Ballistic propagation of interface optical phonons

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We discuss a spatial propagation of interface *optical* phonons in semiconductor heterostructures. Starting from a quantum kinetic equation for the nondiagonal phonon density operator, we derive the wave-packet presentation for the phonon amplitude for the pure initial phonon state. For small wave vectors, the group velocity determining the speed of the packet can exceed substantially the speed of sound and the wave front can propagate a distance of several micrometers before decay of the optical phonons. The possibility of experimental observation of the effect is discussed.

The propagation of acoustic waves and phonons has been studied extensively and has found numerous practical applications. Recently a number of experiments have been performed on the generation and propagation of coherent acoustic phonons with energies in the meV range.[1] On the other hand, the *optical* phonons practically do not propagate because of their weak dispersion. In fact, optical phonon frequencies ω_k are changed substantially for the wave vectors $k \sim \pi/d$, where d is a lattice constant, and the optical phonons could propagate only at few lattice constants before their decay.

The situation is different for heterostructures where the optical phonons can be quantized.[2] While confined and semibound phonon modes have practically the same constant frequencies as the materials to which they are confined, there also exist *interface* optical phonon modes with frequencies intermediate between those of the materials forming the interface. In heterosystems with a characteristic distance a, such as multiple heterostructure (a is the separation between the interfaces) or a quantum wire (a is the diameter), the phonon frequency changes substantially as the wave vector k changes from zero to $1/a$. Therefore, the interface optical phonons can possess comparatively high group velocities of order of $\Delta\omega a$.

In this paper, we consider theoretically the effect of a free spatial propagation of the interface optical phonons in semiconductor double heterostructures. Our results call attention to the possibility of the high-speed *propagation* of the interface phonons in spatially inhomogeneous structures. Furthermore, this treatment suggests that interface phonon propagation velocities are large enough to cause a substantial modification in the temporal evolution of the electronic and optical processes in heterostructures.

We consider the evolution of a spatially localized distribution of phonons with the dispersion $\omega_{\mathbf{k}}$. In order to obtain an adequate description of phonon propagation we derive the kinetic equation for the phonon density matrix in the plane wave basis. It is important to note that the inhomogeneous distribution of phonons is governed by *nondiagonal* components of the density operator.

The Hamiltonian of noninteracting phonons is equal to

$$\mathcal{H} = \sum_{\mathbf{k}} \hbar\omega_{\mathbf{k}}\, b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} \,, \tag{1}$$

where $b_{\mathbf{k}}^{\dagger}$ and $b_{\mathbf{k}}$ are phonon creation and annihilation operators. The equation of motion for the phonon microscopic density operator

$$\hat{n}_{\mathbf{k}\mathbf{k}'} = b_{\mathbf{k}'}^{\dagger} b_{\mathbf{k}} \tag{2}$$

is given by

$$i\hbar\, \frac{\partial \hat{n}_{\mathbf{k}\mathbf{k}'}}{\partial t} = [\hat{n}_{\mathbf{k}\mathbf{k}'},\, \mathcal{H}] \,. \tag{3}$$

Substituting Eqs. (1) and (2) into (3) and performing the necessary commutations, we find the kinetic equation for the density operator $\hat{n}_{\mathbf{k}\mathbf{k}'}$:

$$i\, \frac{\partial \hat{n}_{\mathbf{k}\mathbf{k}'}}{\partial t} = (\omega_{\mathbf{k}} - \omega_{\mathbf{k}'})\, \hat{n}_{\mathbf{k}\mathbf{k}'} \,. \tag{4}$$

Note that Eq. (4) describes the kinetics of any noninteracting boson or fermion system described by the Hamiltonian in Eq. (1).

