# Hot-electron cooling and second-generation phonons in polar semiconductors

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The acoustic-phonon bottleneck is studied theoretically within the model which includes Fröhlich interaction between electrons and LO phonons, and anharmonic coupling between the LO phonons and second-generation acoustic phonons, in the form of the Klemens interaction operator. Calculations are performed for bulk CdSe and GaAs. The acoustic-phonon bottleneck effect is shown to depend on the amount of energy provided by the laser pulse. The lifetime of the hot-electron cooling in the regime of the acoustic-phonon bottleneck appears to be controlled by the highly efficient channel of the fusion of acoustic phonons into LO phonons.

## I. INTRODUCTION

The process of cooling of the hot-electron–hole plasma in semiconductors, excited by a laser pulse, is one of the important processes in the field of nonequilibrium transport phenomena of electrons and phonons.[1] In polar semiconductors, at not very low temperatures, the hot electrons relay their energy mainly to the longitudinal-optical (LO) phonons.[2] The reduction of the hot-electron cooling rate observed in experiment has been ascribed, on the basis of simple estimates performed with the help of the Golden rule formula, to the effect of the LO phonon bottleneck,[2] although among the possible reasons for this reduction the effect of the electric screening[2] and of the phonon-plasmon coupled modes[3] have also been considered. In the case of the LO-phonon bottleneck the rate of the hot-electron cooling is controlled by the lifetime of the LO phonons, so that the whole electron-phonon system is cooling down with a time constant of picoseconds. The decay channel of the LO phonons is considered[4] to be the decay of an LO phonon into a pair of acoustic phonons, at least in GaAs. Usually, these vibrational modes, into which the LO-phonons decay, are considered to stay at equilibrium with the ambient lattice.

In several experimental papers the hot-electron cooling after a short laser pulse has been reported to have quite a large relaxation time constant.[5–8] In CdS,[5] CdSe,[6] superlattice AlGaAs-GaAs,[7] and in a solid-state solution $CdS_{1-x}Se_x$,[8] it has been observed that several tens of picoseconds after the termination of the laser pulse, typically 30 ps long, the lifetime of the hot-electron cooling is about 150–400 ps. Such a low cooling rate has been tentatively ascribed[6,7] to the effect of acoustic-phonon bottleneck, in which the relaxation of the electron-phonon system is controlled by the lifetime $\tau_A$ of the acoustic phonons. In Ref. 6, the possibility of the acoustic-phonon bottleneck was supported by a simple argument based on balancing the energy relayed from the light pulse to the electrons and from the electrons to the phonon modes. A hot-electron cooling with very long relaxation times was also detected in GaAs-AlGaAs quantum wells, in bulk GaSb,[9,10] and in AlSb-GaSb quantum wells.[11] In Refs. 5–11 the long relaxation times are usually observed under conditions in which large amounts of energy are available to the electron-phonon system via the excitation of electrons.

Within the acoustic-phonon bottleneck effect interpretation,[6] the hot electrons are again assumed to emit LO phonons, the population of which can, in close vicinity to the $\Gamma$ point, become larger than the equilibrium population, corresponding to the temperature of the ambient lattice. The nonequilibrium LO phonons, which can be called "first-generation phonons" in the present context, are assumed to decay into pairs of second-generation phonons, which, in GaAs, are assumed to be identical with the longitudinal branch of the acoustic phonons (LA). Because of the energy and momentum conservation restrictions these second-generation phonons are produced mainly at such wave vectors in the reciprocal space, at which the second-generation phonons have energy equal to the one half of the LO-phonon energy.[6] In the present paper we shall consider the case when the second-generation phonons are the LA phonons, which appears to be a reasonable approximation. This point will be discussed later in this paper.

The nonequilibrium population of the second-generation phonons can, under favorable conditions, become hot, so that the first-generation phonons (LO phonons) cannot cool down efficiently and the acoustic-phonon bottleneck can be expected to develop. The buildup of the acoustic phonons produced by the decay of the LO phonons was demonstrated experimentally.[12] The occurrence of the LO-phonon decay into a pair of LA phonons is supported by the experiments in the GaAs heterojunction.[13]

The second-generation phonons have been assumed[6] to decay anharmonically with a certain lifetime. In particular, in GaAs,[4] the LA phonons are expected to decay into pairs of (third-generation) transverse-acoustic phonons. One can ascribe the LA phonons a lifetime $\tau_A$, due to this channel of decay. In Ref. 6 the lifetime $\tau_A$ of the second-generation phonons has been closely connected with the lifetime of the observed hot-electron cooling.

Let us note that the problem of the considerable slowing down of the relaxation of the electronic subsystem in

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semiconductors, after a long laser pulse, has been studied earlier; see, e.g., Refs. 14 and 15. In Ref. 14 the cooling of the hot-electron plasma in laser excited Ge was measured to have the relaxation time of about 40–75 ps. The reader is also referred to Ref. 16 for references related to the so-called "laser annealing debate" of the early 1980s.

It is clear that the effect of the slowing down of the hot-electron cooling, either alone or in connection with the possibility of a buildup of the second-generation phonons, still deserves attention. In our previous work[17] the effect of the acoustic-phonon bottleneck was studied theoretically in the dispersionless approximation, in which both the energy of the LO phonons and the velocity of the acoustic phonons were assumed to be constant throughout the entire Brillouin zone. The acoustic phonons, as the second-generation phonons, were considered to decay in two ways in Ref. 17. First, they decay into third-generation phonons by an anharmonic mechanism. This decay channel was represented by a relaxation time approximation term in the kinetic equations in Ref. 17. Second, the second-generation acoustic phonons were shown to annihilate very efficiently in the reaction of fusion of two acoustic phonons into a LO phonon. The decay lifetime $\tau_F$ of second-generation acoustic phonons, with respect to the fusion channel, was estimated to be roughly equal to the lifetime of the LO phonon. In part, this high efficiency of the fusion channel was ascribed to the simplified characteristics of the dispersionless model assumed, in which the energy and momentum conservation restrictions are broadly fulfilled. It was suggested in Ref. 17 that although the fusion channel may always contribute to a certain extent to the decay of the nonequilibrium acoustic-phonon population, it can, at least in part, be shut by taking into account the realistic dispersion of the LO phonons.

