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C. Zhang
University of Wollongong, czhang@uow.edu.au

J. C. Cao
Chinese Academy of Sciences, Shanghai, China

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Calculation of the nonlinear free-carrier absorption of terahertz radiation in semiconductor heterostructures

C. Zhang[1] and J. C. Cao[2]
[1]*School of Engineering Physics, University of Wollongong, New South Wales 2522, Australia*
[2]*State Key Laboratory of Functional Materials for Informatics, Shanghai Institute of Microsystem and Information Technology, Chinese Academy of Sciences, Shanghai, China*
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Nonlinear absorption of terahertz waves by electrons in a semiconductor heterostructure is calculated. We solve the quantum transport equation for electrons strongly coupled to terahertz photons. The electrical field of the laser radiation is included exactly, and the electron-impurity interaction is included up to the second order. It is found that Joule heating of the electronic system due to impurity scattering decreases rapidly due to the strong electron-photon interaction. Our result is the dynamic equivalence of electron localization in a strong field. In the limit of weak radiation field, the current is linear in the field strength.

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Electromagnetic radiation in the frequency range of terahertz (THz) plays an important role in probing and studying the optical and transport properties of low-dimensional semiconductor systems. Because of the nearly resonant match between the photon energy (of order of THz) and all characteristic energies of the electronic system such as Fermi energy, cyclotron energy, subband energy, etc. (all of order of meV or a few THz), various resonant phenomena and nonlinear properties in the electronic systems can be investigated. THz lasers have been applied to experimental investigation of nonlinear transport and optical properties in electron gases such as those in semiconductor quantum wells and heterostructures.[1–8] The discovery of a new magnetoresistance oscillation and zero-resistance state in high-mobility electronic systems[9–11] has stimulated new theoretical interest in the transport properties of electrons under electromagnetic radiation.[12–15] A satisfactory understanding of the observed dc magnetoresistance oscillation has been achieved.[12,14,15] There are three different theoretical methods of calculating the electrical transport under a radiation. The first is the balance equation approach[12] which calculates the resistivity directly. The second approach[14,15] employs the Green's function formalism and Kubo formula to calculate the conductivity or resistivity. We have developed a quantum transport equation method[16] to study the frequency-dependent phenomena.

All existing theoretical work deal with the electrical current response of a strongly coupled electron-photon system to a weak ac or dc force. In these works, the terahertz field ($\mathbf{E}_\gamma$) is strongly coupled to the electrons and the coupled system is responding in a linear fashion to another weak potential (or probing field $\mathbf{E}$). In a recent paper,[16] we calculated the linear response of a coupled electron-photon system to a weak ac probing field, $\mathbf{j}(\omega,\Omega)=\sigma(\omega,\Omega)\mathbf{E}$, where ω is the radiation frequency and Ω is the probing frequency. It is found that the resonant absorption of the probing field can be achieved when the probing frequency is in the vicinity of the frequency of $\mathbf{E}_\gamma$. In Ref. 16, we used the terahertz field $\mathbf{E}_\gamma$ to tune the absorption of $\mathbf{E}$ or $\mathbf{j}(\omega,\Omega)$. However, we did not calculate the direct absorption of the terahertz field $\mathbf{E}_\gamma$ by the electrons. The absorption of $\mathbf{E}_\gamma$ is much stronger than the absorption of $\mathbf{E}$. The absorption of $\mathbf{E}$ goes to zero as $\mathbf{E}$ goes to zero. However, in the absence of the probing field $\mathbf{E}$, there is still a strong absorption of $\mathbf{E}_\gamma$. To calculate the absorption of the THz radiation, electrical current in the absence of any probing field must be calculated. Such a current cannot be obtained from the previous theory[12–16] because the current obtained there vanishes when the probing field is removed. Furthermore, in the limit of zero radiation field, all existing theories predicted a quadratic dependence of the current on the radiation field while the experiment[11] clearly shows a linear dependence.

