

# Effect of photon-assisted absorption on the thermodynamics of hot electrons interacting with an intense optical field in bulk GaAs

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(Received 27 February 2004; revised manuscript received 13 July 2004; published 7 January 2005)

The use of a Boltzmann transport equation with a drift term is physically incorrect for optical-field frequencies. Also, the use of a simple energy-balance equation is found to lead to an inaccurate estimation of electron temperature. Therefore, we have established a Boltzmann-scattering equation for the accurate description of the relative scattering motion of electrons interacting with an incident optical field by including impurity- and phonon-assisted photon absorption as well as Coulomb scattering between two electrons. Multiple peaks on the high-energy tail of a Fermi-Dirac distribution are predicted and the effect of pair scattering is analyzed. Moreover, the effective electron temperature is calculated as a function of both the incident-field amplitude and the photon energy so that the thermodynamics of hot electrons may be investigated.

DOI: 10.1103/PhysRevB.71.045204

PACS number(s): 61.82.Fk, 61.80.Az, 72.20.Ht, 72.10.Di

## I. INTRODUCTON

The use of a Boltzmann transport equation with a drift term<sup>1</sup> under a time-dependent electric field can be justified only within the limit of  $\nu_f \tau_p \ll 1$ , where  $\nu_f$  is the frequency of the external field and  $\tau_p$  is the momentum-relaxation time of carriers. This approach can no longer be physically justified<sup>2-5</sup> for an incident electromagnetic field with  $\nu_f \geq 1$  THz, where  $\tau_p = 1$  ps is assumed. This is because electrons are expected to be spatially localized when the time period ( $1/\nu_f$ ) of the electromagnetic field becomes equal to or shorter than the momentum-relaxation time ( $\tau_p$ ) of electrons. As a result, no drift of electrons will occur under such an electromagnetic field.

When the motion of electrons is separated into center-of-mass and relative motions, the incident electromagnetic field is found to be coupled only to the center-of-mass motion but not to the relative motion of electrons.<sup>6-9</sup> This will generate an oscillating drift velocity in the center-of-mass motion, but the time-average value of this drift velocity also remains zero as described above. This oscillating drift velocity will, however, affect the electron-phonon and electron-impurity interactions. On the other hand, the thermodynamics of electrons is determined by the relative motion of electrons.<sup>9-11</sup> This includes the scattering of electrons with impurities, phonons, and other electrons.

When the incident electromagnetic field is spatially uniform, electrons in bulk GaAs cannot directly absorb incident photons through an intraband transition.<sup>9</sup> Because both impurity atoms and lattice ions do not move with the electron center-of-mass, electrons inside a drifting system feel that impurities and ions are oscillating against them due to the Galilean principle of relative motion. This leads to impurity- and phonon-assisted photon absorption in the system.

The relative scattering motion of electrons cannot be fully described by a simple energy-balance equation.<sup>7,8,12</sup> The oscillating drift velocity is accounted for, but the thermal effect of pair scattering on the distribution of electrons is not included in this equation. The peak structures separated by multiples of the photon energy in the distribution of electrons

are also absent in this equation. This leads to an inaccurate estimate of the electron temperature.

The relative scattering motion of electrons can be very well described by a Boltzmann scattering equation (Boltzmann transport equation without a drift term).<sup>9,10</sup> The effect of an incident optical field is reflected in the impurity- and phonon-assisted photon absorption through modifying the scattering of electrons with impurities and phonons. This drives the distribution of electrons away from the thermal-equilibrium distribution to a nonequilibrium one. At the same time, the electron temperature increases with the strength of the incident electromagnetic field, creating hot electrons.

In this paper, we will establish a Boltzmann scattering equation for an accurate description of the relative scattering motion of electrons interacting with an intense optical field by including both the impurity- and phonon-assisted photon absorption processes as well as the Coulomb scattering between two electrons. We will study the thermodynamics of hot electrons by calculating the effective electron temperature as a function of both the amplitude of the optical field and the incident photon energy.

