one unequally spaced WS states, which together with their sidebands generate the bounded Floquet states (see the dressed WS ladder picture explained below).

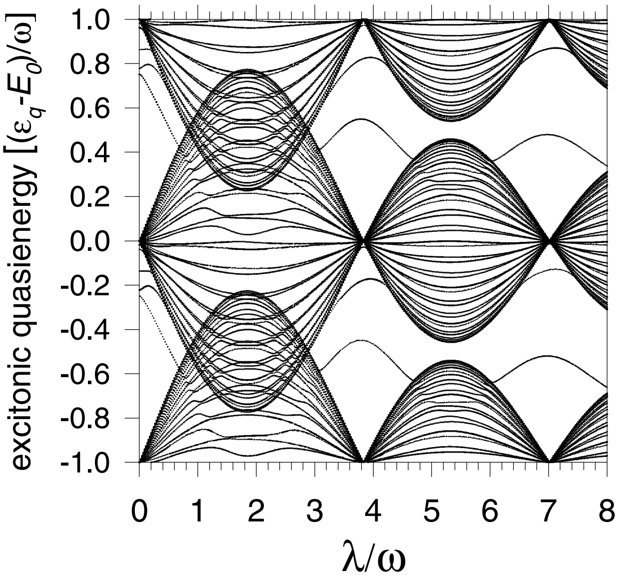

Figure 1. Quasienergy sideband structure of excitons as functions of the ac field strength.

delocalization, the miniband broadens and the discrete excited states§ merge into the continuum one by one. Meanwhile, the discrete ground state is repelled by its continuum towards lower energy, and eventually dips from above into the ionization continuum of its neighbour sideband, causing a series of anticrossings in the spectrum. Further enhancement of the ac field may suppress the quasienergy miniband, then the discrete exciton states can be released from the miniband one by one. At a certain strength, namely $\lambda = \lambda_{\mathrm{DL1}}$, the miniband collapses and dynamical localization takes place. After that the evolution described above will repeat with increasing ac field.

For an intuitive understanding, let us briefly review the picture of dressed WS ladders introduced in [15]. In weak ac field limit, only one-photon-assisted hopping between neighbour WS states $|n\rangle_x$ takes effect [15, 22], thus the time-periodic states $\{\exp(-\mathrm{i}n\omega t)|n\rangle_x\}$ form the Wannier basis set in a tight-binding 'lattice' with the nearest-neighbour hopping coefficient of $\lambda\Delta/(8\omega_{\mathrm{BO}})$ [15]. Those Wannier states with small n can be viewed as 'impurities' in a crystal since their 'on-site energy' $E_n^x - n\omega$ deviates from zero significantly due to the Coulomb interaction [25]. The discrete states in the quasienergy spectrum are concentrated on these 'impurities'; on the other hand, the remote WS states are almost equally spaced and the resonant photon-assisted hopping results in the formation of the continuum as wide as without the Coulomb interaction. As the dynamical delocalization is enhanced by an ac field, the discrete excited states may be ionized, and merge into the continuum. Calculation agrees well with the exact results for $\lambda < \omega/2$ (not shown). When the ac field is strong enough, the inter-sideband interaction with multi-photon processes [15] has to be invoked to account for the mixing between the discrete ground state and the continuum, the band suppression, and the dynamical localization.

The evolution of the linear absorption spectrum is displayed in figure 2 for several ac-field strengths ranging from zero up to that where the first dynamical localization takes place. The absorption spectrum in the absence of an ac field presents just the well known excitonic WS

§ Here the ground and excited states are designated according to their positions in a quasienergy Brillouin zone.