In the present paper our previous theoretical analysis[17] of the relaxation of the laser pulse excited electron-phonon system is continued. The process of the hot-electron cooling and the effect of the acoustic-phonon bottleneck are studied taking into account the dispersion of both the longitudinal-optical and longitudinal-acoustic phonons. Also in contrast to the previous work, the finite duration of the exciting laser pulse is simulated here. In Sec. II the model of the physical system is introduced. The kinetic equations are formulated in Sec. III. In Sec. IV a simple but realistic approximation to the dispersion of the lattice modes is presented, and in Sec. V we describe the process that we analyze. Our results are presented and discussed in Sec. VI.

## II. THE PHYSICAL MODEL

In the experiments with which we wish to compare our theoretical results, the electrons of the valence band are excited over the band gap by the laser pulse, thus leaving the holes in the valence band and the electrons in the conduction band. In order to simplify the theory we ignore completely the process of the hole cooling, expecting that this neglect does not alter significantly the conclusions of the present work.

The Hamiltonian of the unperturbed conduction-band electrons is

$$H_e=\sum_{\mathbf{k},\sigma}\epsilon(\mathbf{k})c^{\dagger}_{\mathbf{k},\sigma}c_{\mathbf{k},\sigma}\ , \tag{1}$$

where $\epsilon(\mathbf{k})=\hbar^2k^2/(2m^*)$, $k=|\mathbf{k}|$ is a nondegenerate and parabolic electronic energy band with the effective mass $m^*$. The particle operator $c_{\mathbf{k},\sigma}$ annihilates an electron in the state with the wave vector $\mathbf{k}$ and the spin projection $\sigma$.

We assume that the hot electrons emit only LO phonons (first generation). The free LO phonons have the Hamiltonian

$$H_{\mathrm{LO}}=\sum_{\mathbf{q}}\hbar\omega_{\mathrm{LO}}(\mathbf{q})b^{\dagger}_{\mathbf{q}}b_{\mathbf{q}}\ . \tag{2}$$

In (2) $b_{\mathbf{q}}$ is LO-phonon particle operator. In the approximation of the present paper the electron and phonon systems are assumed to be isotropic and the Brillouin zone is approximated by a sphere having the same volume. The simplified dispersion of $\omega_{\mathrm{LO}}(\mathbf{q})$ will be specified in a later section.

The second-generation phonons can be, according to the crystal structure of III-V and II-VI compounds, acoustic phonons or optical phonons. So, in GaAs the second-generation phonons are acoustic phonons,[4] while in wurtzite modifications of CdS and CdSe, the second-generation phonons are identified with a certain branch of longitudinal-optical phonons.[6,18] It is well known, however, that within good accuracy,[19,20] the crystal modifications of CdS and CdSe can be approximated by the corresponding sphalerite modifications. In particular, both the first-, and second-generation phonons in wurtzite structures, including the main characteristics of the Brillouin zone, can be approximated by those of the corresponding sphalerite structure. Namely, as a good approximation, we can view the second-generation optical modes of wurtzite structure simply as a part of the branch of the longitudinal-acoustic modes of the corresponding sphalerite structure. As a prototype of the sphelerite structure we take the one of GaAs. Our results thus refer to both the sphalerite and the wurtzite modifications of the crystals in question, although we must be aware of a certain inaccuracy caused by the assumption made above.

The free-particle Hamiltonian of the second-generation acoustic phonons is

$$H_A=\sum_{\mathbf{q}}\hbar\omega_A(\mathbf{q})a^{\dagger}_{\mathbf{q}}a_{\mathbf{q}}\ . \tag{3}$$

The dispersion of the acoustic-phonon frequency $\omega_A(\mathbf{q})$ will be discussed later in this paper.

Even if we took into account the realistic phonon dispersions, together with the proper shape of the Brillouin zone, we would still be seriously limited by the present state of knowledge[21,22] of the operator $V_K$ of the anharmonic coupling, which provides the channel of the decay of LO phonons into pairs of the acoustic ones,

$$V_K=gV^{-1/2}\sum_{\mathbf{q},\mathbf{q}'}\phi(\mathbf{q},\mathbf{q}')(b^{\dagger}_{\mathbf{q}+\mathbf{q}'}a_{\mathbf{q}}a_{\mathbf{q}'}+b_{\mathbf{q}+\mathbf{q}'}a^{\dagger}_{\mathbf{q}}a^{\dagger}_{\mathbf{q}'})\ , \tag{4}$$

in which $V$ is the volume of the sample and[23,24]

$$\phi(\mathbf{q},\mathbf{q}')=\frac{2}{\omega_0^{3/2}}[\omega_{\mathrm{LO}}(\mathbf{q}+\mathbf{q}')\omega_A(\mathbf{q})\omega_A(\mathbf{q}')]^{1/2}\ , \tag{5}$$