The organization of the paper is as follows. In Sec. II, we introduce a Boltzmann scattering equation describing the scattering of electrons with impurities, phonons, and other electrons. Numerical results are displayed in Sec. III for the effect of pair scattering on the distribution of electrons and the dependence of electron temperature on the strength and the frequency of the incident optical field. The paper is briefly concluded in Sec. IV.

## II. BOLTZMANN SCATTERING EQUATION

The nonequilibrium distribution  $n_k$  for electrons in a conduction band satisfies the Boltzmann scattering equation<sup>9,10</sup>

$$\frac{d}{dt} n_k = \mathcal{W}_k^{(\text{in})} (1 - n_k) - \mathcal{W}_k^{(\text{out})} n_k, \quad (1)$$

where  $k$  is a wave number for electrons,  $n_k$  is the nonequilibrium distribution of electrons, and a Markovian process is

assumed for the dynamics of electron scattering. In the above equation,

$$\mathcal{W}_k^{(\alpha)} = \mathcal{W}_k^{(\alpha)(\text{im})} + \mathcal{W}_k^{(\alpha)(\text{ph})} + \mathcal{W}_k^{(\alpha)(c)}, \quad (2)$$

where  $\mathcal{W}_k^{(\alpha)}$  with  $\alpha=\text{in}$  or  $\text{out}$  represents the scattering-in/scattering-out rate for electrons staying in/leaving the  $k$  state. The superscripts  $\text{im}$ ,  $\text{ph}$ ,  $c$  represent the impurity, phonon, and Coulomb scattering of electrons.

The electron scattering-in/scattering-out rates due to impurities are given by<sup>9</sup>

$$\begin{aligned} \mathcal{W}_k^{(\text{in})(\text{im})} &= N_I \frac{2\pi}{\hbar} \sum_{\vec{q}} |U^{\text{im}}(q)|^2 \sum_{M=-\infty}^{\infty} J_{|M|}^2(\mathcal{M}_q) \\ &\times [n_{|\vec{k}-\vec{q}|} \delta(\varepsilon_k - \varepsilon_{k-q} - M\hbar\Omega_{\text{op}}) \\ &+ n_{|\vec{k}+\vec{q}|} \delta(\varepsilon_k - \varepsilon_{k+q} + M\hbar\Omega_{\text{op}})], \end{aligned} \quad (3)$$

$$\begin{aligned} \mathcal{W}_k^{(\text{out})(\text{im})} &= N_I \frac{2\pi}{\hbar} \sum_{\vec{q}} |U^{\text{im}}(q)|^2 \sum_{M=-\infty}^{\infty} J_{|M|}^2(\mathcal{M}_q) \\ &\times [(1 - n_{|\vec{k}+\vec{q}|}) \delta(\varepsilon_{k+q} - \varepsilon_k - M\hbar\Omega_{\text{op}}) \\ &+ (1 - n_{|\vec{k}-\vec{q}|}) \delta(\varepsilon_{k-q} - \varepsilon_k + M\hbar\Omega_{\text{op}})], \end{aligned} \quad (4)$$

where  $J_M(x)$  is the  $m$ th order first-kind Bessel function,  $\varepsilon_k = \hbar^2 k^2 / 2m^*$  is the kinetic energy of an electron in a conduction band,  $m^*$  is the effective mass of electrons,  $n_I = N_I / \mathcal{V}$  is the impurity concentration with sample volume  $\mathcal{V}$ ,  $\hbar\Omega_{\text{op}} (\Omega_{\text{op}} = 2\pi\nu_f)$  is the incident photon energy,

$$\mathcal{M}_q = \frac{eqE_{\text{op}}}{\sqrt{2m^*}\Omega_{\text{op}}^2} \quad (5)$$

for a nonpolarized incident optical field,  $E_{\text{op}}$  is the amplitude of the incident optical field, and the impurity scattering potential is

$$U^{\text{im}}(q) = \frac{Ze^2}{\epsilon_0 \epsilon_r (q^2 + Q_s^2) V}. \quad (6)$$

Here,  $\epsilon_r$  is the average dielectric constant of GaAs,  $Z$  is the charge number of an ionized impurity atom,  $Q_s^2 = (e^2 / \epsilon_0 \epsilon_r) \times (m^* / \pi^2 \hbar^2) (3\pi^2 \sigma_{3D})^{1/3}$  represents the static Thomas-Fermi screening effect,<sup>13</sup> and  $\sigma_{3D}$  is the concentration of conduction electrons in a bulk GaAs.