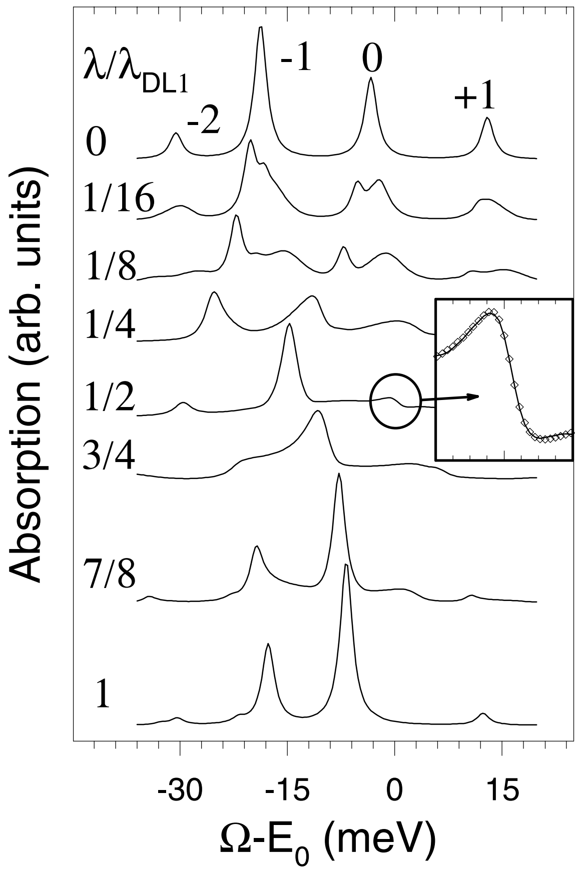

Figure 2. Absorption spectra in the dc- and ac-driven superlattice at various ac field strength as indicated by λ/λ_{DL1}. The integer numbers at the top denote the index of the WS states at vanishing ac field. Inset is an enlarged example of the lineshape due to the DFR, where the squares are fitted with (2).

ladder ($\lambda/\lambda_{DL1} = 0$). When the ac field is turned on, the WS states are coupled through photon-assisted hopping, so their sidebands start to gain in oscillator strength [15, 22], which looks as if the WS peaks were split ($\lambda/\lambda_{DL1} = 1/16$). As the excited discrete states merge into the continuum, the *intra-sideband* DFR occurs, and the oscillator strength is shared among the continuum states which otherwise have negligible optical strength [15]. This manifests itself in the spectrum as broadening of the original discrete line ($\lambda/\lambda_{DL1} = 1/16$ and $1/8$). When the ground exciton state couples to an energetically degenerate continuum associated with its neighbour sideband ($\lambda/\lambda_{DL1} = 1/8$, 1/4, 1/2, and 3/4), the *inter-sideband* DFR occurs, as characterized in the absorption spectra by asymmetric peaks with FWHM larger than $2\gamma_2$. As shown in the inset of figure 2, these peaks have perfect Fano lineshape [1]

$$\alpha(\Omega) = \alpha_0 + \alpha_c \frac{(q\gamma + \Omega - E_x)^2}{\gamma^2 + (\Omega - E_x)^2} \tag{2}$$

where α_0 is the background constant, α_c represents the continuum absorption without the inter-sideband coupling, q is the lineshape parameter, γ is related to the resonance broadening, and E_x denotes the position of the discrete state. When the ac field is enhanced further, the quasienergy-band shrinks, the discrete states emerge out of the continuum, and finally the absorption spectrum evolves from the DFR to Lorentzian-shaped discrete lines ($\lambda/\lambda_{DL1} = 7/8$ and 1). With strengthening the ac field even further, the evolution of the spectrum described above will roughly repeat (not shown).