Here $\omega_0 \equiv \omega_{\rm LO}(\mathbf{q}=0)$. The presence of $\omega_0$ in (5) makes $\phi(\mathbf{q},\mathbf{q}')$ dimensionless. The uncertainty in the knowledge of $V_K$ is connected with the dependence of the coupling factor $\phi(\mathbf{q},\mathbf{q}')$ on the wave vectors of the phonons. The functional dependence (5) was originally introduced on the basis of a certain generalization and has not been given any great quantitative reliance upon.[21–24] With regard to the isotropy of the phonon dispersions assumed above and because of the fact that the hot optical phonons are concentrated near the center of the Brillouin zone, the acoustic phonons are generated at the states which have the energy near $\hbar\omega_0/2$. With good accuracy, the coupling factor $\phi(\mathbf{q},\mathbf{q}')$ could then be reduced to a constant. However, in the case when the acoustic phonons become hot and thus the reverse processes of the fusion of two LA phonons into a single LO phonon become important, then the knowledge of a proper shape of $\phi(\mathbf{q},\mathbf{q}')$ would be desirable. The reason for this is[17] that the momentum conservation law permits the LO phonons to be produced by the fusion processes in a quite broad range of $\mathbf{q}$ space, not only in the close vicinity of the $\Gamma$ point. The restriction imposed on the fusion channel by the energy conservation may depend critically on the wave-vector dependence of the optical-phonon energy. In addition, the dependence of the coupling function $\phi$ on the wave vectors of the phonons taking part in the fusion can further influence the efficiency of the fusion. The calculation will nevertheless be performed with the operator (4) including the form (5) of the coupling constant.

The electrons will be assumed to interact with the LO phonons via the Fröhlich interaction operator

$$V_F = \sum_{\mathbf{k},\mathbf{q},\sigma} A_q (b_{\mathbf{q}} - b^{\dagger}_{-\mathbf{q}}) c^{\dagger}_{\mathbf{k},\sigma} c_{\mathbf{k}-\mathbf{q},\sigma} \ , \tag{6}$$

in which the coupling constant of the statically screened interaction[25,26] is

$$A_q = -ie(\hbar\omega_0)^{1/2}(2\varepsilon_0 V)^{-1/2}(\kappa_\infty^{-1}-\kappa_0^{-1})^{1/2} q^{-1} f_q \ , \tag{7}$$

in which the screening factor $f_q$ is ($q=|\mathbf{q}|$)

$$f_q = \frac{q^2}{q^2+q_D^2} \ . \tag{8}$$

The Debye screening parameter $q_D$ is given in Ref. 27,

$$q_D^2 = e^2 n_e/(\varepsilon_0 \kappa_\infty k_B T_e) \ . \tag{9}$$

$T_e$ is the temperature of the electronic subsystem at a given instant of time, $e>0$ is the electronic charge, $\varepsilon_0$ is the permittivity of the free space, $\kappa_\infty$ and $\kappa_0$ are, respectively, the high- and low-frequency dielectric constants, and $k_B$ is the Boltzmann constant. Consistent with making the assumption that Fröhlich coupling provides a significant energy relaxation channel of the hot electrons, we consider only moderate concentrations of the conduction electrons and assume that they obey classical statistics, taking into account that at high densities the Fröhlich coupling is seriously screened and its effect is only minor.[16]

## III. KINETIC EQUATIONS

The electrons will be assumed to be nondegenerate and thermalized at each instant of time, being characterized by the dimensionless reverse temperature parameter $x$, $x=\hbar\omega_0/(k_B T_e)$. The phonon subsystem will be characterized by the single-phonon distribution functions. The corresponding kinetic equations are derived within the framework of the nonequilibrium statistical operator theory.[28]

The LO phonons, characterized by the mean number $\nu_{\mathbf{q}}$ of the LO phonons in the state with the wave vector $\mathbf{q}$, interact with the conduction electrons and acoustic phonons. In the lowest order in $V_F$ and $V_K$ we confine ourselves to the following two terms of the rate of change $d\nu_{\mathbf{q}}/dt$:

$$d\nu_{\mathbf{q}}/dt = (d\nu_{\mathbf{q}}/dt)_{e\text{-LO}} + (d\nu_{\mathbf{q}}/dt)_{\text{LO-}A} \ , \tag{10}$$

where the term $(d\nu_{\mathbf{q}}/dt)_{e\text{-LO}}$ denotes the generate rate of LO phonons produced by the hot-electron cooling. The second term, $(d\nu_{\mathbf{q}}/dt)_{\text{LO-}A}$, is the rate of the decay of LO phonons into the acoustic ones. At the level of approximation of the Boltzmann equation we get

$$(d\nu_{\mathbf{q}}/dt)_{\text{LO-}A} = -\frac{2\pi}{\hbar}\frac{2g^2}{V}\sum_{\mathbf{q}_1}\phi^2(\mathbf{q}_1,\mathbf{q}-\mathbf{q}_1)[(1+N_{\mathbf{q}_1})(1+N_{\mathbf{q}-\mathbf{q}_i})\nu_{\mathbf{q}} - N_{\mathbf{q}_1}N_{\mathbf{q}-\mathbf{q}_1}(\nu_{\mathbf{q}}+1)]$$
$$\times\delta[\hbar\omega_{\rm LO}(\mathbf{q})-\hbar\omega_A(\mathbf{q}-\mathbf{q}_1)-\hbar\omega_A(\mathbf{q}_1)] \ . \tag{11}$$

Here $N_{\mathbf{q}}$ is the number of acoustic phonons in the state $\mathbf{q}$.

