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Quasienergy Spectroscopy of Excitons

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We theoretically study nonlinear optics of excitons under intense THz irradiation. In particular, the linear near-infrared absorption and resonantly enhanced nonlinear sideband generation are described. We predict a rich structure in the spectra which can be interpreted in terms of the quasienergy spectrum of the exciton, via a remarkably transparent expression for the susceptibility, and show that the effects of strongly avoided quasienergy crossings manifest themselves directly, both in the absorption and transmitted sidebands.

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Excitons, strongly correlated pairs of electrons and holes, dominate the band-edge optics of semiconductors: their bound states are observed optically as resonances in the absorption spectrum in the gap of the semiconductor and in the continuum as enhanced absorption [1]. The interband susceptibility describes the linear optical properties of the semiconductor and can be expressed in terms of the well-known equation [2],

$$\chi(\omega) = -\sum_n \frac{|\psi_n(\vec{r} = \vec{0})|^2}{\hbar\omega - E_n + i0^+}, \qquad (1)$$

where $\psi_n(\vec{r})$ are the Wannier wave functions of the exciton with spectrum E_n. This compact formula is extremely useful since it relates the spectrum of the exciton directly to the resonances occurring in the absorption spectrum, which is proportional to the imaginary part of (1).

The development of coherent sources of intense electromagnetic radiation in the THz regime has opened up a new exciting area of semiconductor physics [3]: e.g., two color spectroscopy of excitons in the dynamical Franz-Keldysh (DFK) regime [4]. One color has weak intensity in the near infrared (NIR) regime inducing interband transitions, creating the excitons. The other color, which has high intensity, is in the far infrared regime (FIR), which corresponds to transitions between the internal degrees of freedom of the created excitons. In this regime the field strength, $\vec{E}_{\mathrm{FIR}}$, and the frequency, Ω, of the FIR are such that both strong field effects [dc Franz-Keldysh (FK) effect [5]] and multiphoton processes (MP) [6] are important. This regime is quantified by [7]

$$\gamma = \frac{e^2 E_{\mathrm{FIR}}^2}{4 m_r \hbar \Omega^3}, \qquad (2)$$

which is the ratio between the ponderomotive energy and the energy of a FIR photon. Here m_r is the reduced mass of the exciton and e is the electronic charge. The regime of FK corresponds to $\gamma \gg 1$, MP corresponds to $\gamma \ll 1$, while $\gamma \sim 1$ corresponds to the regime we consider presently, the DFK regime.

Study of the response of the weak NIR probe as a function of the two frequencies and the intensity of the FIR beam yields considerable insight into the nature of excitons and the fundamental electro-optical processes occurring in semiconductors. Experiments have been reported on the effects of the FIR on luminescence [8], absorption [4], and resonantly enhanced nonlinear mixing of the fundamental NIR with the FIR [9], even in the presence of strong magnetic fields [10]. The free particle properties of such experiments have been studied intensively [6,11–14], while excitonic effects on absorption have only been reported recently [4,15,16].

The goal of this Letter is to report a generalization of the textbook result (1) to the new dynamical regime $\gamma \sim 1$. We find that the macroscopic polarization can be expressed as

$$\vec{P}(t) = d^2 \vec{E}_{\mathrm{NIR}} \sum_n \chi_n(\omega) e^{i(\omega + n\Omega)t}, \qquad (3)$$

where

$$\chi_n(\omega) = -\sum_{\alpha n'} \frac{\phi^*_{\alpha n' + n} \phi_{\alpha n'}}{\hbar\omega - \tilde{\epsilon}_\alpha - n'\hbar\Omega + i0^+}. \qquad (4)$$

Here ω is the frequency of the NIR, d is the interband dipole matrix element, assumed to be constant, and $\phi_{\alpha n}$ are the temporal Fourier components of the Floquet states $\phi_\alpha(\vec{r} = \vec{0}, t)$ of the exciton, with *quasienergy* $\tilde{\epsilon}_\alpha$ [17]. A Floquet state is the temporal analog to a Bloch state in a spatially periodic potential. The spectral decomposition of the polarization [Eq. (3)] shows that the linear absorption of the NIR is proportional to $\mathrm{Im}\chi_0(\omega)$, while $I_n(\omega) = |\chi_n(\omega)|^2$ is proportional to the intensity of photonic sidebands which radiate at frequency $\omega + n\Omega$. Equation (4) thus relates the nonequilibrium optical properties directly to the excitonic quasienergy spectrum.