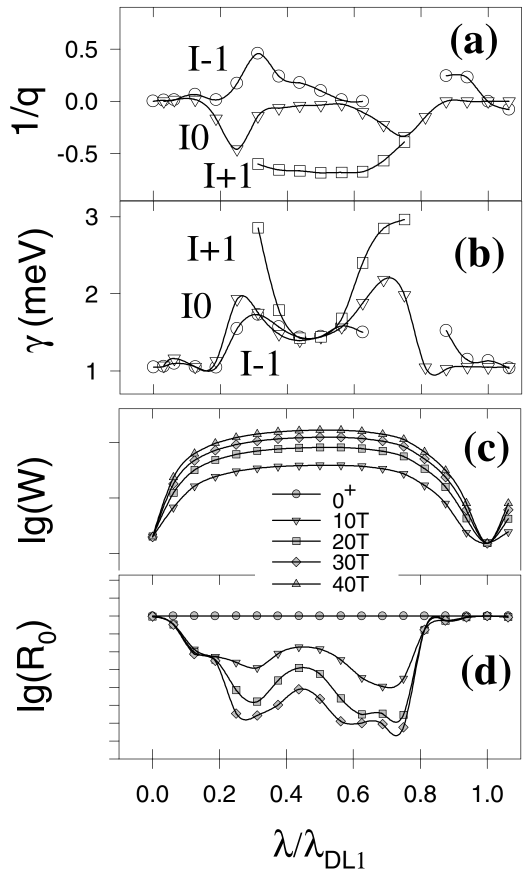

Figure 3. Dependence on the ac field strength of (a) the inverse lineshape parameter $1/q$ and (b) the Fano coupling parameter γ obtained by fitting with (2) the absorption peaks associated with the exciton ground state in figure 2, and (c) the mean-square root of the exciton radius W and (d) e-h overlap probability R_0 at $t = 0^+$, $10T$, $20T$, $30T$, and $40T$ after a δ-pulse excitation. In (a) and (b), the index 'Im' denotes the exciton ground state in the quasienergy Brillouin zone $[(m-1)\omega, m\omega]$ [$m = -1$ (circle), 0 (triangle), and +1 (square)].

The lineshape of the absorption peaks vary drastically when the ac field strength approaches the field at which the DFR occurs. As shown in figure 3, when the exciton ground state meets the continuous sideband, $1/q$ becomes nonzero, demonstrating the asymmetrical non-Lorentzian lineshape. Meanwhile the Fano coupling parameter, γ, is larger than the static dephasing rate of excitons, γ_2, indicating that the peak is further broadened by the DFR. According to [1],

$$\gamma = \pi |V_E|^2 \tag{3}$$

(in the present calculation, γ also includes the contribution from γ_2), and

$$1/q^2 = |\pi V_E\,(\mu_E/\mu_X)|^2 \tag{4}$$

where V_E denotes the coupling between the discrete and the continuum states, and μ_E and μ_X are the optical transition matrix element of the continuum and the discrete states, respectively. V_E is determined mainly by the overlap integral of the bound and extended excitons, which can be modulated by the ac field through changing the localization property of the excitons. Besides, the ac field redistributes the oscillator strength among the quasienergy states and their sidebands, changing the relative optical strength of the continuum. Thus both $1/q$ and γ, depending on the sideband index, are readily tuned by the ac field (see figure 3 (a) and (b)).

To study the localization property of excitons in the dc- and ac-driven superlattices, by integrating the Schrödinger equation in the Wannier representation, we have calculated the mean-square root of the exciton radius $W \equiv \langle \hat{r}^2 \rangle^{1/2}$ and the probability R_0 of finding the electron and hole at the same site, at various delay times after a weak δ-pulse excitation (see figure 3 (c) and (d)). W and R_0 can virtually be related to intraband and interband optical response, respectively. At $t = 0^+$, a δ-pulse excites the e–h pair at the same site, thus $W = 0$ and $R_0 = 1$. W increases unboundedly with time as a result of the resonant photon-assisted hoping between the evenly spaced remote WS states, whenever the quasienergy miniband width remains finite. This dynamical delocalization effect is handicapped somewhat by the Coulomb binding. The R_0 may saturate with time as long as the discrete state survives outside the continua. When the inter-sideband DFR occurs, however, the excitons are dynamically ionized with disappearance of such saturation behavior for R_0.

In summary, the dynamic Fano resonance between a discrete exciton and the sideband of ionization continuum is predicted in dc- and ac-driven superlattices, which, well controlled by the ac field strength, manifests itself as broadened asymmetric lineshape in absorption spectra and leads to the dynamic ionization for bound excitons. All these effects stem essentially from the unequal spacing of the excitonic WS ladder together with the dynamical delocalization effect of the ac field. This DFR effect was absent in the absorption spectra calculated in [13], because the sidebands of quasienergy miniband are not effectively overlapped for parameters chosen there. It should be pointed out that the DFR is quite different in nature from the FR between WS states and the degenerate in-plane continua associated with low-lying WS states in dc-biased superlattices [4]. The DFR is expected to be experimentally distinguishable from the in-plane FR in its ac field dependence. Moreover, it is possible to adjust the dc-field strength to minimize the in-plane FR effect [4], or, alternatively, to apply a quantizing magnetic field along the growth direction [26] to eliminate the in-plane FR effect, so as to emphasize the dynamic interference.

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