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LETTER TO THE EDITOR

Coherent effects induced by dc–ac fields in semiconductor superlattices: the signature of fractional Wannier–Stark ladders

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Abstract. Using the semiconductor Bloch equations, we investigate the coherent effects induced by combined dc and ac fields through monitoring the square of the third-order nonlinear polarization (four-wave mixing signals). We find that when the ratio of the Stark frequency of the dc field ω_B to the ac-field frequency ω is a fractional number, the four-wave mixing signals reveal fractional Wannier–Stark ladders.

Since the development of semiconductor superlattices (SL), there has been intensive investigation of such novel quantum structures under the influence of static and time-dependent electric fields [1–6]. For example, one of the well-known semiclassical predicted phenomena in solid-state physics—Bloch oscillations (BO)—is induced by the dc field and has been observed by experimentalists in SL. Besides BO, many other new interesting phenomena have been found theoretically or/and experimentally; dynamic localization is a prominent such effect generated by ac fields [1]. The effects produced by dc–ac fields have drawn attention recently because of the many interesting phenomena inherent in such combined fields. Among these, multi-photon absorption [2], strong terahertz (THz) photon-current resonance at the Bloch frequency—the inverse Bloch oscillator [3], absolute negative conductance [4], and Rabi flop between the two minibands [5] are particular examples induced by such combined fields. The recent suggestion of fractional Wannier–Stark ladders [6] generated by the dc–ac field has also been made by Zhao *et al* on the basis of the application of quasi-energy eigenstates and perturbative calculations. They obtained the optical absorption strength, and found that, when the ratio of the Stark frequency of the dc field ω_B ($=eF_0d/\hbar$, where F_0 is the amplitude of the dc electric field, and d is the SL lattice constant) to the ac-field frequency ω is a fraction (say, p/q, where p and q are relatively prime numbers), the additional absorption lines appear equidistantly at $1/p, 2/p, \ldots, (p-1)/p$ throughout the frequency (energy) interval of ω_B (see figure 2 of reference [6]). In particular, these fractional Wannier–Stark ladders have been theoretically demonstrated to arise in a simulation by using an ultra-cold atom ensemble driven by accelerated optical potentials [7].

It has been suggested that one could observe coherent effects induced by external electric fields by probing the square of the third-order nonlinear optical polarizations: degenerate four-wave mixing (DFWM) signals [8, 9]; this technique is often used to investigate bulk and quantum-confined semiconductor structures [10]. Using the semiconductor Bloch equations (SBE) [11, 12], Meier *et al* found by monitoring the THz emission and DFWM signals that photo-excited carriers driven by the dc field can still experience BO even in the presence of excitonic Coulomb interaction that is comparable to the miniband widths of SL [11], while the carriers driven by the ac field reveal dynamic localization provided that $edF_1/\hbar\omega$ coincides with the roots of the ordinary Bessel function of order zero [1, 11], where F_1 is the amplitude of the ac field.

In the following, proceeding along the same lines as Meier *et al*, we investigate the coherent effects produced by the combined dc–ac field. We find in this letter that time-resolved DFWM can reveal fractional Wannier–Stark ladders. In other words, in the time domain, when $\omega_B/\omega = p/q$, the DFWM signals have periods that are multiples of q times the period of the ac field, $T_{ac} = 2\pi/\omega$ (this point will be expounded on in the following discussion).

Our starting point is the following pair of SBEs:

$$\left[\frac{\partial}{\partial t} - \frac{e}{\hbar}\boldsymbol{F}(t)\cdot\boldsymbol{\nabla}_{\boldsymbol{k}} - \frac{\mathrm{i}}{\hbar}[e_c(\boldsymbol{k},t) - e_v(\boldsymbol{k},t)]\right]P(\boldsymbol{k},t) = \frac{\mathrm{i}}{\hbar}[n_c(\boldsymbol{k},t) - n_v(\boldsymbol{k},t)]\Omega(\boldsymbol{k},t) + \left.\frac{\partial P(\boldsymbol{k},t)}{\partial t}\right|_{\mathrm{coll}} \tag{1}$$