We first outline the derivation of (3) and then present a numerical study of a quantum well (QW) exciton, which illustrates the applicability of the theory. We show that *avoided crossings* in the quasienergy spectrum lead to two clear experimental consequences: (i) the sideband intensities I_n have a resonant behavior and (ii) the $1s$ resonance in optical absorption displays a strong Autler-Townes splitting [18].

In the following we describe the semiconductor via a simple spinless effective mass two-band model. The Hamiltonian is given by

$$H = \sum_{\vec{k},\mu\in\{c,v\}} \epsilon_\mu[\hbar\vec{k} + e\vec{A}(t)]c^\dagger_{\mu\vec{k}}c_{\mu\vec{k}} + H_i\,, \quad (5)$$

where μ labels conduction or valence bands, and H_i describes the Coulomb interaction via the potential $v(|\vec{k}|)$. Using nonequilibrium Green function methods, one can show that the retarded susceptibility obeys the integral equation [19],

$$\chi^r(\vec{k};t,t') = \bar{\chi}^r(\vec{k};t,t') + \int \frac{d^n\vec{k}'}{(2\pi)^n}\int dt''\,\bar{\chi}^r(\vec{k};t,t'') \times v(|\vec{k}-\vec{k}'|)\chi^r(\vec{k}';t'',t')\,, \quad (6)$$

where $\bar{\chi}^r(\vec{k};t,t')$ is the free-particle susceptibility [20].

Equation (6) can be transformed into an infinite-dimensional matrix equation and solved numerically by introducing a cutoff frequency [4,16]. This method has successfully described some effects in the linear absorption [4], but has failed to describe the nonlinear mixing adequately. The representation introduced here does not introduce cutoffs, is numerically much faster, and clarifies the physics involved. In the rotating wave approximation with respect to the NIR probe, the irreducible interband susceptibility for an undoped semiconductor obeys [13]

$$\left\{i\hbar\frac{\partial}{\partial t} - \epsilon[\hbar\vec{k} + e\vec{A}(t)]\right\}\bar{\chi}^r(\vec{k};t,t') = \delta(t-t')\,, \quad (7)$$

where $\epsilon(\hbar\vec{k}) = \epsilon_c(\hbar\vec{k}) - \epsilon_v(\hbar\vec{k})$ and $e = |e|$. The uniform FIR field is described by the vector potential $\vec{A}(t) = -\vec{E}_{\rm FIR}\sin(\Omega t)/\Omega$. With (7) we transform (6) to

$$\left\{i\hbar\frac{\partial}{\partial t} - \epsilon\left[\frac{\hbar}{i}\vec{\nabla}_{\vec{r}} + e\vec{A}(t)\right] + \frac{e^2}{4\pi\kappa r}\right\}\chi^r(\vec{r};t,t') = \delta(\vec{r})\delta(t-t')\,, \quad (8)$$

where κ is the effective dielectric constant in the semiconductor. To proceed, we first consider the solutions to the homogeneous part of (8),

$$\left\{\frac{[\frac{\hbar}{i}\vec{\nabla}_{\vec{r}} + e\vec{A}(t)]^2}{2m_r} + \epsilon_g - \frac{e^2}{4\pi\kappa r}\right\}\Psi(\vec{r},t) = i\hbar\frac{\partial\Psi(\vec{r},t)}{\partial t}\,. \quad (9)$$