In this paper, we study the nonlinear absorption of an intense THz electrical field by electrons. We calculate the electrical current driven directly by the THz radiation field (i.e., without any probing field) to the second order of the electron-impurity interaction. It is found that photon absorption due to impurity scattering decreases very rapidly at high THz field, indicating a suppression of electron-impurity scattering by intense photon radiation. Our result of the electrical current is linear in the radiation field as $\mathbf{E}_\gamma$ approaches zero.

Our model system is a two-dimensional electron gas under an intense laser radiation. We choose the laser field to be along the x direction, $\mathbf{E}_\gamma(t)=E_0\cos(\omega t)\mathbf{e_x}$, where E_0 and ω are the amplitude and frequency of the laser field. For the notational convenience, both $\hbar$ and the speed of light c have been set to unity. Let us choose the vector potential for the laser field to be in the form

$$\mathbf{A}_\gamma=(E_0/\omega)\sin(\omega t)\mathbf{e_x}. \tag{1}$$

The time-dependent Schrödinger equation for a single electron is given as

$$i\frac{\partial}{\partial t}\psi(\mathbf{r},t)=H\psi(\mathbf{r},t)=\frac{(\mathbf{p}-e\mathbf{A}_\gamma)^2}{2m^*}\psi(\mathbf{r},t). \tag{2}$$

The time-dependent wave function can be written as

$$\psi_{\mathbf{k}}(\mathbf{r},t) = \exp(-i2\gamma_1\omega t)\exp\{i\gamma_0 k_x[1-\cos(\omega t)]\} \times\exp[i\gamma_1 \sin(2\omega t)]\exp(-i\epsilon_k t)\exp(i\mathbf{k}\cdot\mathbf{r}), \quad (3)$$

where $\gamma_0=(eE_0)/m^*\omega^2$ and $\gamma_1=(eE_0)^2/(8m^*\omega^3)$. The effect of electrical field is included in this wave function exactly.

Electrical current of the system driven by the terahertz laser due to electron–random-impurity scattering can now be calculated. The Hamiltonian of the system can be written as

$$H = H_0 + H_{ee} + H_{eI}, \quad (4)$$

where H_0 is the Hamiltonian of a noninteracting many-electron system,

$$H_0 = \frac{1}{2m^*}\sum_{\mathbf{p}} [\mathbf{p}+e\mathbf{A}_\gamma]^2 b_{\mathbf{p}}^\dagger(t) b_{\mathbf{p}}(t) \quad (5)$$

where

$$b_{\mathbf{k}}(t) = b_{\mathbf{k}}\exp(-i2\gamma_1\omega t)\exp\{i\gamma_0 k_x[1-\cos(\omega t)]\} \times\exp[i\gamma_1\sin(2\omega t)]$$

and $b_{\mathbf{k}}^\dagger(b_{\mathbf{k}})$ is the creation (annihilation) operator for an electron with momentum $\mathbf{k}$ in the absence of THz photon radiation. The term H_{ee} is the electron-electron interaction

$$H_{ee} = \frac{1}{2}\sum_{\mathbf{p},\mathbf{p}',\mathbf{q}} V_q b_{\mathbf{p}+\mathbf{q}}^\dagger(t) b_{\mathbf{p}'-\mathbf{q}}^\dagger(t) b_{\mathbf{p}'}(t) b_{\mathbf{p}}(t), \quad (6)$$

where $V_q=2\pi e^2/q$ is the Fourier transform of the electron-electron interaction in two dimensions. H_{eI} is the interaction between the electrons and random impurities:

$$H_{eI} = -\sum_{\mathbf{p},\mathbf{q}} V_q b_{\mathbf{p}+\mathbf{q}}^\dagger(t) b_{\mathbf{p}}(t)\sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i}, \quad (7)$$

where $\mathbf{R}_i$ is the position of the ith impurity which is assumed to be singly charged. Now the total average two-dimensional (2D) current density of the system is defined as

$$\mathbf{j} = \left\langle \frac{\delta H}{\delta \mathbf{A}_\gamma}\right\rangle = \frac{e}{m^*}\sum_{\mathbf{p}}\langle[\mathbf{p}+e\mathbf{A}_\gamma] b_{\mathbf{p}}^\dagger(t) b_{\mathbf{p}}(t)\rangle. \quad (8)$$

