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.

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I. INTRODUCTION

Since the early 1990s, thanks mainly to the emergence of free electron lasers operating in the terahertz (THz) wave band,[1] the interaction between semiconductors and a strong THz field has been brought under intensive investigations. Nonlinear transport[2–5] and linear optics[6–12] are the two main themes of these 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 multifrequency 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,[13] and a theory based on Floquet states[14] of time-periodic systems has been developed to consider the nonperturbative effects of the THz dipole field.[15] Recently, the difference-frequency processes were proposed to generate THz emission, and the estimated strength of THz field could be of the order of kV/cm,[16,17] 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 nonperturbative driving of the THz field. To this end, a good starting point is the eigenstates of the THz-driven systems, the Floquet states,[14] which have the nonperturbative 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.[8] In the 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.[18,19] In Sec. III, some examples will be given to illustrate how to calculate the nonlinear optical susceptibility from the Feynman diagrams. 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

$$H=H_0(t)-\sum_p \hat{\boldsymbol{\mu}}\cdot\mathbf{F}_p(t), \tag{1}$$

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

$$\mathbf{F}_p(t,\mathbf{R})=\mathbf{F}_p e^{i\mathbf{K}_p\cdot\mathbf{R}-i\Omega_p t}+\text{c.c.} \tag{2}$$

is the perturbative optical field.

With the density matrix of the system denoted by $\hat{\rho}(t)$, the optical polarization is $\mathbf{P}(t)=\mathrm{Tr}[\hat{\rho}(t)\hat{\boldsymbol{\mu}}]$. As the THz field, with photon energy much smaller than the band gap, induces no interband excitation, the system is assumed in the semiconductor ground state before optical excitation, i.e., $\hat{\rho}(-\infty)=|0\rangle\langle 0|$. Thus the jth component of the Nth order $[\chi^{(N)}]$ nonlinear optical response to the optical fields is[18,19]

$$\begin{aligned}
P_j^{(N)}(t)=&\sum_{\mathrm{P}\{j_1,j_2,\ldots,j_N,j\}}\int_{-\infty}^{+\infty}\mathrm{Tr}\Big[U(t,t_n)\,\theta(t-t_n)\frac{i}{\hbar}\hat{\mu}_{j_n}F_{j_n}(t_n)\times U(t_n,t_{n-1})\,\theta(t_n-t_{n-1})\frac{i}{\hbar}\hat{\mu}_{j_{n-1}}F_{j_{n-1}}(t_{n-1})\cdots U(t_2,t_1)\\
&\times\theta(t_2-t_1)\frac{i}{\hbar}\hat{\mu}_{j_1}F_{j_1}(t_1)|0\rangle\langle 0|\left(-\frac{i}{\hbar}\right)\hat{\mu}_{j_{n+1}}F_{j_{n+1}}(t_{n+1})U(t_{n+1},t_{n+2})\,\theta(t_{n+2}-t_{n+1})\cdots\left(-\frac{i}{\hbar}\right)\\
&\times\hat{\mu}_{j_N}F_{j_N}(t_N)U(t_N,t)\,\theta(t-t_N)\times\hat{\mu}_j\Big]dt_1dt_2\cdots dt_N\\
\equiv&\int_{-\infty}^{+\infty}\chi^{(N)}_{j;j_1,j_2,\ldots,j_N}(t;t_1,t_2,\ldots,j_N)F_{j_1}(t_1)F_{j_2}(t_2)\cdots F_{j_N}(t_N)dt_1dt_2\cdots dt_N,
\end{aligned} \tag{3}$$

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

$$U(t,t')\equiv\hat{\mathrm{T}}e^{-i/\hbar\int_{t'}^{t}H_0(t_1)dt_1}=U^{\dagger}(t',t)$$

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 ω denoting the angular frequency of the THz field. The eigenstates of the time-periodic Hamiltonian is the Floquet states $\{|q,t\rangle\}$, which are time periodic and satisfy the secular equation

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

where E_q, in analogy of quasimomentum of Bloch states, is termed quasienergy. Obviously, the sidebands of the Floquet states $|q,m,t\rangle\equiv\exp(im\omega t)|q,t\rangle$ are also eigenstates of Eq. (4) with quasienergy $E_{q,m}\equiv E_q+m\hbar\omega$. Now the propagator can be expanded into the Floquet states as

$$U(t,t')=|q,t\rangle e^{-(i/\hbar)E_q(t-t')}\langle q,t'|, \qquad (5)$$

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

$$\boldsymbol{\mu}_{q;q'}(t)=\langle q,t|\hat{\boldsymbol{\mu}}|q',t\rangle=e^{im\omega t}\boldsymbol{\mu}_{q,m;q'}, \qquad (6)$$