This is a Schrödinger equation for a hydrogenlike particle, the exciton, in the presence of the intense FIR field. When the FIR field is absent this equation reduces to the Wannier equation [1]. We have thus arrived at a generalized Wannier equation with a time periodic Hamiltonian, $H(t) = H(t+T)$, where $T = 2\pi/\Omega$. Such a discrete time symmetry gives rise to the temporal analog of the Bloch wave functions, the Floquet states [17]: $\psi_\alpha(\vec{r},t) = e^{-i\tilde{\epsilon}_\alpha t/\hbar}\phi_\alpha(\vec{r},t)$, where $\tilde{\epsilon}_\alpha$ are the quasienergies and $\phi_\alpha(\vec{r},t) = \phi_\alpha(\vec{r},t+T)$. The Floquet states can be viewed as the stationary states of the periodically driven system. They form complete sets of solutions to the Schrödinger equation, which is to say that any wave function obeying (9) is of the form $\Psi(\vec{r},t) = \sum_\alpha c_\alpha e^{-i\tilde{\epsilon}_\alpha t/\hbar}\phi_\alpha(\vec{r},t)$, where c_α are c numbers. Furthermore, at equal times the states fulfill the closure relation $\sum_\alpha \phi^*_\alpha(\vec{r},t)\phi_\alpha(\vec{r}',t) = \delta(\vec{r}-\vec{r}')$. Using these properties, we expand the susceptibility in terms of the Floquet states and their quasienergies, and find

$$\chi^r(\vec{r};t,t') = \frac{\theta(t-t')}{i\hbar}\sum_\alpha e^{-i\tilde{\epsilon}_\alpha(t-t')/\hbar}\phi^*_\alpha(\vec{0},t')\phi_\alpha(\vec{r},t)\,. \quad (10)$$

We next set $\vec{r} = \vec{0}$, define $\phi_\alpha(t) = \phi_\alpha(\vec{0},t)$, and expand the states as $\phi_\alpha(t) = \sum_n \phi_{\alpha n}e^{-in\Omega t}$. The macroscopic polarization is then readily expressed as (3) and (4). This concludes the outline of our derivation.

We next apply the present theory to the nonlinear optical properties of a quantum well exciton. Specifically we choose $ea_0E_{\rm FIR} = 1E_{\rm Ry}$, where $E_{\rm Ry} = \hbar^2/(2m_ra_0^2)$ is the effective Rydberg energy of the medium, and $a_0 = \hbar^2\kappa/(e^2m_r)$ is the effective Bohr radius of the exciton. As an example, for InGaAs, $E_{\rm Ry} \sim 2$–3 meV and $a_0 \sim 200$ Å which leads to $E_{\rm FIR} \sim 10^5$ V/m, well within the range of free electron lasers [3]. The FIR is linearly polarized and the field oscillates in the plane of the QW. We sweep the THz frequency such that it probes the various internal resonances of the exciton and study the absorption and the nonlinear sideband generation.

A two-dimensional exciton in equilibrium has the bound state spectrum $E_n = E_{\rm gap} - E_{\rm Ry}/(n-0.5)^2$, $n > 0$. The $n = 1$ state is a nondegenerate s state, while $n \neq 1$ are degenerate containing also p states, etc. [21]. With linear polarization of the FIR the doubly degenerate p states may be decomposed into $p_\perp$ and $p_\parallel$, where the $p_\parallel$ has the same spatial symmetry as the field. Only $p_\parallel$ contributes to the dynamics. We include both s and $p_\parallel$ states in our calculations since both are physically important as they couple strongly in the presence of the THz field [22]. The Floquet states and their quasienergies are determined numerically. In the figures, we have introduced a phenomenological damping of the resonances in (4). In Fig. 1 we show the results of our calculation for fixed $\hbar\Omega = 2.55E_{\rm Ry}$ as a function of NIR frequency, illustrating the basic effects due to the THz field. Considering the absorption, the THz frequency is below the $1s \to 2p_\parallel$ equilibrium transition frequency, $\hbar\omega^0_{12} \approx 3.56E_{\rm Ry}$. The redshift of the $1s$ resonance, away from its equilibrium position, is due to the ac-Stark effect [23], which is pronounced even though the frequency is considerably detuned from the $1s \to 2p_\parallel$ transition. The effect is maximal in the $\gamma \sim 1$ regime. The $2s$