$$\left[\frac{\partial}{\partial t} - \frac{e}{\hbar}\boldsymbol{F}(t)\cdot\boldsymbol{\nabla}_{\boldsymbol{k}}\right]n_{c,(v)}(\boldsymbol{k},t) = \frac{\mp 2}{\hbar}\,\mathrm{Im}[\Omega(\boldsymbol{k},t)P^*(\boldsymbol{k},t)] + \left.\frac{\partial n_{c,(v)}(\boldsymbol{k},t)}{\partial t}\right|_{\mathrm{coll}} \tag{2}$$

where $n_{c,(v)}(\boldsymbol{k},t)$ are the electron (hole) populations in the conduction band and valence band respectively, and $P(\boldsymbol{k},t)$ is the interband polarization. Also

$$e_{c,(v)}(\boldsymbol{k},t) = \epsilon_{c,(v)}(\boldsymbol{k},t) - \sum_{\boldsymbol{k}'} V(\boldsymbol{k},\boldsymbol{k}')n_{c,(v)}(\boldsymbol{k}',t)$$

are the electron and hole energies with the Coulomb interaction taken into account, while $\epsilon_{c,(v)}(\boldsymbol{k},t)$ are the corresponding energies of the conduction and (valence) bands with no Coulomb interaction. Additionally,

$$\Omega(\boldsymbol{k},t) = \mu E(t) + \sum_{\boldsymbol{k}'} V(\boldsymbol{k},\boldsymbol{k}')P(\boldsymbol{k}',t)$$

is the renormalized Rabi frequency, and $V(\boldsymbol{k},\boldsymbol{k}')$ is the Coulomb potential in the quasi-momentum space. $E(t)$ and $\boldsymbol{F}(t)$ are the optical field and the dc–ac field respectively. The optical field is defined as

$$E(t) = \sum_{j=1,2} E_j(t)\exp[-\mathrm{i}(\boldsymbol{k}_j\cdot\boldsymbol{r} - \omega_j t)]$$

where $E_j(t)$ is Gaussian laser pulse defined by

$$E_j(t) = E_0\exp(-t^2/\tau^2).$$

The last two terms on the right-hand side of equations (1) and (2) describe the dephasing processes phenomenologically. In the SBEs, the time-dependent Hartree–Fock approximation is adopted, and exciton–exciton correlations are neglected, while the full description of the coherent effects in crystalline semiconductors needs the inclusion of the higher-order correlation effect [13]. In this letter, we simply want to show qualitatively, by using SBEs, that the combined dc–ac fields can generate fractional Wannier–Stark ladders.

In the low-excitation regime, perturbative expansion of equations (1) and (2) in terms of the optical field can be performed by means of the following procedure [11, 14, 15]: we use $n_{c,(v)}(\boldsymbol{k},t) = n^{(0)}_{c,(v)}(\boldsymbol{k},t) + n^{(2)}_{c,(v)}(\boldsymbol{k},t) + \cdots$ and $P(\boldsymbol{k},t) = P^{(1)}(\boldsymbol{k},t) + P^{(3)}(\boldsymbol{k},t) + \cdots$, with the following initial conditions: $n_c(\boldsymbol{k}, t=0) = 0, n_v(\boldsymbol{k}, t=0) = 1$. Up to the third order, we get the following equations of which the first two have already been given in reference [11]:

$$\left[\frac{\partial}{\partial t} - \frac{e}{\hbar}\boldsymbol{F}(t)\cdot\nabla_{\boldsymbol{k}} - \frac{\mathrm{i}}{\hbar}\left[\epsilon_c(\boldsymbol{k},t) - \epsilon_v(\boldsymbol{k},t) + \sum_{\boldsymbol{k}'} V(\boldsymbol{k},\boldsymbol{k}')\right]\right]P^{(1)}(\boldsymbol{k},t)$$
$$= \frac{-\mathrm{i}}{\hbar}\left[\mu E(t) + \sum_{\boldsymbol{k}'} V(\boldsymbol{k},\boldsymbol{k}')P^{(1)}(\boldsymbol{k}',t)\right] + \left.\frac{\partial P^{(1)}(\boldsymbol{k},t)}{\partial t}\right|_{\mathrm{coll}} \tag{3}$$