The electron scattering-in/scattering-out rates due to phonons, including phonon-assisted photon absorption, are given by<sup>9</sup>

$$\begin{aligned} \mathcal{W}_k^{(\text{in})(\text{ph})} &= \frac{2\pi}{\hbar} \sum_{\vec{q}, \lambda} |C_{q\lambda}|^2 \sum_{M=-\infty}^{\infty} J_{|M|}^2(\mathcal{M}_q) \\ &\times [n_{|\vec{k}-\vec{q}|} N_{q\lambda} \delta(\varepsilon_k - \varepsilon_{k-q} - \hbar\omega_{q\lambda} - M\hbar\Omega_{\text{op}}) \\ &+ n_{|\vec{k}+\vec{q}|} (N_{q\lambda} + 1) \delta(\varepsilon_k - \varepsilon_{k+q} + \hbar\omega_{q\lambda} + M\hbar\Omega_{\text{op}})], \end{aligned} \quad (7)$$

$$\begin{aligned} \mathcal{W}_k^{(\text{out})(\text{ph})} &= \frac{2\pi}{\hbar} \sum_{\vec{q}, \lambda} |C_{q\lambda}|^2 \sum_{M=-\infty}^{\infty} J_{|M|}^2(\mathcal{M}_q) \\ &\times [(1 - n_{|\vec{k}+\vec{q}|}) N_{q\lambda} \delta(\varepsilon_{k+q} - \varepsilon_k - \hbar\omega_{q\lambda} - M\hbar\Omega_{\text{op}}) \\ &+ (1 - n_{|\vec{k}-\vec{q}|}) (N_{q\lambda} + 1) \\ &\times \delta(\varepsilon_{k-q} - \varepsilon_k + \hbar\omega_{q\lambda} + M\hbar\Omega_{\text{op}})]. \end{aligned} \quad (8)$$

Here,  $\hbar\omega_q$  represents the phonon energy, phonons are assumed to be in thermal equilibrium with an external heat bath at a fixed temperature  $T$ . The distribution of phonons is a Bose-Einstein function, given by

$$N_{q\lambda} = \frac{1}{\exp(\hbar\omega_{q\lambda}/k_B T) - 1}. \quad (9)$$

For optical phonon scattering, we find from the Fröhlich electron-phonon coupling<sup>14</sup> (with  $\lambda=\text{LO}$ )

$$|C_{q\text{LO}}|^2 = \left( \frac{\hbar\omega_{\text{LO}}}{2\mathcal{V}} \right) \left( \frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_s} \right) \frac{e^2}{\epsilon_0 (q^2 + Q_s^2)}, \quad (10)$$

where  $\omega_{\text{LO}}$  is the frequency of dominant longitudinal-optical (LO) phonon modes at high temperatures,  $\epsilon_\infty$  and  $\epsilon_s$  are the high-frequency and static dielectric constants of GaAs. For acoustic phonon scattering, we find from the deformation-potential approximation<sup>9</sup> (with  $\lambda = \ell, t$ )

$$|C_{q\ell}|^2 = \frac{\hbar\omega_{q\ell}}{2\rho c_\ell^2 \mathcal{V}} \left[ D^2 + \frac{9}{32q^2} (eh_{14})^2 \right] \left( \frac{q^2}{q^2 + Q_s^2} \right)^2, \quad (11)$$

$$|C_{qt}|^2 = \frac{\hbar\omega_{qt}}{2\rho c_t^2 \mathcal{V}} \frac{13}{64q^2} (eh_{14})^2 \left( \frac{q^2}{q^2 + Q_s^2} \right)^2, \quad (12)$$

where  $\lambda = \ell, t$  corresponds to one longitudinal and two transverse acoustic-phonon modes,  $c_\ell$  and  $c_t$  are the sound velocities for these modes,  $\rho$  is the ion mass density,  $D$  is the deformation-potential coefficient, and  $h_{14}$  is the piezoelectric constant. Applying the Debye model to low-energy acoustic phonons, we get  $\omega_{q\lambda} = c_\lambda q$  with  $\lambda = \ell, t$ .