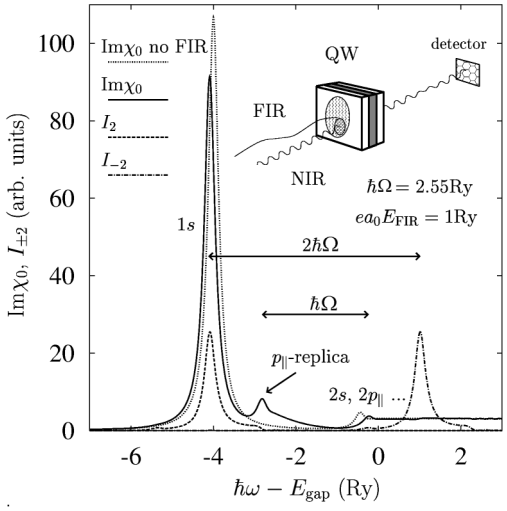

FIG. 1. The main effects due to intense FIR irradiation, colinear with weak NIR field. Linear optical absorption Imχ_0: (i) ac-Stark redshift of the $1s$ resonance, (ii) DFK blueshift of the $2s$ resonance and the band edge, (iii) suppression of oscillator strength, and (iv) emergence of a photon replica of the $p_\parallel$ state. Nonlinear mixing $I_{\pm2}$: sideband emission at $\omega \pm n\Omega$. The inset illustrates the experimental geometry.

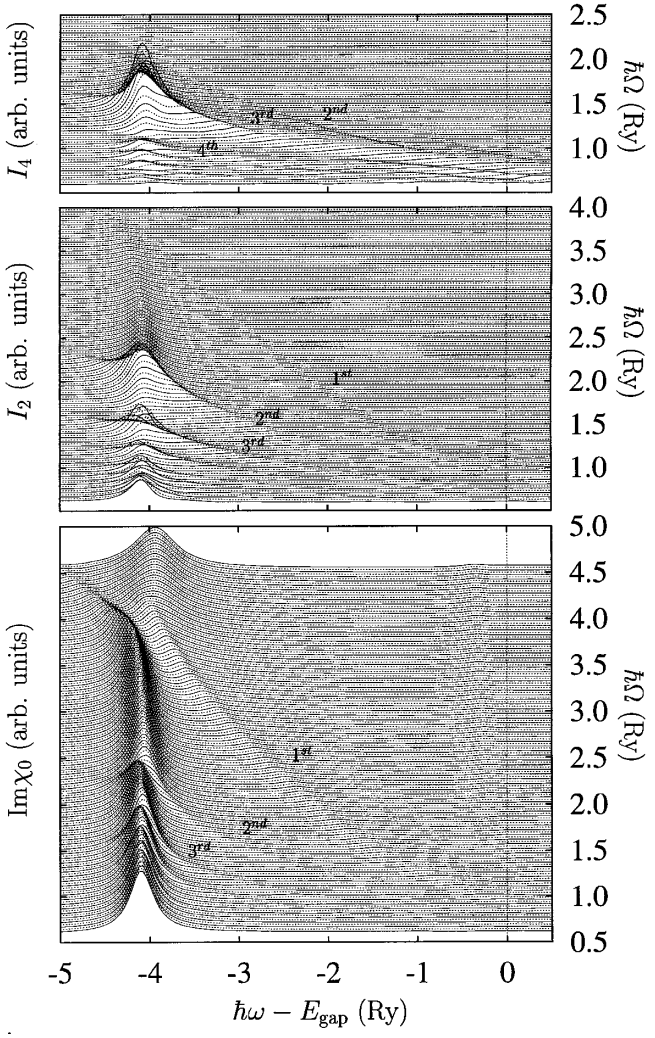

FIG. 2. The effect of the THz frequency at fixed field strength. From below; the absorption Imχ_0, two-, and four-photon sideband generation. The THz frequency is indicated on the right vertical axis. Here several avoided crossings in the quasienergy spectra can be distinguished, as splittings in the absorption and as resonant enhancement in the sideband generation. The order of the processes involved is indicated. I_4 is multiplied by 3 compared to I_2.