The term $(d\nu_{\mathbf{q}}/dt)_{e\text{-LO}}$ is the rate of generation of LO phonons by hot-electron cooling. Because the inclusion of the LO-phonon dispersion into this generation rate causes only negligible corrections to it, which is due to the fact that the LO phonons are generated only in a very small neighborhood of the $\Gamma$ point by the hot-electron cooling, we use here the formula given already in the previous paper,[17] namely

$$(d\nu_q/dt)_{e\text{-LO}} = \frac{e^2}{\varepsilon_0\hbar^2}\sqrt{\pi/2}\,n_e\sqrt{m\hbar\omega_0}(\kappa_\infty^{-1}-\kappa_0^{-1})\sqrt{x}\,\frac{f_q^2}{q^3}e^{-(x/4)(\tilde{q}+1/\tilde{q})^2}[1+\nu_q-\nu_q e^x] \ , \tag{12}$$

where $\tilde{q}=q[\hbar/(2m\omega_0)]^{1/2}$.

The rate of change $dN_{\mathbf{q}}/dt$ of the acoustic phonons is a sum of two terms,

$$dN_q/dt=(dN_q/dt)_{A\text{-LO}}+(dN_q/dt)_R \ , \tag{13}$$

where the first term contains the decay of the LO phonons into the acoustic ones and also the terms giving the reverse process of the fusion,

$$(dN_{\mathbf{q}}/dt)_{A\text{-LO}}=\frac{2\pi}{\hbar}\frac{4g^2}{V}\sum_{\mathbf{q}_1}\phi^2(\mathbf{q},\mathbf{q}_1)[(1+N_{\mathbf{q}})(1+N_{\mathbf{q}_1})\nu_{\mathbf{q}+\mathbf{q}_1}-N_qN_{\mathbf{q}_1}(1+\nu_{\mathbf{q}+\mathbf{q}_1})]$$
$$\times\delta[\hbar\omega_{\mathrm{LO}}(\mathbf{q}+\mathbf{q}_1)-\hbar\omega_A(\mathbf{q})-\hbar\omega_A(\mathbf{q}_1)] \ . \tag{14}$$

The second term in (13) is the decay rate of the longitudinal acoustic-phonon population into third-generation phonons. We shall express this term simply in the relaxation time approximation taking the third-generation phonons as a reservoir having a constant temperature of the ambient lattice $T_L$. The second-generation phonons are thus decaying with the time constant $\tau_A$ according to the rule

$$(dN_q/dt)_R=-\frac{N_q-N_q^{(0)}}{\tau_A} \ . \tag{15}$$

$N_{\mathbf{q}}^{(0)}$ is the equilibrium population of the second-generation acoustic modes corresponding to the temperature $T_L$,

$$N_q^{(0)}=\{\exp[\hbar\omega_A(\mathbf{q})/(k_BT_L)]-1\}^{-1} \ . \tag{16}$$

The evolution of the electronic temperature is given by

$$dx/dt=\frac{1}{3\pi^2n_e}x^2\int_0^\infty q^2(d\nu_q/dt)_{e\text{-LO}}dq \ . \tag{17}$$

Equations (10), (13), and (17) describe time evolution of the system in question, providing the initial conditions are given which simulate the experimental conditions we wish to compare with. These will be specified in a later section.

Before finishing this section let us show how the coupling constant $g$ in $V_K$ is deduced from the experimental data in the LO-phonon lifetime. Generally, the rate of change of the LO-phonon population depends on the state of the electrons, acoustic phonons, and the LO phonons in a complicated way. However, in the special case, when the electrons can be excluded out from consideration, when the acoustic phonons are at equilibrium having the temperature $T_L$, and when the nonequilibrium population of the LO phonons is small and close to the equilibrium population at $T_L$ and confined to a small area near the $\Gamma$ point, then the LO-phonon lifetime $\tau_{\mathrm{LO}}$ is given by the equation

$$\frac{1}{\tau_{\mathrm{LO}}}=\frac{1}{\tau_{\mathrm{LO}}^{(0)}}\left[1+\frac{2}{\exp\left[\dfrac{\hbar\omega_0}{2k_BT_L}\right]-1}\right] , \tag{18}$$

where

$$\frac{1}{\tau_{\mathrm{LO}}^{(0)}}=\frac{g^2q_0^2}{\pi\hbar^2|\omega_A'(q_0)|} \ , \tag{19}$$

in which $\omega_A'(q_0)=[d\omega_A(q)/dq]_{q=q_0}$ and $q_0$ is such that $\omega_0=2\omega_A(q_0)$. $\tau_{LO}^{(0)}$ is the lifetime of the LO phonons at absolute zero of temperature. We assume here that our model is isotropic so that $\omega_A(\mathbf{q})=\omega_A(q)$, $q=|\mathbf{q}|$. The equations reduce obviously to the results given in Ref. [17], in the dispersionless limit.

Equation (18) can be derived from the general equation (11) and is a generalization of the Klemens[23] formula to the case in which the dispersion of the acoustic phonons is included. It gives the temperature dependence of the LO-phonon lifetime. Equation (19) then allows us to obtain the constant $g$ from the LO-phonon lifetime at $T=0$. The simple form of (18) is a result of rather serious simplifications of assuming that the acoustic-phonon dispersion relation has a center of symmetry. Therefore the use of (18) assumes a reasonable estimate of an average dispersion model. This will be discussed in the next section.

## IV. SIMPLIFIED DISPERSION MODEL

In connection with what has been said in Sec. II concerning the plausible approximation of the wurtzite structure of CdSe by a sphalerite model, we approximate the Brillouin zones of both materials considered, GaAs and CdSe, by a sphere, the volume of which is equal to that of the so-called Jones zone[19,29] of the corresponding crystal. Let us note that the volume of the Jones zone of wurtzite is equal to twice the Brillouin-zone volume, while in the sphalerite crystal the volumes of both zones are the same. The Jones zone of wurtzite can be constructed by doubling the Brillouin zone in $\Gamma$-$A$ direction. This direction then corresponds, in this approximation, to the $\Gamma$-$L$ direction of sphalerite. Thus, when we unfold the dispersion curves, those given for wurtzite modification of CdSe in Ref. 18, we obtain, within an approximation, the dispersion curves of the sphalerite approximation in the $\Gamma$-$L$ direction. The other crystal directions are not given in Ref. 18.