The electron scattering-in/scattering-out rates due to the Coulomb interaction between electrons are given by<sup>15</sup>

$$\begin{aligned} \mathcal{W}_k^{(\text{in})(c)} &= \frac{2\pi}{\hbar} \sum_{\vec{k}', \vec{q}} |V^c(q)|^2 (1 - n_{k'}) n_{|\vec{k}-\vec{q}|} n_{|\vec{k}'+\vec{q}|} \\ &\times \delta(\varepsilon_k + \varepsilon_{k'} - \varepsilon_{k-q} - \varepsilon_{k'+q}), \end{aligned} \quad (13)$$

$$\begin{aligned} \mathcal{W}_k^{(\text{out})(c)} &= \frac{2\pi}{\hbar} \sum_{\vec{k}', \vec{q}} |V^c(q)|^2 n_{k'} (1 - n_{|\vec{k}-\vec{q}|}) (1 - n_{|\vec{k}'+\vec{q}|}) \\ &\times \delta(\varepsilon_{k-q} + \varepsilon_{k'+q} - \varepsilon_k - \varepsilon_{k'}), \end{aligned} \quad (14)$$

where the Coulomb scattering potential is

$$V^c(q) = \frac{e^2}{\epsilon_0 \epsilon_r (q^2 + Q_s^2) \mathcal{V}}. \quad (15)$$

Coulomb scattering of electrons is a relative motion between electrons. As a result, the external optical field does not directly couple to it.

We know that the electron temperature  $T_e$  is generally a reflection of the magnitude of the average kinetic energy of all electrons even in a nonequilibrium state. Therefore, we can define an effective electron temperature at each moment by using a quasiequilibrium Fermi-Dirac function ( $T_e \neq T$ ) in the equation of the average kinetic energy of electrons<sup>9</sup>

$$\int_0^{+\infty} \frac{k^4 dk}{1 + \exp\{[\varepsilon_k - \mu(T_e)]/k_B T_e\}} = \int_0^{+\infty} n_k k^4 dk, \quad (16)$$

where the chemical potential  $\mu(T_e)$  can be determined for given  $T_e$  and  $\sigma_{3D}$  by

$$\frac{1}{\pi^2} \int_0^{+\infty} \frac{k^2 dk}{1 + \exp\{[\varepsilon_k - \mu(T_e)]/k_B T_e\}} = \sigma_{3D}. \quad (17)$$

For a thermal equilibrium distribution of electrons, the electron temperature equals the lattice temperature. On the other hand, the electron temperature for a quasiequilibrium distribution of electrons is not the same as the lattice temperature. However, the functional form of the quasiequilibrium distribution is assumed to be the Fermi-Dirac function with the electron temperature determined by the additional energy-balance equation.<sup>9,16,17</sup> For a general transient or steady-state distribution of electrons, there is no simple quantum-statistical definition for the electron temperature in all ranges. However, at high electron temperatures we can still define an *effective* electron temperature through the Fermi-Dirac function according to Eq. (17) with the conservation of the total number of electrons.

The Fermi-Dirac function describes the quantum statistics of degenerate electrons at low electron temperatures and high electron densities. When either the electron temperature is high or the electron density is low, the Fermi-Dirac function reduces to the Boltzmann function for nondegenerate electrons with conservation of the total number of electrons. In the nondegenerate case, the average kinetic energy of electrons is proportional to the electron temperature. The numerically calculated distribution of electrons in this paper is not the Fermi-Dirac function. We only use the Fermi-Dirac function to define an effective electron temperature in the high-temperature range in Eq. (16) by equating the numerically calculated average kinetic energy of electrons with that of the Fermi-Dirac function for the same number of electrons.