resonance is blueshifted with respect to the equilibrium position. The band edge is blueshifted as well. This is due to the DFK effect which shifts all main features by the ponderomotive energy [6,11,12]. The oscillator strength of the $1s$ resonance is suppressed. These effects have been observed in quantum well excitons [4]. Furthermore, a new resonance appears in the absorption spectrum: a single photon replica of the dark $p_\parallel$ state which under irradiation becomes optically active. The tail to the blue from the resonance is due to replicas of the $p_\parallel$ symmetric states in the continuum. The sideband intensity for $I_{\pm2}$ is shown as well. In reflection symmetric systems only sidebands with n even appear: the optical properties are invariant under the transformation $\vec{E}_{\rm FIR} \rightarrow -\vec{E}_{\rm FIR}$, and hence the "effective" frequency is 2Ω. Breaking the reflection symmetry introduces odd sidebands as well. The sideband generation is at a maximum if either ω or $\omega - n\Omega$, $n > 0$, is tuned to the $1s$ resonance. We find that $I_n(\omega) = I_{-n}(\omega + n\Omega)$. However, in an experiment using a multiple quantum well sample the radiation due to I_{-n} will tend to be reabsorbed, its maximum being tuned to the main absorption resonance while I_n has its maximum $2n\Omega$ away from it. This behavior of the sideband generation has been observed [9].

In Fig. 2 we show the spectra for a constant field strength with a THz frequency sweep $\hbar\Omega = (0.5\text{–}5)E_{\rm Ry}$. In Im$\chi_0$, one observes how photon replicas traverse from $\hbar\omega \sim E_2$ in a fan as the THz frequency is increased. The fan blades have been tagged with the order in the THz frequency involved, which is roughly proportional to the inverse slope of the blade. In view of (4), when the replicas reach the main resonance a strongly avoided crossing in the quasienergy spectrum results, which is directly visible in the spectra. For the first order process an Autler-Townes splitting results. The sidebands, in the upper two panels, show clear evidence of the fan shape. In this case, however, the underlying avoided crossing in the quasienergy spectrum results in resonantly enhanced sideband generation. Generation of sideband I_n calls for optical processes of order $|n| + 1$ or higher and thus the predominant order determines if a strongly avoided crossing results in a resonantly enhanced sideband.

The oscillator strength reflects the resonant conditions. In Fig. 3 we compare the oscillator strength to the maximum of the generated sidebands at each frequency. We remark in passing that the $n = 2$ sideband was recently detected for $\hbar\Omega \sim 4E_{\rm Ry}$ [9], which in view of Fig. 3

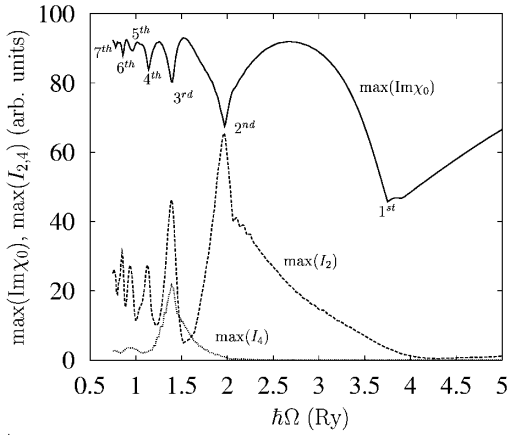

FIG. 3. The maximum absorption and sideband generation as a function of the FIR frequency illustrating the relation between the absorption suppression and the resonantly enhanced sideband generation.

suggests that the predicted results should be detectable with present technology. Resonant suppression of the oscillator strength occurs when the sideband generation is enhanced. The resonance conditions are met by the quasienergies according to (4). The range in which resonant enhancement occurs is determined by the coupling of the internal exciton levels. The enhancement mainly involves the $1s \rightarrow 2p_{\parallel}$ transition for odd processes, while it is mainly the $1s \rightarrow 2s$ transition which is responsible for even resonances. The first seven resonances are indicated in Fig. 3.

In summary, we have studied the nonlinear optics of excitons subject to intense THz radiation. We have shown analytically that observed resonances in the sideband generation may be viewed as manifestations of strongly avoided crossings in the quasienergy spectrum of the exciton. Our theory is consistent with experiment, where available, and it leads to a number of predictions for the outcome of new experiments. An example is the strong correlation between the oscillator strength suppression in absorption and the associated resonant enhancement in sideband generation, respectively.

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