Introducing the ensemble-averaged macroscopic density matrix $n_{\mathbf{k}\mathbf{k}'} \equiv \langle \hat{n}_{\mathbf{k}\mathbf{k}'} \rangle$, where the average is taken over the phonon ensemble, we write the solution of Eq. (4) in the form

$$n_{\mathbf{k}\mathbf{k}'}(t) = n_{\mathbf{k}\mathbf{k}'}(0)\, e^{i(\omega_{\mathbf{k}} - \omega_{\mathbf{k}'})t} \,. \tag{5}$$

Subsequently, the observable phonon density (per unit

volume or area) is given by the diagonal part of the density matrix in the coordinate representation

$$N(\mathbf{r},t) = n(\mathbf{r},\mathbf{r}',t) = \mathcal{L}^{-\nu} \sum_{\mathbf{k},\mathbf{k}'} e^{i\mathbf{r}(\mathbf{k}'-\mathbf{k})}\, n_{\mathbf{k}\mathbf{k}'}(t) \,. \quad (6)$$

Here $\nu = 1,2,3$ is the dimensionality of the system and $\mathcal{L}$ is a normalization length.

Equations (5) and (6) give the complete description of phonon evolution from any initial state $n_{\mathbf{k},\mathbf{k}'}(0)$. In this study, we limit ourselves by the consideration of a *pure* initial state and choose the initial density matrix in the form

$$n_{\mathbf{k}\mathbf{k}'}(0) = n^*_{\mathbf{k}}(0)\, n_{\mathbf{k}'}(0) \,. \quad (7)$$

Substitution of Eq. (7) into Eqs. (5) and (6) produces the following result for the phonon density:

$$N(\mathbf{r},t) = |u(\mathbf{r},t)|^2 \,, \quad (8)$$

where the phonon amplitude is given by a wave packet of the plane wave components

$$u(\mathbf{r},t) = (2\pi)^{-\nu} \int d^\nu k\; u_0(\mathbf{k})\, e^{i(\mathbf{k}\cdot\mathbf{r}-\omega_{\mathbf{k}}t)} \quad (9)$$

with the spectral density equal to $u_0(\mathbf{k}) = \mathcal{L}^{\nu/2} n_{\mathbf{k}}(0)$.

We normalize the density matrix with respect to the total energy E injected into the system:

$$\hbar \sum_{\mathbf{k}} \omega_{\mathbf{k}} n_{\mathbf{k}\mathbf{k}} = E \,. \quad (10)$$

Introducing the mean phonon energy $\bar{\omega}$ by

$$\bar{\omega} \equiv \sum_{\mathbf{k}} \omega_{\mathbf{k}} n_{\mathbf{k}\mathbf{k}} \Big/ \sum_{\mathbf{k}} n_{\mathbf{k}\mathbf{k}} \,,$$

we can rewrite Eq. (10) in the form

$$\sum_{\mathbf{k}} n_{\mathbf{k}\mathbf{k}} = N_0 \,, \quad (11)$$

where $N_0 = E/\hbar\bar{\omega}$ is the actual number of phonons created. In terms of the phonon density $N(\mathbf{r},t)$, the normalization condition given in Eq. (11) takes the form

$$\int d^\nu r\; N(\mathbf{r},t) = N_0 \,. \quad (12)$$

Let us analyze in more detail the asymptotic behavior of the phonon amplitude $u(\mathbf{r},t)$ at a large time t in the case of two-dimensional propagation ($\nu = 2$). Rewriting Eq. (9) in polar coordinates we obtain

$$u(\mathbf{r},t) = (2\pi)^{-2} \int_0^\infty k\, dk \int_0^{2\pi} d\varphi\; u_0(k,\varphi)\, e^{it\psi(k,\varphi)} \,, \quad (13)$$

where the phase in the exponent is given by

$$\psi(k,\varphi) = \xi k \cos\varphi - \omega(k) \quad (14)$$

and the vector $\boldsymbol{\xi} = \mathbf{r}/t$ is introduced to facilitate the limiting transition $t, r \to \infty$, while r/t is a constant. At a large time t the dominant contribution to the double integral in Eq. (13) occurs in the vicinity of the stationary phase points, where

$$\frac{\partial\psi}{\partial\varphi} = 0 \,, \qquad \frac{\partial\psi}{\partial k} = 0 \,. \quad (15)$$

These conditions given in Eq. (15) imply $k = k_\xi$, where k_ξ is a solution of equation

$$|\omega'(k_\xi)| = \xi \quad (16)$$

and $\varphi = 0$ ($\varphi = \pi$) for an increasing (decreasing) dispersion $\omega(k)$.