In the spherical zone we thus consider one LO and one LA branch of the lattice phonons in both materials under consideration. Assuming thus the complete isotropy of the crystal properties we take the phonon frequencies as dependent only on the magnitude $q$ of the wave vector $\mathbf{q}$. Then we assume to be valid the following dependences on $q$, of the phonon frequencies:

$$\omega_{\mathrm{LO}}(q)=\tfrac{1}{2}(\omega_0-\omega_1)\cos\frac{\pi q}{Q}+\tfrac{1}{2}(\omega_0+\omega_1) \tag{20}$$

and

$$\omega_A(q)=\omega_2\sin\frac{\pi q}{2Q} \ . \tag{21}$$

TABLE I. Parameters of the wave-vector dependence of phonon frequencies.

| Quantity | Unit | CdSe | GaAs |
|---|---|---|---|
| $\omega_0$ | $10^{13}$ s$^{-1}$ | 4.000 | 5.502 |
| $\omega_1$ | $10^{13}$ s$^{-1}$ | 3.200 | 4.370 |
| $\omega_2$ | $10^{13}$ s$^{-1}$ | 2.315 | 3.641 |
| $Q$ | $10^{10}$ m$^{-1}$ | 1.0187 | 1.0945 |
| $a$ | Å | 4.295 | 3.997 |
| $v$ | ms$^{-1}$ | 3569 | 5226 |

Here $Q$ is the radius of the spherical zone. The volume of the Jones zone of both structures is $V_{Jz}=(2\pi/a)^3\sqrt{2}$, where $a$ is the length of the basic lattice vector. The ideal wurtzite structure with the ratio of the basic vector lengths $c/a=\sqrt{8/3}$ is assumed. We put $V_{Jz}=\frac{4}{3}\pi Q^3$. Also, $\omega_1=\omega_{\rm LO}(Q)$ and $\omega_2=\omega_A(Q)$ (zone boundary). The magnitudes of $\omega_1$ and $\omega_2$ are chosen such that an overall agreement is achieved with the available data on the zone boundary optical properties and sound velocity $v$.[18,30–32] The value of the acoustic frequency at the spherical zone boundary, obtained in this way, can be considered as plausible. The parameters of our dispersion model are given in Table I.

## V. THE PROCESS CONSIDERED

In experiments, as a result of the laser excitation of electrons over the band gap, an electron gas in the conduction band is obtained, with density $n_e$ and a certain temperature. After the termination of the laser pulse the electrons cool down while the phonon modes are heated up, thus having the mode temperatures above the temperature of the ambient lattice $T_L$. The whole system cools down finally to the temperature $T_L$.

It is important to recognize that the phonon modes become partly heated already in the course of the finite laser pulse duration. In order to simulate in a simple way the effect of the finite laser pulse duration and of the cooling process which follows after the termination of the pulse, we solve the kinetic equations for the following process: Namely, we assume that at $t=0$ the electrons have a density $n_e$, which is assumed to be independent of time, and a temperature $T_{e0}$, while all the phonon modes have the temperature $T_L$. We assume that the whole process consists of two periods of time: a heating period and a relaxation period. Thus, in the course of the heating period $0<t<t_p$, the length of which simulates here as the length of the laser pulse, the electrons are kept at a constant temperature $T_{e0}$, while the phonon modes are being heated up via the interaction among the electron and phonon subsystems. After the instant of time $t_p$, in the relaxation period $t>t_p$, the condition of the electronic temperature being constant is abandoned and the whole electron-phonon system is let to be relaxing spontaneously towards the equilibrium with the ambient lattice. Let us remark that in the experiments under consideration the pulse lengths of several hundreds of picoseconds are not expected to cause any substantial heating of the ambient lattice.[33]

## VI. NUMERICAL RESULTS AND DISCUSSION

The material parameters used in solution of the kinetic equation are compiled[34] in Table II. The lifetimes of the LO phonons $\tau_{\rm LO}^{(0)}$ and of the acoustic phonons $\tau_A$ (for CdSe) are taken from Refs. 2 and 6. For the lack of experimental data we have chosen $\tau_A=150$ ps for GaAs, which is the same value as in CdSe.

In CdSe the numerical calculations were performed taking $T_{e0}=2000$ K and $T_L=77$ K. The density of the conduction-band electrons is taken to be $n_e=1.1\times10^{24}$ m$^{-3}$ (see Ref. 6). At this density and under the conditions considered in this work the electrons remain nondegenerate. The distribution functions in this work are displayed in the form of the effective temperatures $T$ of the modes which are connected with the mode population by means of the Bose-Einstein distribution function. Thus, we introduce $T_q^{\rm LO}$ and $T_q^A$ as mode temperatures of optical and acoustic modes, respectively. In all the figures the temperatures $T$ are displayed as relative temperatures with respect to $T_L$. We introduce therefore $\Delta T_q^{\rm LO}=T_q^{\rm LO}-T_L$, $\Delta T_q^A=T_q^A-T_L$, and $\Delta T_e=T_e-T_L$, with obvious meaning.