The current given in Eq. (8) is directly driven by the radiation field; i.e., it is not a linear response to any probing field. The method developed in Ref. 16 can be used to obtain nonlinear current for the present system. There are two basic equations to describe the quantum transport of the system. The first equation is the equation of motion for the single-electron density matrix $F(\mathbf{p},\mathbf{p}+\mathbf{k})=\langle b_{\mathbf{p}}^\dagger(t) b_{\mathbf{p}+\mathbf{k}}(t)\rangle$,

$$i\frac{\partial}{\partial t}F(\mathbf{p},\mathbf{p}+\mathbf{k}) = [\epsilon_{\mathbf{p}+\mathbf{k}} - \epsilon_{\mathbf{p}} + k_x\gamma_0\omega\sin(\omega t)]F(\mathbf{p},\mathbf{p}+\mathbf{k}) + \sum_{\mathbf{q}} V_q\left[n(\mathbf{q},t) - \sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i}\right][F(\mathbf{p},\mathbf{p}+\mathbf{k}-\mathbf{q}) - F(\mathbf{p}+\mathbf{q},\mathbf{p}+\mathbf{k})]. \quad (9)$$

Here $\epsilon_{\mathbf{p}}=p^2/2m$ is the kinetic energy of an electron having momentum $\mathbf{p}$ and

$$n(\mathbf{q},t) = \sum_{\mathbf{p}} F(\mathbf{p},\mathbf{p}+\mathbf{k}). \quad (10)$$

The second equation describes time-dependent current density $\mathbf{j}_1(t)$:

$$i\frac{d\mathbf{j}_1(t)}{dt} = \frac{-e}{m^*}\sum_{\mathbf{q}} V_q \mathbf{q} n(-\mathbf{q},t)\sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i}. \quad (11)$$

The key difference between this work and Ref. 16 is that the present work does not seek a linear current response to a probing field. Instead, we solve the exact current density driven by the radiation field. The solution of Eqs. (9) and (11), up to the second order in electron-impurity scattering, can be written as

$$\mathbf{j}_1(\omega) = \frac{-ne}{m^*\omega}\sum_{\mathbf{q}} \mathbf{q} V_q \sum_{m\neq 0}\frac{(-i)^m}{m} J_m(q_x\gamma_0) e^{iq_x\gamma_0}\frac{V_q Q(q,m\omega)}{D(q,m\omega)}, \quad (12)$$

where

$$D(q,m\omega) = 1 - V_q Q(q,m\omega) \quad (13)$$

is the dielectric function in the random-phase approximation and

$$Q(q,\omega) = \int \frac{d\mathbf{p}}{(2\pi)^2}\frac{f_{\mathbf{p}+\mathbf{q}} - f_{\mathbf{p}}}{\epsilon_{\mathbf{p}+\mathbf{q}} - \epsilon_{\mathbf{p}} - \omega} \quad (14)$$

is the polarizability for free electrons. Here, f_p is the Fermi-Dirac distribution and $J_m(x)$ is the Bessel function of the first kind. The terms with different m represent various photon emission and absorption processes.

Here it is necessary to point out that the appearance of the equilibrium distribution f_p in the final result does not mean that the original nonequilibrium density matrix $F(\mathbf{p},\mathbf{p}+\mathbf{k})$ $=\langle b_{\mathbf{p}}^\dagger(t) b_{\mathbf{p}+\mathbf{k}}(t)\rangle$ has been treated in an equilibrium fashion. Our theory involves decomposing the nonequilibrium density matrix into successive density matrices for photon side bands. Such a density matrix for the mth photon sideband is written in the form of the $F_0^{(m)}R^{(m)}(t)$ where $F_0^{(m)}$ is the density matrix in the absence of radiation field.[16] Because of the different time dependences of $R^{(m)}$ for different photon sidebands, $F_0^{(m)}R^{(m)}(t)$ is still a nonequilibrium quantity and has to be determined by the equation of motion including the electron-impurity scattering. Such an equation of motion is then solved to the lowest order in electron-impurity scattering. In the equation of motion, for those F_0 terms multiplied by a term proportional to impurity potential, only a zeroth-order solution is required. Our choice of a zeroth-order solution is the Fermi-Dirac distribution function.