We consider two most important cases corresponding to the saddle points of the phase ψ: (i) $\omega'(k) > 0$ and $\omega''(k) < 0$ and (ii) $\omega'(k) < 0$ and $\omega''(k) > 0$. In the first case the Hessian of the phase is equal to $\psi'' = \xi k_\xi\, \omega''(k_\xi)$ and the phonon amplitude at $t \to \infty$ is equal to

$$u(\mathbf{r},t) = \frac{u_0(k_\xi,0)}{2\pi} \sqrt{\frac{k_\xi}{rt|\omega''(k_\xi)|}}\; e^{i[k_\xi r - \omega(k_\xi)t]} \,; \quad (17)$$

the amplitude for the second case can be obtained from Eq. (17) by changing $u_0(k_\xi,0)$ to $u_0(k_\xi,\pi)$. For an isotropic initial phonon distribution $u_0(k,\varphi) = u_0(k)$, the phonon distribution function of Eq. (8) then reduces to

$$N(\mathbf{r},t) = \frac{k_\xi\, |u_0(k_\xi)|^2}{(2\pi)^2\, rt\, |\omega''(k_\xi)|} \,. \quad (18)$$

To illustrate the applications of the obtained expressions, we consider the propagation of interface phonons in a double heterostructure with a distance a between the interfaces. The inner material has dielectric function $\varepsilon_1(\omega)$ and the outer ones are characterized by the dielectric function $\varepsilon_2(\omega)$, where

$$\varepsilon_i(\omega) = \varepsilon_{\infty i}\, \frac{\omega^2 - \omega_{Li}^2}{\omega^2 - \omega_{Ti}^2} \,, \quad (19)$$

$i = 1,2$ is a material index, $\varepsilon_{\infty i}$ stand for the high-frequency dielectric constants, and ω_{Li} and ω_{Ti} are the longitudinal and tranverse bulk phonon frequencies in the corresponding materials. In the dielectric continuum model,[2] the frequencies of the two *symmetric* interface phonon modes $\omega_{S\pm}$ are found from the solutions of the dispersion equation

$$\varepsilon_1(\omega) \tanh\frac{ka}{2} + \varepsilon_2(\omega) = 0 \quad (20)$$

and the dispersion relation for two *antisymmetric* modes $\omega_{A\pm}$ is obtained from Eq. (20) upon interchanging subscripts 1 and 2.

We choose the initial phonon spectral density in the form of Gaussian with effective width δ:

$$u_0(k) = \sqrt{4\pi N_0}\; \delta\, \exp(-\delta^2 k^2/2) \,. \quad (21)$$

The corresponding initial spatial distribution is also a Gaussian,

$$N(\mathbf{r},0) = (N_0/\pi\delta^2)\exp(-r^2/\delta^2)\ . \qquad (22)$$

Note that $u_0(k)$ and $N(\mathbf{r},0)$ in Eqs. (21) and (22) are normalized to the total number of created phonons N_0 according to conditions in Eqs. (11) and (12). Substituting the spectral density in Eq. (21) into Eq. (18), we find the final expression for the evolution of the Gaussian distribution in the limit of large time t:

$$N(\mathbf{r},t) = \frac{N_0\delta^2}{\pi}\,\frac{k_\xi\exp(-\delta^2k_\xi^2)}{rt\,|\omega''(k_\xi)|}\ . \qquad (23)$$

Here $\xi = r/t$ and k_ξ is defined by Eq. (16).

For numerical calculations we choose the parameters of the AlAs/GaAs/AlAs double heterostructure:[2] $\omega_{L1} = 36.2$ meV, $\omega_{T1} = 33.3$ meV, $\omega_{L2} = 50.1$ meV, $\omega_{T2} = 44.8$ meV, $\varepsilon_{\infty1} = 10.8$, and $\varepsilon_{\infty2} = 8.16$. In Fig. 1 we present (a) the frequencies $\omega_j(k)$ and (b) the group velocities $\omega_j'(k)$ of the interface phonon modes. The subscript j denotes four existing interface phonon modes in the double heterostructure, $j = S+, S-, A+$, and $A-$. Note that the group velocities $\omega_j'(k)$ are proportional to the distance a between the interfaces. As a is increased, the frequencies $\omega_j(k)$ will approach the values $\omega_\pm$ of the single heterostructure everywhere except in the wave vector region $k \lesssim 1/a$. Therefore, an increase of the separation distance a leads to the narrowing of the spectral region of the propagating waves with a simultaneous increase in the propagation velocities $\omega_j'(k)$. For a heterostructure with a on the order of a few hundred angstroms, the group velocities of the interface phonon modes can be substantially larger than the sound velocity of the material.