### A. CdSe

Presenting the results for CdSe, let us first consider the evolution of the system during the heating period $0<t<t_p$. In Fig. 1 we display the temperature of the LO-phonon mode with the wave vector $q_m=1.2\times10^8$ m$^{-1}$ in the heating period for $t_p=200$ ps. The quantity $q_m$ is chosen here to be the magnitude of such a wave vector at which the mode temperature reaches approximately an overall maximum. The dashed horizontal line denotes the electronic temperature $\Delta T_e=T_{e0}-T_L$, which is constant within this period of time. We observe that the LO-phonon population sharply increases during the first several picoseconds and stabilizes after about 50 ps at a constant value.

As it is well known,[6] the LA phonons produced in the course of the decay of the hot LO-phonon population are concentrated in the close vicinity of a spherical surface in $\mathbf{q}$ space. The radius $q_0$ of the sphere, near which the LA-phonon population reaches the maximum, is given approximately by the condition $\omega_0=2\omega_A(q_0)$. In Fig. 1 we also display the LA-phonon temperature $\Delta T_{q0}^A$ taken at $q_0$ in this heating period. The acoustic-phonon temperature increases during about the first 50 ps. Also in Fig. 1 is the temperature $\Delta T_0^{\rm LO}$ of the LO-phonon mode with $q=0$ displayed as a function of time. The $q=0$ LO phonons are produced solely by the acoustic-phonon fusion. This is why the LO-phonon curve $\Delta T_0^{\rm LO}$ is slight-

TABLE II. Material parameters.

| Quantity | Unit | CdSe | GaAs |
|---|---|---|---|
| $m^*/m_0$ | | 0.13 | 0.067 |
| $\kappa_\infty$ | | 6.2 | 10.91 |
| $\kappa_0$ | | 9.6 | 12.91 |
| $\tau_{\rm LO}^{(0)}$ | ps | 4.4 | 8 |
| $\tau_A$ | ps | 150 | 150 |

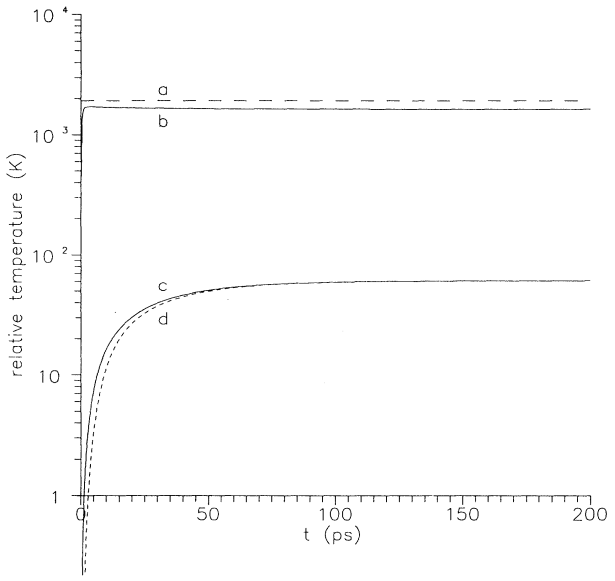


FIG. 1. The time dependence of the relative temperatures in CdSe in the heating period, computed at $T_{e0}=2000$ K, $T_L=77$ K, and $t_p=200$ ps. Curve $a$ (dashed line), electronic temperature $\Delta T_e$; curve $b$ (solid line), LO-phonon temperature $\Delta T_{q_m}^{\mathrm{LO}}$, $q_m=1.2\times10^8$ m$^{-1}$; curve $c$ (solid line), LA-phonon temperature $\Delta T_{q0}^{A}, q_0=68\times10^8$ m$^{-1}$; curve $d$ (dashed line), LO-phonon temperature $\Delta T_0^{\mathrm{LO}}$ at $q=0$.

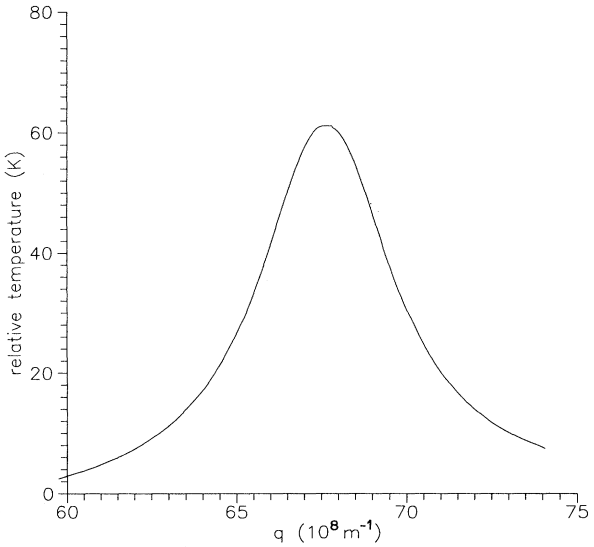


FIG. 3. $q$ dependence of the LA-phonon relative temperature $\Delta T_q^A$ at $t=200$ ps in CdSe. $T_{e0}=2000$ K and $T_L=77$ K.

ly retarded behind the LA-phonon curve. The time of retardation reflects the finite lifetime of the nonequilibrium LA-phonon population with respect to the fusion channel.

The $q=0$ LO-phonon mode becomes heated considerably after about the first 50 ps. From the reasons connected with the fusion processes, all the long-wavelength LO-phonon modes with approximately $q<0.2\times10^8$ m$^{-1}$ are heated to about the same temperature as the $q=0$ mode. This is seen in Fig. 2, giving the optical-phonon temperature $\Delta T_q^{\mathrm{LO}}$ at $t=t_p=200$ ps. In a more simple model with the hot LA phonons neglected, the heating of the long-wavelength LO phonons would be only due to the hot-electron cooling and thus $\Delta T_q^{\mathrm{LO}}$ would be negligible because of the energy and momentum conservation in the long-wavelength region of the optical phonons.