In earlier work,<sup>16,17</sup> the temperature of plasma electrons was defined through a simplified energy-balance equation, including inelastic scattering and joule heating of electrons. The inelastic scattering of electrons was treated within a perturbation theory, and the electron current was calculated using a Drude-type model including displacement current. The electron temperature was found to be independent of time when an optical field was applied to the plasma. However, in the current theory, the distribution of electrons is calculated exactly, and the effective electron temperature is obtained based on this calculated distribution. The heating from the incident optical field is included through the phonon-assisted photon absorption process.

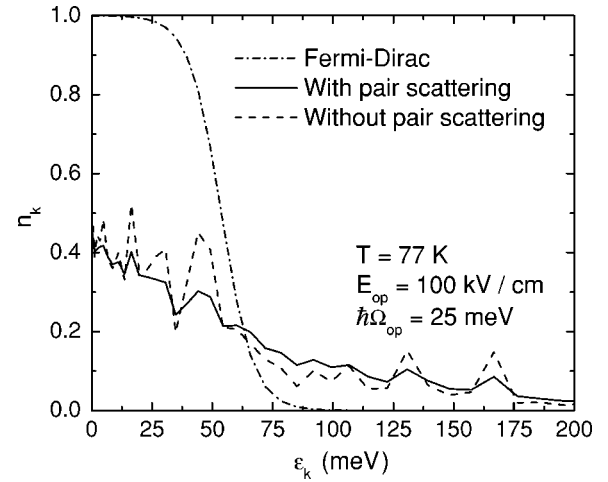


FIG. 1. Calculated electron distribution  $n_k$  for a bulk GaAs as a function of the kinetic energy  $\varepsilon_k$  of electrons at  $T=77$  K with  $\hbar\Omega_{op}=25$  meV and  $E_{op}=100$  kV/cm. The dash-dotted curve is for the thermal-equilibrium Fermi-Dirac function. The solid curve is for the calculated nonequilibrium electron distribution with the effect of pair scattering, while the dashed curve represents that without the effect of pair scattering. The other parameters are given in the text.

### III. NUMERICAL RESULTS AND DISCUSSIONS

In our numerical calculations, we have chosen GaAs as the host material. For GaAs we have taken parameters<sup>13</sup> as follows:  $m^*=0.067m_0$  with free-electron mass  $m_0$ ,  $\sigma_{3D}=1 \times 10^{18}$  cm<sup>-3</sup>,  $\hbar\omega_{LO}=36$  meV,  $\varepsilon_r=12$ ,  $\varepsilon_s=13$ ,  $\varepsilon_\infty=11$ ,  $\rho=5.3$  g/cm<sup>3</sup>,  $c_t=5.14 \times 10^5$  cm/sec,  $c_l=3.04 \times 10^5$  cm/sec,  $D=-9.3$  eV,  $h_{14}=1.2 \times 10^7$  V/cm, and  $T=300$  K. Other parameters, such as  $E_{op}$  and  $\hbar\Omega_{op}$ , will be given directly in the figure captions. In the numerical calculations below, we will only show results for steady-state cases.

In our numerical calculation, we have employed the finite-difference method for solving the evolution of the distribution of conduction electrons under an intense optical field. The time step  $\Delta t$  is taken to be 0.5 fs. The steady-state electron distribution is obtained by ensuring the relative change of the results at two successive times smaller than  $10^{-6}$ . For discrete states of electrons, we have taken a total of eighty-one points for electron wave number  $k$  with  $\Delta k = 0.05(3\pi^2\sigma_{3D})^{1/3}$ . With this precision, which is limited by our current computation ability, we find that the dominant peaks in Fig. 1 can be resolved. We expect the curves in Fig. 1 will be smoother if a higher precision is taken.