Figure 2 demonstrates the spatial phonon distribution function $N_j(\mathbf{r},t)$ for four interface modes j at time $t =$ 10 ps. In accordance with Fig. 1(b) the fastest modes are $S+$ and $A+$, while $S-$ is the slowest one. Figure 3(a) presents the spatial distribution for the fastest $S+$ mode at initial moment t=0 and t =5, 7, and 10 ps. The peak values gradually become smaller with time due to the propagation in a two-dimensional structure as indicated

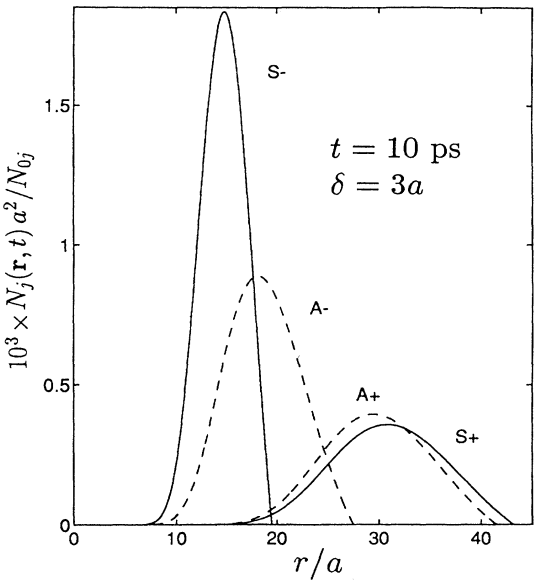

FIG. 2. Dimensionless phonon densities $N_j(\mathbf{r},t)\,a^2/N_0$ for the interface modes versus dimensionless distance r/a. Time $t = 10$ ps; initial wave-packet width $\delta = 3a$.

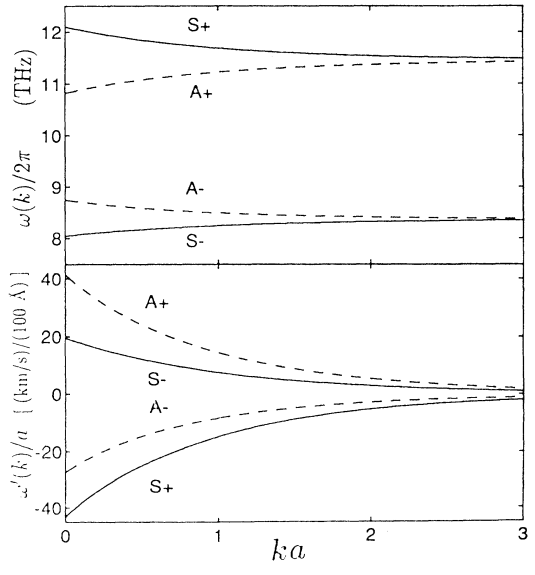

FIG. 1. (a) Cyclic frequencies $\omega_j/2\pi$ and (b) normalized group velocities $(d\omega_j/dk)/a$ for interface phonons versus dimensionless wave vector ka. Solid and dashed lines correspond to $j = S+, S-$ and $j = A+, A-$, respectively.

