Nonlinear optics of semiconductors under an intense terahertz field

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A theory for nonlinear optics of semiconductors in the presence of an intense terahertz electric field is constructed based on the double-line Feynman diagrams, in which the nonperturbative effect of the intense terahertz field is fully taken into account through using the Floquet states as propagating lines in the Feynman diagrams.

PACS numbers: 78.20.Bh, 42.65.An, 71.35.Cc

I. INTRODUCTION

Since early 1990’s, thanks mainly to the emergence of free electron lasers operating in the terahertz (THz) waveband\(^1\) the interaction between semiconductors and a strong THz field has been brought under intensive investigations. Nonlinear transport\(^2\) and linear optics of semiconductors under an intense THz field is desired. To this end, a good starting point is the eigen states of the THz field induced by Bloch oscillation in biased semiconductor superlattices\(^1\) and a theory based on Floquet states\(^1\) of time-periodic systems has been developed to consider the non-perturbative effects of the THz dipole field. Recently, the difference-frequency processes were proposed to generate THz emission, and the estimated strength of a strong THz field has been brought under intensive investigations (here we use the terminology of “linear optics” or “nonlinear optics” in the sense that the intense THz field is treated as a part of the system but not an optical excitation, otherwise, if the THz field is viewed as an external optical field, even the so-called “linear optics” here would be highly nonlinear). To thoroughly understand the physics in THz-field-driven semiconductors, as well as to develop novel devices based on these systems, nonlinear optical spectroscopies are a powerful and sometimes necessary method due to their accessibility both in ultrafast time-resolution and in multi-frequency mixing. For example, the four-wave mixing spectroscopy has been adopted to study the effect of the strong THz field induced by Bloch oscillation in biased semiconductor superlattices\(^1\) and a theory based on Floquet states\(^1\) of time-periodic systems has been developed to consider the non-perturbative effects of the THz dipole field. Recently, the difference-frequency processes were proposed to generate THz emission, and the estimated strength of a strong THz field could be of the order of kV/cm\(^1\), which is so large that the feedback effect of the THz field on the nonlinear difference-frequency process may be important. So, as a common theoretical basis, a nonlinear optical theory of semiconductors in the presence of an intense THz field is desired.

To construct such a theory, it is essential to include the non-perturbative driving of the THz field. To this end, a good starting point is the eigen states of the THz-driven systems, the Floquet states\(^1\) which have the non-perturbative effect of the THz field fully included. In fact, a compact theory for linear optics of THz-driven semiconductors has been formulated in the Floquet-state basis\(^2\). In next section, the general formalism for nonlinear optics of semiconductors under a strong THz field will be constructed with the double-line Feynman diagrams frequently used in textbooks\(^2\). In Section II some examples will be given to illustrate how to calculate the nonlinear optical susceptibility from the Feynman diagrams. And the conclusions are given in the last section.

II. GENERAL THEORY

The system to be considered is a semiconductor irradiated by an intense cw THz laser. The Hamiltonian of this system under excitation of additional weak lasers can be expressed as

\[
H = H_0(t) - \sum_p \hat{\mu} \cdot F_p(t),
\]

where \(H_0(t)\) is the unperturbated Hamiltonian of the semiconductor with the THz-field-driving included, \(\hat{\mu}\) is the dipole operator, and

\[
F_p(t, \mathbf{R}) = F_p e^{iK_p \cdot \mathbf{R} - i\Omega_p t} + c.c.
\]

is the pertubative optical field.

With the density matrix of the system denoted by \(\hat{\rho}(t)\), the optical polarization is \(P(t) = \text{Tr} [\hat{\rho}(t) \hat{\mu}]\). As the THz field, with photon energy much smaller than the band gap, induces no inter-band excitation, the system is assumed in the semiconductor ground state before optical excitation, i.e. \(\hat{\rho}(-\infty) = \langle 0 | 0 \rangle\). Thus the \(j\)th component of the \(N\)th order \(\chi^{(N)}\) nonlinear optical response to the optical fields is\(^1\):
\[ \cdots \times \left( \frac{i}{\hbar} \hat{\mu}_{j\mu} F_{j\mu}(t_N) U(t_N, t) \theta(t - t_N) \times \hat{\mu}_j \right) dt_1 dt_2 \cdots dt_N \]
\[ = \int_{-\infty}^{+\infty} \chi^{(N)}_{j_1j_2 \ldots j_N}(t; t_1, t_2, \ldots, j_N) F_{j_1}(t_1) F_{j_2}(t_2) \cdots F_{j_N}(t_N) dt_1 dt_2 \cdots dt_N, \] (3)

where the summation is over all permutations of the indices as indicated, and

\[ U(t, t') \equiv \hat{T} e^{-\frac{i}{\hbar} \int_{t'}^{t} H_0(t_1) dt_1} = U(t', t) \]

is the unperturbed propagator of the system. The system in the presence of a THz field is time-periodic, i.e. \( H_0(t) = H_0(t + T), \) where \( T \equiv 2\pi/\omega \) with \( \omega \) denoting the angular frequency of the THz field. The eigen states of the time-periodic Hamiltonian is the Floquet states \( \{q, t\} \), which are time-periodic and satisfy the secular equation

\[ [H_0(t) - i\hbar \partial_t] |q, t\rangle = E_q |q, t\rangle = E_q |q, t + T\rangle, \] (4)

where \( E_q \) is the quasi-energy. Obviously, the sidebands of the Floquet states \( |q, m, t\rangle \equiv \exp(i m \omega t) |q, t\rangle \) are also eigen states of Eq. (4) with quasi-energy \( E_{q,m} \equiv E_q + m \hbar \omega \).