For isotropic systems, the electric current is along the direction of polarization of the laser field. The real part of the electric current is given as

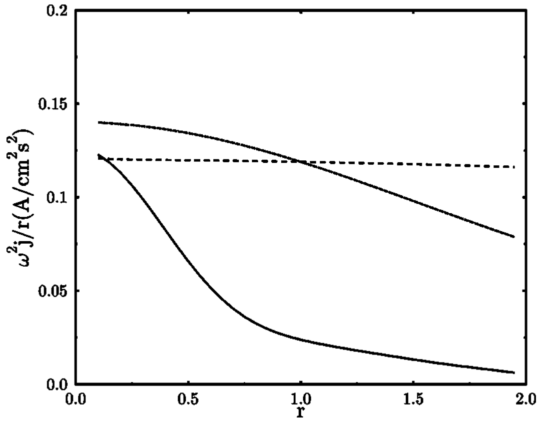

FIG. 1. Plot of $R=\omega^2\,\mathrm{Re}(j)/r$ versus the reduced electric field r for several different frequencies: the solid line is for $\omega=1$ Thz, the dotted line is for $\omega=2$ Thz, and the dashed line is for $\omega=5$ Thz.

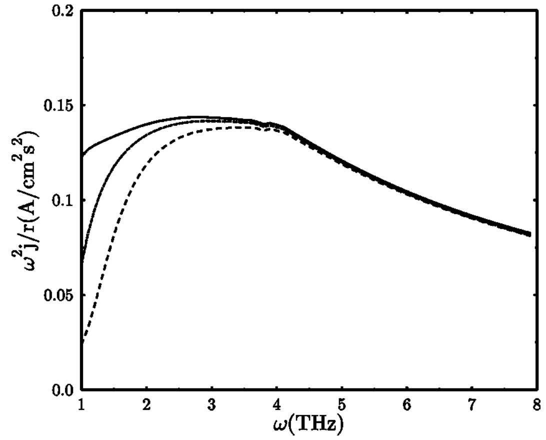

FIG. 2. Plot of $R=\omega^2\,\mathrm{Re}(j)/r$ versus the laser frequency. The solid line is for $r=0.1$, the dotted line is for $r=0.5$, and the dashed line is for $r=1.0$.

$$\mathrm{Re}[\mathbf{j}_{1x}(\omega)] = -\frac{ne}{m^*\omega}\sum_{\mathbf{q}} q_x V_q \sum_{m\neq 0}\frac{J_m(q_x\gamma_0)}{m} \times\left\{\mathrm{Im}\left[\frac{1}{D(q,m\omega)}\right]\sin(q_x\gamma_0 - m\pi/2) - \mathrm{Re}\left[\frac{1}{D(q,m\omega)}\right]\cos(q_x\gamma_0 - m\pi/2)\right\}. \tag{15}$$

We now make use of the following facts: (a) The dielectric function is only dependent on the magnitude of q; (b) the Bessel functions are symmetric for even m and antisymmetric for odd m with respect to the argument q_x. Therefore the integration over the direction of q will be zero for the second term in the curly brackets. The real part of the electric current is now written as

$$\mathrm{Re}[\mathbf{j}_{1x}(\omega)] = -\frac{ne}{m^*\omega}\sum_{\mathbf{q}} q_x V_q \sum_{m\neq 0}\frac{J_m(q_x\gamma_0)}{m} \times\mathrm{Im}\left[\frac{1}{D(q,m\omega)}\right]\sin(q_x\gamma_0 - m\pi/2). \tag{16}$$