$$\left[\frac{\partial}{\partial t} - \frac{e}{\hbar}\boldsymbol{F}(t)\cdot\nabla_{\boldsymbol{k}}\right]n^{(2)}_{c,(v)}(\boldsymbol{k},t)$$
$$= \frac{\mp 2}{\hbar}\,\mathrm{Im}\left[\left(\mu E(t) + \sum_{\boldsymbol{k}'} V(\boldsymbol{k},\boldsymbol{k}')P^{(1)}(\boldsymbol{k}',t)\right)P^{(1)*}(\boldsymbol{k},t)\right] + \left.\frac{\partial n^{(2)}_{c,(v)}(\boldsymbol{k},t)}{\partial t}\right|_{\mathrm{coll}} \tag{4}$$

$$\left[\frac{\partial}{\partial t} - \frac{e}{\hbar}\boldsymbol{F}(t)\cdot\nabla_{\boldsymbol{k}} - \frac{\mathrm{i}}{\hbar}\left[\epsilon_c(\boldsymbol{k},t) - \epsilon_v(\boldsymbol{k},t) + \sum_{\mathrm{k}'} V(\boldsymbol{k},\boldsymbol{k}')\right]\right]P^{(3)}(\boldsymbol{k},t)$$
$$= \frac{\mathrm{i}}{\hbar}[n^{(2)}_c(\boldsymbol{k},t) - n^{(2)}_v(\boldsymbol{k},t)]\left[\mu E(t) + \sum_{\boldsymbol{k}'} V(\boldsymbol{k},\boldsymbol{k}')P^{(1)}(\boldsymbol{k}',t)\right]$$
$$- \frac{\mathrm{i}}{\hbar}\sum_{\boldsymbol{k}'}\left(V(\boldsymbol{k},\boldsymbol{k}')[n^{(2)}_c(\boldsymbol{k}',t) - n^{(2)}_v(\boldsymbol{k}',t)]\right)P^{(1)}(\boldsymbol{k},t)$$
$$- \frac{\mathrm{i}}{\hbar}\sum_{\boldsymbol{k}'} V(\boldsymbol{k},\mathrm{k}')P^{(3)}(\boldsymbol{k}',\boldsymbol{t}) + \left.\frac{\partial P^{(3)}(\boldsymbol{k},t)}{\partial t}\right|_{\mathrm{coll}}. \tag{5}$$

For simplicity, we concentrate on the one-dimensional case, where the dc–ac field is assumed to be along the growth direction of the SL: $F(t) = F_0 + F_1\cos(\omega t)$. We use the tight-binding model: $\epsilon_c = E_c + (\Delta_c/2)\cos(kd), \epsilon_v = E_v - (\Delta_v/2)\cos(kd)$, and the contact Coulomb potential. The feasibility of applying these has been demonstrated in several previous studies [11, 13, 16].

Equations (3)–(5) are partial differential equations, but the partial derivative with respect to the quasi-momentum k can be eliminated by transforming to an accelerated basis for k [17] (by changing k to $k - \eta(t)$, where $\eta(t)$ is defined as $\eta(t) = (e/\hbar)\int_0^t F(t')\,\mathrm{d}t'$). For convenience, we introduce the following new notation:

$$\tilde{P}^{(1)}(k,t) = P^{(1)}(k-\eta(t),t) \tag{6}$$
$$\tilde{n}^{(2)}_{c,(v)}(k,t) = n^{(2)}_{c,(v)}(k-\eta(t),t) \tag{7}$$
$$\tilde{P}^{(3)}(k,t) = P^{(3)}(k-\eta(t),t). \tag{8}$$

In the accelerated basis, the three partial differential equations can be rewritten as the following coupled integro-differential equations:

$$\left[\frac{\partial}{\partial t} - \frac{\mathrm{i}}{\hbar}[\epsilon_c(k-\eta(t),t) - \epsilon_v(k-\eta(t),t) + V]\right]\tilde{P}^{(1)}(k,t)$$

$$= -\frac{\mathrm{i}}{\hbar}\left[\mu E(t) + V\sum_q \tilde{P}^{(1)}(q,t)\right] - \frac{\tilde{P}^{(1)}(k,t)}{T_2} \tag{9}$$

$$\frac{\partial \tilde{n}^{(2)}_{c,(v)}(k,t)}{\partial t} = \frac{\mp 2}{\hbar}\,\mathrm{Im}\left[\left(\mu E(t) + V\sum_q \tilde{P}^{(1)}(q,t)\right)\tilde{P}^{(1)*}(k,t)\right] - \frac{\tilde{n}^{(2)}_{c,(v)}(k,t)}{T_1} \tag{10}$$