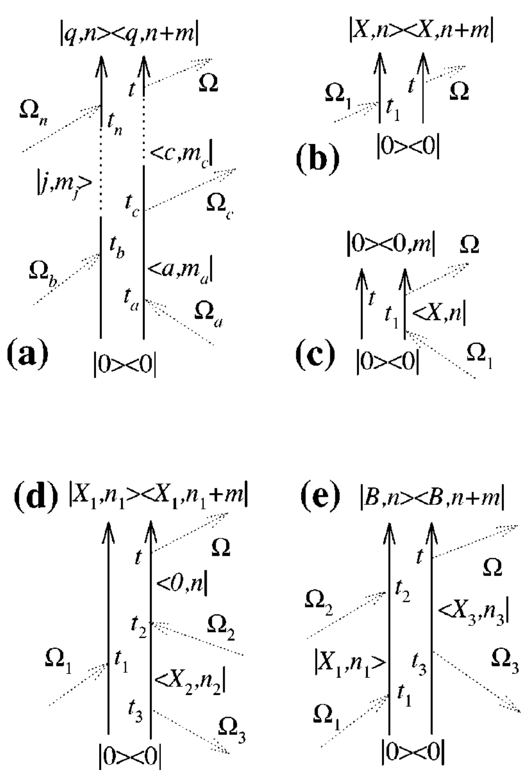

FIG. 1. (a) The general form of double-line Feynman diagrams for nonlinear optics of semicondcutors in the presence of an intense THz electric field. (b) and (c) The double-line Feynman diagrams for linear optics of THz-field-driven semiconductors. (d) and (e) Two of the double-line Feynman diagrams for $\chi^{(3)}$ four-wave mixing in THz-field-driven semiconductors.

where

$$\boldsymbol{\mu}_{q,m;q'}\equiv T^{-1}\int_0^T\langle q,m,t|\hat{\boldsymbol{\mu}}|q',t\rangle dt=\boldsymbol{\mu}_{q;q',-m}$$

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

$$U(t',t)\hat{\boldsymbol{\mu}}=|q',m',t'\rangle e^{-i/\hbar E_{q',m'}(t'-t)}\boldsymbol{\mu}_{q',m';q}\langle q,t|, \quad (7)$$

$$\hat{\boldsymbol{\mu}}U(t,t')=|q,t\rangle\boldsymbol{\mu}_{q;q',m'}e^{-i/\hbar E_{q',m'}(t-t')}\langle q',m',t'|. \quad (8)$$

With Eqs. (7) and (8) 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 $P_j^{(N)}(t)$ are [see Fig. 1 (a)]:

(i) 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 $|q,n\rangle\langle q,n+m|$.

(ii) The N interaction vertices at $t_p<t$ $(p=1,2,\ldots,N)$ consist of a photon line (the dotted arrow) with frequency Ω_p, the Floquet state before interaction ($|a,m_a\rangle$ on the left branch or $\langle a,m_a|$ on the right branch), and the state after interaction ($|c,m_c\rangle$ on the left branch or $\langle c,m_c|$ on the right branch). The photon lines pointing to inside and outside represent photon absorption and emission processes, respectively. The pth vertex contributes a dipole matrix element $(i/\hbar)\boldsymbol{\mu}_{c,m_c;a,m_a}$ [or $-(i/\hbar)\boldsymbol{\mu}_{a,m_a;c,m_c}$] if it is on the left (or right) branch, and a factor from the optical field $\mathbf{F}_p\exp(-i\Omega_pt_p+i\mathbf{K}_p\cdot\mathbf{R})$ for photon absorption or $\mathbf{F}_p^{*}\exp(i\Omega_pt_p-i\mathbf{K}_p\cdot\mathbf{R})$ for photon emission.

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

(iv) 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 $|b,m_b\rangle\langle c,m_c|$, the factor due to the propagator is

$$\theta(t_2-t_1)e^{-(i/\hbar)(E_{b,m_b;c,m_c}-i\Gamma_{b,m_b;c,m_c})(t_2-t_1)},$$

where $E_{b,m_b;c,m_c}\equiv E_{b,m_b}-E_{c,m_c}$ 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.