The parameter that controls the electrical field dependence of the current is γ_0. We consider the two limiting cases. (i) At small γ_0 (low field and high frequency), we only retain the linear and cubic terms in E_0. The real part of the current is given as

$$\mathrm{Re}[\mathbf{j}_{1x}(\omega)] \approx -\frac{ne^2}{(m^*)^2\omega^3}\sum_{\mathbf{q}} q_x^2 V_q\,\mathrm{Im}\left[\frac{1}{D(q,\omega)}\right]E_0 + \frac{ne^4}{(m^*)^4\omega^5}\sum_{\mathbf{q}} q_x^4 V_q\,\mathrm{Im}\left[\frac{1}{D(q,\omega)} - \frac{1}{D(q,2\omega)}\right]E_0^3. \tag{17}$$

The current is linear in E_0 as $E_0\to 0$. This is in contrast to the E_0^2 dependence predicted by other theories. It is consistent with the linear dependence found in the experiment.[11] Since the magnetic field is absent in the current calculation, we cannot make a direct comparison with the experiment. However, we do not expect that application of a constant magnetic field will alter such a linear dependence. It is also important to note that terms of high order in E_0 also represent high-order multiphoton processes.

(ii) At large γ_0, $J_m(q_x\gamma_0)\sim\sqrt{2/\pi q_x\gamma_0}\cos(q_x\gamma_0 - m\pi/2 + \pi/4)$. Therefore the dissipative part of the electrical current vanishes in the limit of strong electrical field. This is the dynamic equivalence of electron localization in a strong static electric field. When electrons are strongly coupled to the photons, electron-impurity scattering approaches zero.

We have numerically calculated the real part of the current. The parameters used in our calculation are those of GaAs, $m^*=0.067m_0$, and $r_s=m^*e^2/\epsilon_0 k_F=1.0$. All energies and frequencies are in units of E_F, and all wave numbers are in units of k_F. Here $\alpha=k_F eE/(m^*\omega^2)$ is the dimensionless electron-photon coupling parameter.

Figure 1 shows electric field dependence of the real part of the electric current divided by the reduced electric field, $r=k_F eE/(m^*\omega_0^2)$ (where $\omega_0=1$ THz). In the plot, we multiply $\mathrm{Re}(j)/r$ by the frequency squared so the values $R=\omega^2\,\mathrm{Re}(j)/r$ at three different frequencies are comparable. The quantity R is a direct measure of the absorption of THz photons by the electronic system due to electron-impurity scattering. At weak electric fields or high frequencies, the current is almost linear in the electric field. The electron-photon coupling is inversely proportional to ω^2. Therefore the absorption increases as frequency decreases. At fixed frequency, as the laser intensity increases, the current starts to deviate from the linear dependence and the absorption coefficient j/E starts to decrease with the field intensity. This behavior is a direct consequence of the electron-impurity scattering time being affected by electron-photon coupling. At low field intensity or weak electron-photon coupling, the scattering time is independent of the electric field. As the field intensity increases, the electron-photon coupling becomes stronger and, as a consequence, the electron-impurity

scattering becomes less effective. This reduced electron-impurity scattering is the origin of the reduction of the dissipative current at high fields.

In Fig. 2, we plot the quantity $R=\omega^2 \mathrm{Re}(j)/r$ versus the laser frequency. At low frequency, the absorption increases with frequency due to the increasing phase space for electron-impurity scattering. The energy loss of the electromagnetic waves is weaker but increases more rapidly at strong field. At high frequency the real part of the current decreases with frequency due to rapidly decreasing electron-impurity scattering at the high-frequency region. At high frequency, electron-photon coupling is weak and therefore the conductivity under the strong field is very close to that under the weak field.

In conclusion, we have presented a study of the nonlinear electrical transport in low-dimensional systems under intense THz laser radiation. Due to the strong electron-photon coupling, the electron-impurity scattering and its effect on the energy loss of the laser field are significantly reduced.

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