In the present case of CdSe the LA phonons are generated near $q=q_0=6.8\times10^9$ m$^{-1}$, as observed in Fig. 3, in which the LA-phonon temperature $\Delta T_q^A$ is given at $t=t_p=200$ ps. It is observed that the maximum value of the LA-mode temperature is close to that of the long-wavelength LO phonons. This is expected, realizing that

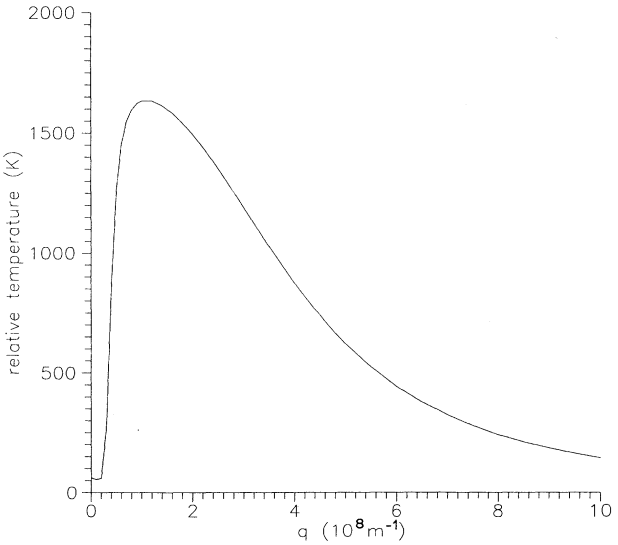


FIG. 2. $q$ dependence of LO-phonon relative temperature $\Delta T_q^{\mathrm{LO}}$ at $t=200$ ps (end of the heating period) in CdSe. $T_{e0}=2000$ K and $T_L=77$ K.

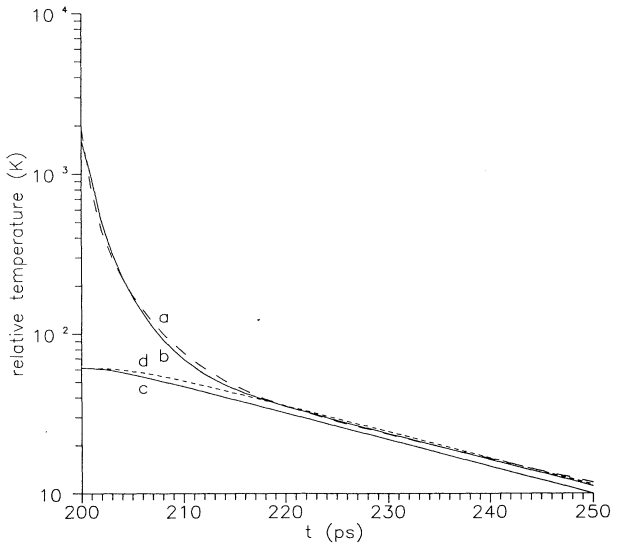


FIG. 4. Time dependence of relative temperatures in CdSe in the relaxation period ($t>t_p$). See caption of Fig. 1 for other data.

the mechanism of the generation of the long-wavelength LO phonons is the fusion of LA phonons.

After the termination of the heating period $(0,t_p)$, with $t_p=200$ ps, the electron-phonon system is left to relax towards $T_L$. In the relaxation period (Fig. 4), at about $t>220$ ps, the electronic temperature and the temperatures of the acoustic and of the long-wavelength optical phonons, together with $T_{qm}^{LO}$, closely follow each other. This shows that the hot-electron cooling is controlled by the cooling of the phonon system at this period of time and the acoustic phonon bottleneck effect is clearly displayed in this figure. The time evolution of $\Delta T_e$ then becomes exponential. At $t=250$ ps the time constant $\tau_e$ of the hot-electron cooling is 30.7 ps. Because the time constant $\tau_e$ is rather small in comparison with the input value of $\tau_A$, we conclude that the acoustic-phonon bottleneck effect is controlled by the fusion channel. Comparing that in the previous work,[17] performed with the dispersion neglected, we obtained $\tau_e=9.2$ ps.

In order that the present calculations may be compared more closely with the experiments,[5–8] we also performed the calculations for $t_p=30$ ps. The results are displayed in Fig. 5. The time constant of the hot-electron cooling is 26.4 ps at the time $t=80$ ps in this case.

Let us note that all the components of the whole system are cooling down with a constant relaxation time, at the temperatures which are well above $T_L$ in both cases of $t_p=200$ and 30 ps. The occurrence of the acoustic bottleneck effect is quite obvious in these results. This is found to be a contrast to the results of the previous work,[17] in which the bottleneck effect was not observed at temperatures reasonably high above $T_L$.

### B. GaAs

We performed similar calculations for GaAs taking $n_e=10^{22}$ m$^{-3}$, $T_{e0}=1000$ K, $T_L=77$ K, and $t_p=200$ ps. We have chosen somewhat lower values of $T_{e0}$ and $n_e$ than we did in the case of CdSe in order to avoid the degeneracy of the electron gas. The results, given in Fig. 6, displaying only the relaxation period, are quantitatively similar to those of CdSe, but the temperatures $\Delta T_0^{LO}$ and $\Delta T_{q0}^{A}$ come out much lower. With $q_0=60\times10^8$ m$^{-1}$ we obtained $\Delta T_{q0}^{A}=2.2$ K at $t_p=200$ ps. This is too low a value of the relative temperature for a significant acoustic-phonon bottleneck effect to be observed.