$$\begin{aligned}\left[\frac{\partial}{\partial t} - \frac{\mathrm{i}}{\hbar}[\epsilon_c(k-\eta(t),t) - \epsilon_v(k-\eta(t),t) + V]\right]\tilde{P}^{(3)}(k,t) \\ = \frac{\mathrm{i}}{\hbar}[\tilde{n}^{(2)}_c(k,t) - \tilde{n}^{(2)}_v(k,t)]\left[\mu E(t) + V\sum_q \tilde{P}^{(1)}(q,t)\right] \\ - \frac{\mathrm{i}V}{\hbar}\sum_q[\tilde{n}^{(2)}_c(q,t) - \tilde{n}^{(2)}_v(q,t)]\tilde{P}^{(1)}(k,t) - \frac{\mathrm{i}V}{\hbar}\sum_q \tilde{P}^{(3)}(q,t) - \frac{\tilde{P}^{(3)}(k,t)}{T_2}.\end{aligned} \tag{11}$$

In these three coupled equations, we have introduced the intraband dephasing time T_1 and interband dephasing time T_2, respectively, to describe the incoherent process phenomenologically. However, it should be emphasized that the choice of T_1 and T_2 is not arbitrary. This important point has been made by Axt and co-workers recently, invoking the method of dynamics-controlled truncation (DCT) of the hierarchy of density matrices for optically excited ordered semiconductors. They found that the DCT approach is more effective than that using the SBE, both theoretically and experimentally, and that the SBE approach is correct only in the coherent limit [13]. In the following numerical calculation, we will set the interband dephasing time T_1 as 1.5 ps and the intraband dephasing time as $T_2 = 2T_1$. This choice fulfils the coherent-limit requirement [13]. Detailed studies on specific dephasing processes beyond the relaxation approximation, e.g. electron–phonon scattering processes, can be made by employing either the method of DCT [18] or Monte Carlo simulation [19].

The periodicity of both $P^{(3)}(k,t)$ and $\tilde{P}^{(3)}(k,t)$ in the quasi-momentum k leads to the following relation:

$$\int_{BZ} P^{(3)}(k',t)\,\mathrm{d}k' = \int_{BZ} \tilde{P}^{(3)}(k',t)\,\mathrm{d}k'.$$

The DFWM signals are proportional to $|P^{(3)}(t)|^2$ $(=|\int_{BZ}\tilde{P}^{(3)}(k',t)\ \mathrm{d}k'|^2)$, and hence we only need to solve the coupled integro-differential equations (9)–(11) directly in the accelerated basis. In our numerical simulation, we use the combined miniband width $\Delta = \Delta_c + \Delta_v = 20$ meV; the Coulomb potential strength V was set as 10 meV. Such a choice is believed to be appropriate to experimental practice [11, 15]. The central frequency of the optical field is assumed to be located 2 meV below the excitonic resonance [20]; the full width at half-maximum of the Gaussian laser pulse envelope $|E(t)|^2$ is chosen to be 100 fs.

The energy quantum of the ac field, $\hbar\omega$, is fixed as 20 meV, and the ratio $edF_1/\hbar\omega$ as 3.832, which is the first root of the Bessel function of the first order. This choice ($edF_1/\hbar\omega = 3.832$) is believed to be favourable for the appearance of fractional structures under the dc–ac field [21], and we *only* vary the Stark frequency ω_B, accordingly.

We show our results in figure 1, where we plot the fractional cases $\omega_B/\omega = \frac{1}{3}, \frac{2}{3}, \frac{4}{3}$ in the upper, middle, and lower panels respectively. From these panels, it can be clearly seen that the DFWM signals are periodic. The periods for these fractional cases are almost identical, approximately 0.8 picoseconds, and are around three times the ac field period $T_{ac} = 2\pi/\omega = 0.21$ ps. In all of these panels, we *only* change the Stark