Now the propagator can be expanded into the Floquet states as

\[ U(t, t') = |q, t\rangle \langle q, t'| e^{-\frac{i}{\hbar} \int_{t'}^{t} E_q(t - t') dt}, \] (5)

(hereafter all superscripts and subscripts appearing only on the righthand side of an equation are assumed dumb indices to be summed over). The dipole matrix element between the Floquet states is

\[ \mu_{q,q'}(t) = \langle q, t| \hat{\mu}| q', t \rangle = e^{i m \omega t} \mu_{q,m,q'}, \] (6)

where

\[ \mu_{q,m,q'} \equiv T^{-1} \int_{0}^{T} \langle q, m, t| \hat{\mu}| q', t \rangle dt = \mu_{q,q',-m} \]

is the time-average of the dipole matrix element. Thus we have

\[ U(t', t) \hat{\mu} = |q', m', t'| e^{-\frac{i}{\hbar} E_q,m',m'(t' - t)} \mu_{q',m',q'}(q, t), \] (7)
\[ \hat{\mu} U(t, t') = |q, t\rangle \mu_{q,q',m'} e^{-\frac{i}{\hbar} E_q,m'(t' - t)} \langle q', m', t'|. \] (8)

With Eq. (4) and (5) used respectively on the left and right sides of \(|0\rangle\langle 0| \) in Eq. (3), the nonlinear optical response can be derived. The double-line Feynman diagrams can be used to assist such a derivation as well as in static cases. For this particular time-periodic system, the rules of the Feynman diagrams for the optical response \( F_q^{(N)}(t) \) are [see Fig. (a)]:

1. The evolution of the diagram starts from the ground state \(|0\rangle\langle 0| \) at \( t = -\infty \) and ends at \( t \) with the final Floquet-state density matrix \(|q, n\rangle\langle q, n + m| \).

2. The \( N \) interaction vertices at \( t_p < t \) (\( p = 1, 2, \ldots, N \)) consist of a photon line (the dotted arrow) with frequency \( \Omega_p \), the Floquet state before interaction \(|a, m_a\rangle \) on the left branch or \(|a, m_a\rangle \) on the right branch, and the state after interaction \(|c, m_c\rangle \) on the left branch or \(|c, m_c\rangle \) on the right branch. The photon lines pointing to inside and outside represent photon absorption and emission processes, respectively. The \( p \)th vertex contributes a dipole matrix element \( \frac{i}{\hbar} \mu_{c,m_a,a,m_a} \) or \( -\frac{i}{\hbar} \mu_{a,m_a,c,m_c} \) if it’s on the left (or right) branch, and a factor from the optical field \( F_p \exp(-i \Omega_p t_p + i \mathbf{K}_p \cdot \mathbf{R}) \) for photon absorption or \( F_p \exp(i \Omega_p t_p - \mathbf{K}_p \cdot \mathbf{R}) \) for photon emission.
from the rules above:

1. The interaction vertex, constituted by the photon line with frequency \( \Omega_p \), the initial state \( |a, m_a \rangle \) (or \( \langle a, m_a | \) ), and the final state \( |c, m_c \rangle \) (or \( \langle c, m_c | \) ), contributes the factor \( \pm \langle \mu_j \rangle_{c,m,c,q,n-m} e^{-im\omega t} \) where \( \langle c, m_c | \) is the state before photon emission.

2. The factor associated with the double-line state \( |b, m_b \rangle |c, m_c \rangle \) between \( t_1 \) and \( t_2 \) \((t_1 < t_2)\) represents the unperturbed propagator of the density matrix. If the state associated with the double-line is \( |b, m_b \rangle \langle c, m_c | \), the factor due to the propagator is

\[
\theta(t_2 - t_1) e^{-\frac{\hbar}{4} (E_{b,m_b} - E_{c,m_c}) (t_2 - t_1)},
\]

where \( E_{b,m_b} \) and \( E_{c,m_c} \) is the transition energy between the Floquet states, and \( \Gamma_{b,m_b,c,m_c} / \hbar \) is the relaxation rate of the density matrix due to the interaction with environment.

3. The photon emission vertex at final time \( t \) contributes the factor \( (\langle \mu_j \rangle_{c,m,c,q,n+m} e^{-im\omega t} \) where \( \langle c, m_c | \) is the state before photon emission.