The relaxation time of the electrons is $\tau_e=12.5$ ps at $t=250$ ps, deduced from the Fig. 6. The relaxation time of acoustic phonons at the same instant of time is 36.7 ps. The calculation shows that the development of the acoustic-phonon bottleneck can be expected to appear at too low values of $\Delta T_e$, in GaAs and under the conditions considered here.

In accordance with the expectations expressed in our previous paper, the relaxation times of the electron cooling in CdSe come out larger than those we obtained with the help of our previous model. In this material and under suitable conditions, one can observe the acoustic-phonon bottleneck effect, at which the whole system cools down with a constant relaxation time already at reasonably high temperatures above $T_L$. We ascribe the observed difference between the previous theoretical results[17] and the present ones to both the inclusion of the realistic dispersion of the both branches of the phonon modes and to the introduction of the finite pulse length represented here by the heating period.

In particular, the fusion channel is now partly closed. This can be ascribed first of all to the significant depen-

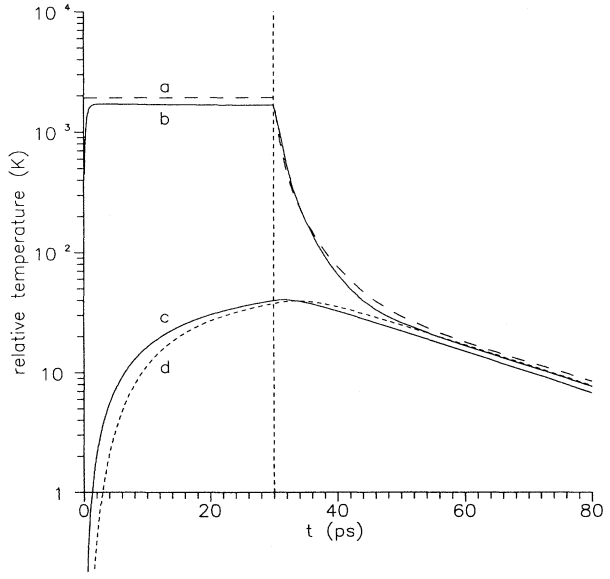


FIG. 5. The time dependence of the relative temperatures in CdSe in both the heating and relaxation periods of time, for $t_p=30$ ps. $t_p$ is marked by the vertical dashed line. See caption of Fig. 1 for other data.

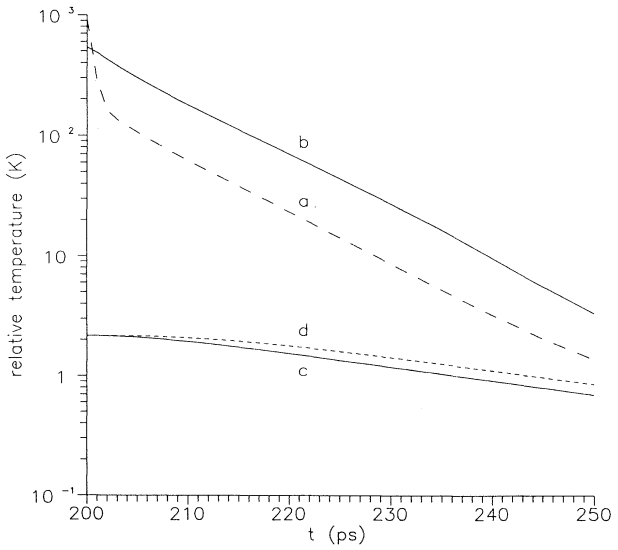


FIG. 6. The relative temperatures in the relaxation period in GaAs. $t_p=200$ ps, $q_m=0.7\times10^8$ m$^{-1}$, $q_0=60\times10^8$ m$^{-1}$, $T_{e0}=1000$ K, and $T_L=77$ K. See caption of Fig. 1 for other data.

dence of the LO-phonon energy on $q$. In this case the energy conservation law brings a restriction of the $q$ space available for the products of the fusion channel, although the fusion channel remains open to such an extent that the hot-electron cooling time is still lower than the observed experimental values[5–8] reported to be of the order of hundreds of picoseconds. The inability of the present theory to approach the experimental values of the hot-electron cooling rates could be partly ascribed to the choice[23,24] of the coupling function $\phi$ in Klemens interaction operator, which does not seem to be understood enough at present.

As far as the inclusion of the finite pulse length simulation is concerned, the significance of it appears to be comparable with the importance of the dispersions. We conclude, therefore, that the laser pulse length, which is reported in Refs. 5–8 to be about 30 ps, may be an important factor in numerical simulation of those relaxation processes, which should lead to large relaxation times. In other words, the amount of the energy provided to the electronic subsystem by the laser pulse may thus be a significant factor.

The numerical calculation shows that the results are sensitive to some extent to the magnitude of the spherical zone radius. This is not a surprise because the magnitudes of $q_0$ are rather large on the scale of the zone radius. In this context the importance of umklapp processes is unresolved as yet.

Summing up, we have solved the kinetic equations for the system of hot electrons and hot optical and acoustic phonons, taking into account the anharmonic coupling between the optical and acoustic phonons and realistic dispersions of both. The conditions were found to be more favorable for the occurrence of the acoustic-phonon bottleneck in the case of the realistic dispersions and finite laser pulse lengths included than in the case of the dispersionless approximation and short pulses. The relaxation time constant in CdSe, being about 30 ps, is larger than the value of 9.2 ps obtained previously in the dispersionless approximation, although it is still lower than the experimentally observed time constants. The value of the relaxation time constant obtained in the present case appears to be controlled by the fusion processes of the acoustic phonons.


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