(v) 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 $2^NN!$ 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

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

The rules for constructing susceptibility in frequency domain from the Feynman diagrams can be easily derived from the rules above:

(i) The interaction vertex, consitituted by the photon line with frequency Ω_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 $(i/\hbar)(\mu_{j_p})_{c,m_c;a,m_a}$ [or $-(i/\hbar)(\mu_{j_p})_{a,m_a;c,m_c}$] if the vertex is on the left (or right) branch.

(ii) The factor associated with the double-line state $|b,m_b\rangle\langle c,m_c|$ between t_1 and t_2 ($t_1<t_2$) is

$$-i\hbar\left(E_{b,m_b;c,m_c} - i\Gamma_{b,m_b;c,m_c} - \sum_{t_n<t_2} \Omega_n\right)^{-1}.$$

(iii) The factor associated with the final photon emission vertex at t is

$$2\pi\delta\left(\Omega - m\omega - \sum_{n=1}^{N} \Omega_n\right)(\mu_j)_{c,m_c;q,n+m},$$

where $\langle c,m_c|$ is the state before photon emission.

(iv) All factors are multiplied together and all intermediate Floquet states and their sidebands are summed over. Then all possible diagrams are summed. For a certain optical configuration, the frequency Ω_p is positive or negative depending on whether the corresponding vertex is an absorption or emission process.

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] The difference lies on three aspects: First, the dipole matrix element here is the time average of that between Floquet states. Second, the THz field induces new resonances at THz-photon-assisted transitions, as the sidebands of the Floquet states act as intermediate states in the nonlinear optical process. And third, there is an extra dynamic phase factor $e^{-im\omega t}$ at the final photon-emission vertex, which makes the energy of the emitted photon differ from the total input energy by an integer multiple of the THz-photon energy, corresponding to the physical process of THz-photon-sideband generation.[9,10] In the time-periodic systems, the energy-conserving condition is relaxed to the quasienergy conservation. We reiterate that the nonperturbative effect of the THz field has been fully included through the renormalization of dipole matrix element and transition energy.

III. EXAMPLES

As an illustrative example, the Feynman diagrams for linear optics are plotted in Figs. 1(b) and 1(c). From the rules for the Feynman diagrams, the linear susceptibility can be formulated as

$$\chi^{(1)}_{j;j_1}(\Omega;\Omega_1) = 2\pi\delta(\Omega - \Omega_1 - m\omega) \times \left[\frac{(\mu_j)_{0;X,n+m}(\mu_{j_1})_{X,n;0}}{E_{X,n;0} - i\Gamma_{X,n;0} - \Omega_1} - \frac{(\mu_j)_{X,n;0,m}(\mu_{j_1})_{0;X,n}}{E_{0;X,n} - i\Gamma_{0;X,n} - \Omega_1}\right], \tag{10}$$

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. 8 derived with nonequilibrium Green's-function technique.

Now we consider another example, the $\chi^{(3)}$ four-wave mixing, in which three input beams propagate in the directions $\mathbf{K}_1$, $\mathbf{K}_2$, and $\mathbf{K}_3$, respectively, and the signal is detected in the direction $\mathbf{K}_1+\mathbf{K}_2-\mathbf{K}_3$. 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). Figure 1(d) is a Feynman diagram for resonant excitation of excitons, which contributes to the susceptibility as

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

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

$$\frac{-2\pi\delta(\Omega-\Omega_1-\Omega_2+\Omega_3-m\omega)(\mu_{j_3})_{0;X_3,n_3}(\mu_j)_{X_3,n_3;B,n+m}(\mu_{j_2})_{B,n;X_1,n_1}(\mu_{j_1})_{X_1,n_1;0}}{(E_{B,n;X_3,n_3}-i\Gamma_{B,n;X_3,n_3}-\Omega_1-\Omega_2+\Omega_3)(E_{X_1,n_1;X_3,n_3}-i\Gamma_{X_1,n_1;X_3,n_3}-\Omega_1+\Omega_3)(E_{X_1,n_1;0}-i\Gamma_{X_1,n_1;0}-\Omega_1)},$$

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 side-band generation, as indicated by the δ 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. 15.

IV. SUMMARY

In summary, based on the double-line Feynman diagrams similar to the static case in textbooks, we have constructed a general theory for nonlinear optics of semiconductors under an intense THz field. The basis of the Feynman diagram is the eigenstates of the time-periodic systems, i.e., the Floquet states, so the nonperturbative 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.

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