4. The double-line between two neighbor vertices at \( t_1 \) and \( t_2 \) \((t_1 < t_2)\) represents the unperturbed propagator of the density matrix. If the state associated with the double-line is \( |b, m_b \rangle |c, m_c \rangle \), the factor due to the propagator is

\[
\theta(t_2 - t_1) e^{-\frac{\hbar}{4} (E_{b,m_b} - E_{c,m_c}) (t_2 - t_1)},
\]

where \( E_{b,m_b} \) and \( E_{c,m_c} \) is the transition energy between the Floquet states, and \( \Gamma_{b,m_b,c,m_c} / \hbar \) is the relaxation rate of the density matrix due to the interaction with environment.

5. For each diagram, all factors described above should be multiplied together and all Floquet states and their sidebands are summed over. Then all different diagrams, which are determined both by how the \( N \) vertices are grouped into two branches and by the time-ordering, should be summed. For a certain optical configuration (which determines each vertex is a photon absorption or emission process), there are totally \( 2^N N! \) different diagrams, among which there are \( 2^N \) elementary diagrams and the others can be derived from them by permutation of the \( N \) vertices. And still, there are \( 2^N \) different optical configurations. In a certain experiment, only one optical configuration needs to be considered.

The nonlinear optical susceptibility in frequency domain is defined as

\[
\chi^{(N)}_{j_1,j_2,\ldots,j_N}(\Omega, \Omega_1, \Omega_2, \ldots, \Omega_N) = \int dt_1 dt_2 \cdots dt_N \chi^{(N)}_{j_1,j_2,\ldots,j_N} (t; t_1, t_2, \ldots, t_N) e^{i\Omega t - i\Omega_1 t_1 - i\Omega_2 t_2 - \cdots - i\Omega_N t_N}. \tag{9}
\]

From the rules described above, we can see that the nonlinear optical response of THz-field-driven semiconductors, or generally speaking, time-periodic systems, takes the form very similar to the textbook formalism for static systems.\(^{18,19} \)

\[ \text{The difference lies on three aspects:} \]

First, the dipole matrix element here is the time-average element and transition energy. The THz field has been fully included through the renormalization of dipole matrix element and transition energy.

As an illustrative example, the Feynman diagrams for linear optics are plotted in Fig. 1(b) and (c). From the rules for the Feynman diagrams, the linear susceptibility can be formulated as
where $|X, n\rangle$ denotes the THz-photon sidebands of the Floquet-state excitons and $|0, n\rangle$ denotes the sidebands of the ground state. This result is identical to that in Ref. 3 derived with non-equilibrium Green’s function technique.

Now we consider another example, the $\chi^{(3)}$ four-wave mixing, in which three input beams propagate in the directions $K_1$, $K_2$, and $K_3$, respectively, and the signal is detected in the direction $K_1 + K_2 - K_3$. Corresponding to this optical configuration, there are 48 different Feynman diagrams, among which only 16 are under resonant excitation condition. We calculate two typical diagrams as examples of resonant excitation of excitons and bi-excitons (exciton molecules constituted by two excitons). Fig. 1(d) is a Feynman diagram for resonant excitation of excitons, which contributes to the susceptibility as

$$\chi^{(1)}_{j;i}(\Omega; \Omega_1) = 2\pi\delta(\Omega - \Omega_1 - m\omega) \frac{(\mu_j)_0;X,n+m (\mu_{j_1})_{X,n,0}}{E_{X,n,0} + i\Gamma_{X,n,0} - \Omega_1} - \frac{(\mu_j)_{X,n,0+m} (\mu_{j_1})_{0;X,n}}{E_{0;X,n} + i\Gamma_{0;X,n} - \Omega_1},$$

(10)

and Fig. 1(c) is a diagram for resonant excitation of bi-excitons, which contributes to the susceptibility as

$$-2\pi\delta(\Omega - \Omega_1 - \Omega_2 + \Omega_3 - m\omega) \frac{(\mu_{j_3})_{0;X_2,n_2} (\mu_{j_2})_{X_2,n_2;0,n} (\mu_j)_{0;n;X_1,n_1+m} (\mu_{j_1})_{X_1,n_1;0}}{(E_{X_1,n_1,0,n} - i\Gamma_{X_1,n_1,0,n} - \Omega_1 - \Omega_2 + \Omega_3) (E_{X_2,n_2,0,n} - i\Gamma_{X_2,n_2,0,n} + \Omega_3)}.$$

where $|B, n\rangle$ denotes the sidebands of Floquet-state bi-excitons. From the two terms above, we can easily identify the resonances associated with the THz-photon-assisted Floquet-state exciton and bi-exciton transitions. The sideband generation, as indicated by the $\delta$-function with argument containing an integer multiple of THz-photon energy, accounts for the four-wave mixing signal out of the excitation spectrum observed in the numerical calculations in Ref. 11.

#### IV. SUMMARY

In summary, based on the double-line Feynman diagrams similar to the static case in textbooks, we have construct a general theory for nonlinear optics of semiconductors under an intense THz field. The basis of the Feynman diagram is the eigen states of the time-periodic systems, i.e., the Floquet states, so the non-perturbative effect of the THz field has been fully taken into account. Many phenomena, including the THz-photon-assisted exciton or bi-exciton resonances and the THz-photon sideband generation, are naturally accounted for in this theory.

**Acknowledgments**

This work was supported by the National Science Foundation of China.

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