Figure 1 displays a comparison between calculated non-equilibrium electron distributions  $n_k$  with/without the effect of pair scattering and the thermal-equilibrium Fermi-Dirac distribution of electrons (dash-dotted curve) at  $T=77$  K,  $E_{op}=100$  kV/cm, and  $\hbar\Omega_{op}=25$  meV. The impurity- and phonon-assisted photon absorption processes create multiple small peaks (dashed curve) on the high-energy tail (above 54 meV) of the Fermi-Dirac distribution. The multiple peaks predicted by our theory were completely missed by the simple energy-balance equation.<sup>12</sup> The occurrence of these high-energy peaks are attributed to electrons that have predominantly been scattered-out of low-energy states below

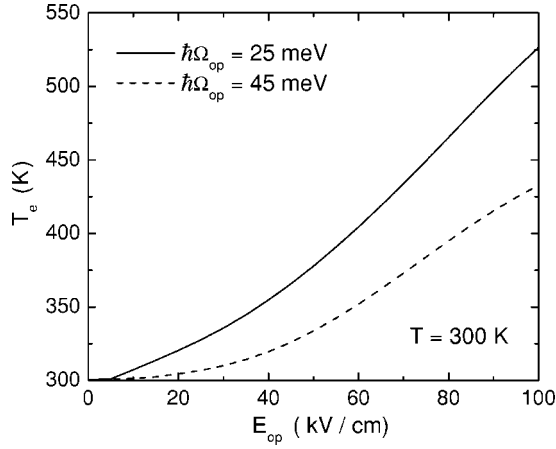


FIG. 2. Calculated effective electron temperature  $T_e$  for a bulk GaAs as a function of the amplitude  $E_{op}$  of incident electromagnetic field at  $T=300$  K with  $\hbar\Omega_{op}=25$  meV (solid curve) and  $\hbar\Omega_{op}=45$  meV (dashed curve). The other parameters are given in the text.

the Fermi energy (at 54 meV), which can be seen from terms with  $M \neq 0$  in Eqs. (3) and (7). At the same time, the combination of the electrons that have been scattered out from below the Fermi energy and the electrons that have been scattered out from the band edge leave an oscillation of the electron distribution there. From the conservation of electron number in the conduction band, we expect that the electron distribution below the Fermi energy will drop as the distribution spreads towards high-energy states. This can be equivalently viewed as an increase of an effective electron temperature through assisted photon absorption. The effect of pair scattering was zero in the energy-balance equation due to conservation of total energy. However, when the effect of pair scattering is included (solid curve) in our theory, the multiple peaks both above and below the Fermi energy are found to be reduced in size and broadened. As a price, the overall electron distribution above/below the Fermi energy is enhanced/suppressed, leading to an even higher electron temperature through pair scattering. This analysis shows that the simple energy-balance equation<sup>12</sup> will generally lead to an inaccurate estimation of the electron temperature.

We show in Fig. 2 the calculated effective electron temperature  $T_e$  as a function of the amplitude  $E_{op}$  of an incident electromagnetic field at  $T=300$  K for  $\hbar\Omega_{op}=25$  meV (solid curve) and  $\hbar\Omega_{op}=45$  meV (dashed curve). From the figure we find that  $T_e$  increases with increasing  $E_{op}$ . This is because the rate of the dominant assisted photon absorption with  $M \neq 0$  is proportional to  $E_{op}^2/\Omega_{op}^4$ . The increase of  $E_{op}$  for fixed  $\Omega_{op}$  implies the enhancement of photon absorption, which increases the electron temperature by pushing up the average electron kinetic energy. Moreover, we find that the increase of  $T_e$  with  $E_{op}$  is reduced when the photon energy  $\hbar\Omega_{op}$  changes from 25 meV (solid curve) to 45 meV (dashed curve). This is because the rate of the dominant assisted photon absorption with  $M \neq 0$  is inversely proportional to photon energy.