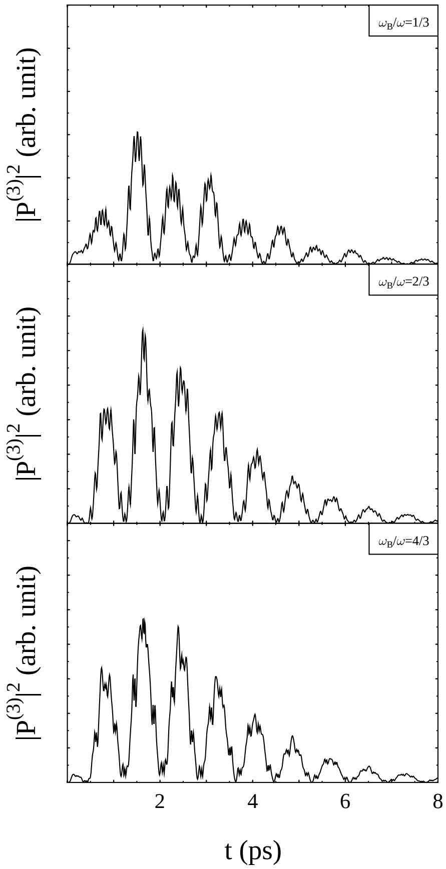

Figure 1. Time-resolved DFWM signals, showing the cases where $\omega_B/\omega = 1/3, 2/3, 4/3$ in the upper, middle, and lower panels, respectively.

frequency ω_B, and the ratios of the Stark frequency ω_B to the ac-field frequency ω span from $\frac{1}{3}$, which is less than unity, to $\frac{4}{3}$ which is greater than unity. All of these features can be explained by the following inference, which is the conclusion reached concerning optical absorption in reference [6]: when $\omega_B/\omega = p/q$, additional resonant peaks appear at $1/p, 2/p, \ldots, p - 1/p$ in the ω_B-range; that is to say, the period T_f for these new fractional peaks is p times that of usual Wannier–Stark ladders, $2\pi/\omega_B$. The ratio of the Stark frequency ω_B to the ac-field frequency ω is p/q; a simple arithmetic calculation leads to the assertion that T_f is q times the ac-field period $2\pi/\omega$ ($T_f = p \times 2\pi/\omega_B = p \times q/p \times 2\pi/\omega = q \times 2\pi/\omega$). We also calculated the DFWM signals for several other values of $\omega_B/\omega = p/q$ and confirmed this conclusion; we have not described this calculation here, to save space. Because all of the above findings are for the time domain rather than for the frequency (energy) domain as in references [6] and [7], we can conclude that fractional Wannier–Stark ladders can be obtained from the FWM signals.

Closer inspection of these panels makes it clear that these signals rise to maxima at around 1.5 picoseconds, and that the first small peaks in all of these panels are related to the duration of Gaussian laser pulses.

In summary, in the low-excitation regime, we perturbatively expand the SBEs involving the combined dc and ac fields up to the third order in the optical field E. By appropriately selecting the parameters—the ones that we choose are close to the experimental ones—and, particularly, by selecting the dephasing time T_1 and T_2 in such a way as to fulfil the coherent-limit requirement [13], we find that when the ratio of the Stark frequency ω_B of the dc field to the ac-field frequency ω is a fraction $\omega_B/\omega = p/q$, the periods T_f of the time-resolved DFWM signals are multiples of q times the period of the ac fields $T_{ac} = 2\pi/\omega$, even in the presence of excitonic interaction comparable to the miniband width. This fact is consistent with the optical absorption calculation for the frequency domain [6], and is believed to be a manifestation of fractional Wannier–Stark ladders, but in the time domain. In a more recent study on fractional Wannier–Stark ladders induced by dc–ac fields involving the use of the Boltzmann equation, it was found from the I–V characteristics and the differential conductivity that resonant peaks appear near the locations at which ω_B/ω is a fraction [21]. Both of these phenomena indicate the existence of fractional Wannier–Stark ladders. In fact, the introduction of next-nearest-neighbour interaction into the model will enhance the fractional Wannier–Stark ladders, especially for THz signals; this will be reported on elsewhere.

From an experimental point of view, the fractional Wannier–Stark ladder can be probed either by using a FWM experiment [8, 9, 11] in a SL driven by dc and a THz-range free-electron laser or by using an ultra-cold atom ensemble driven by a chopped optical potential to simulate a particle driven by external fields in spatially periodic structures, such as (and especially) crystalline solids [7].

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