The decrease of  $T_e$  with  $\hbar\Omega_{op}$  can be seen even more clearly in Fig. 3, where the calculated effective electron tem-

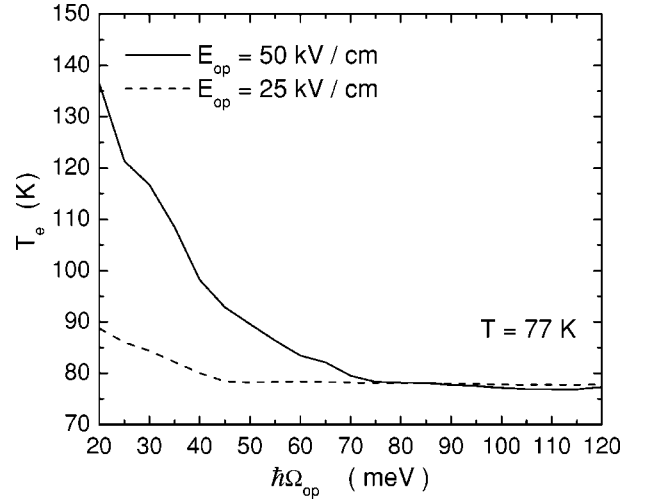


FIG. 3. Calculated effective electron temperature  $T_e$  for a bulk GaAs as a function of the energy  $\hbar\Omega_{op}$  of incident photons at  $T=77$  K with  $E_{op}=25$  kV/cm (dashed curve) and  $E_{op}=50$  kV/cm (solid curve). The other parameters are given in the text.

perature  $T_e$  is presented as a function of the incident photon energy  $\hbar\Omega_{op}$  at  $T=77$  K for  $E_{op}=25$  kV/cm (dashed curve) and  $E_{op}=50$  kV/cm (solid curve). From this figure, we find that  $T_e$  decreases with increasing  $\hbar\Omega_{op}$  initially and levels out at the given lattice temperature  $T=77$  K finally (solid curve). As described above, the rate of the dominant assisted photon absorption is proportional to  $E_{op}^2/\Omega_{op}^4$ . The increase of  $\Omega_{op}$  for fixed  $E_{op}$  means the reduction of photon absorption. As we can see from this figure, the initial drop of  $T_e$  is a result of the decreased assisted photon absorption. The final leveling out of  $T_e$  is attributed to electrons and the lattice gradually approaching a thermal-equilibrium state due to complete suppression of assisted photon absorption. This correct asymptotic behavior was not seen from the numerical result of the energy-balance equation.<sup>12</sup> When the incident electromagnetic field changes from 50 to 25 kV/cm (dashed curve), the initial drop of  $T_e$  is partially suppressed and the asymptotic approach of  $T_e$  to  $T$  occurs at an even lower photon energy as expected.

#### IV. CONCLUSIONS

In conclusion, we have established a Boltzmann scattering equation for the accurate description of relative scattering motion of electrons interacting with an incident optical field by including the impurity- and phonon-assisted photon absorption as well as the Coulomb scattering between two electrons. We have quantified multiple peaks on the high-energy tail of a Fermi-Dirac distribution and analyzed the effect of pair scattering. Moreover, we have calculated the effective electron temperature as functions of both the incident-field amplitude and the photon energy. We have found that the use of a transport equation with a drift term for such optical-field frequencies cannot be physically justified. We have further found that the use of a simple energy-balance equation will lead to an inaccurate estimation of the electron temperature.

The effect of pair scattering is sometimes included as a homogeneous broadening in a phenomenological theory. However, the effect of pair scattering in this paper is found to be inhomogeneous, which depends on the wave number of electrons. Solving a Boltzmann equation beyond the relaxation-time approximation is a difficult numerical procedure. The exact solution in this paper provides a tool for testing the justification of simplified theories, e.g., relaxation-time approximation and linearized Fokker-Planck

equation, in certain parameter ranges. Although the appearance of the multiple peaks in the distribution of electrons can be foreseen through the phonon-assisted photon absorption process, the relative strength of these peaks is very hard to predict. The relative strength of peaks at high electron kinetic energies turns out to be extremely important for understanding the laser damage of the semiconductor material through impact ionization of valence electrons<sup>11</sup> and for hot-electron transport.<sup>9</sup>

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