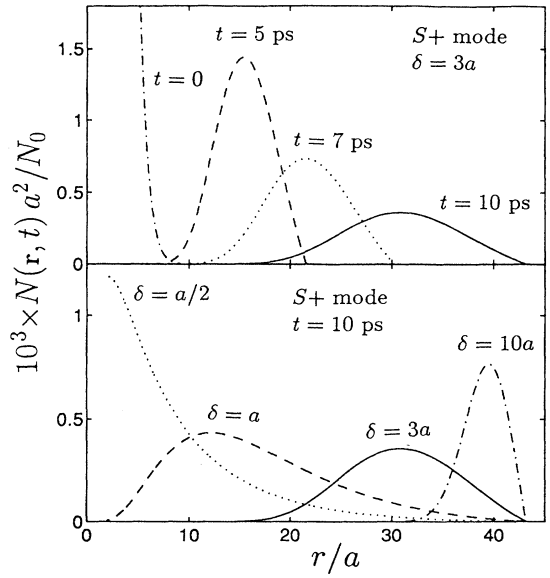

FIG. 3. Dimensionless phonon density $N(\mathbf{r},t)\,a^2/N_0$ for the $S+$ mode versus dimensionless distance r/a. (a) Time $t = 0, 5, 7$, and 10 ps; initial wave-packet width $\delta = 3a$. (b) Time $t = 10$ ps and $\delta = a/2, a, 3a$, and $10a$.

in Eq. (12). Figure 3(b) illustrates the influence of the initial width δ on the form of the wave packet for the $S+$ interface mode. An interesting point to note is the fact that the spatial spread of wave packet at $t = 10$ ps is smaller with a larger δ (see $\delta = 10a$). This is because the wave packet with a larger initial width in real space has a smaller spread in momentum space. As δ increases, the partial contribution of wave components with small q having velocities close to the maximal one $\omega'(0)$ increases and the wave packet becomes more concentrated near the wave front at $r = \omega'(0)\, t$.

Let us discuss now the possibility of the experimental observation of the proposed effect. The spatially localized phonon wave packet can be generated due to emission by hot electrons,[3] directly by a laser beam in a pump-and-probe Raman-type experiment,[4] or by the impulse method.[5] Since these methods involve sample illumination by a spatially localized light beam, the width of the packet cannot be substantially smaller than one-half of the light wavelength $\lambda/2$.[6] For an experimental observation, the propagation distance of the phonons should exceed the initial spread of the packet, therefore, the distance a between the interfaces should be comparable to $\lambda/2$. Note that only the long-wavelength phonons with $k \lesssim 1/a$ propagate substantially.

In reality the propagation of the optical phonons is limited by their decay and scattering. Since we consider long-wavelength phonons, the scattering by short-scale structural imperfections is negligible and the propagation length is limited by the decay of the interface optical phonon into two acoustic phonons with a characteristic decay time τ of the order of that for the bulk optical phonons:[7] $\tau_{\text{bulk}} \sim 10$ ps. Taking the AlAs/GaAs/AlAs double heterostructure with the distance between the interfaces $a = 1000$ Å, we find that the group velocity for the $S+$ interface mode is $d\omega/dk \sim 400$ km/s for the phonon wave vectors $k \lesssim 10^5$/cm. Therefore the wave front of the packet will propagate a distance of a few micrometers before its decay.

Another possibility for an experimental verification of the effect is a time-resolved observation of emitted secondary acoustic phonons. Because the acoustic phonons are generated by the decaying interface phonons which propagate at large speed, the secondary acoustic phonons will reach an observation point at times smaller than that for an acoustic wave propagating from the excitation source. Other approaches may include a strongly localized one-dimensional wave packet of interface phonons that can be generated by a one-dimensional electron gas confined vertically between the interfaces and horizontally by electrostatic modulation of the potential by a corrugated gate. An exchange of the interface optical phonons between two one-dimensional electron gases can provide an efficient mechanism of the momentum and energy transfer between two spatially separated electron subsystems.[8] The effect of the mutual drag between electron gases in parallel quantum wells due to the Coulomb interaction and acoustic phonons exchange has been experimentally observed in Ref. 9.

Finally, we discuss the limiting transition to large distances between the interfaces, $a \to \infty$. With an increase of a the phonon frequencies tend to their limits for the single interfaces $\omega_{\pm}$ and the group velocities tend to zero for all wave vectors k except the vanishing region $k \lesssim 1/a$. The group velocity of phonons from this region increases proportional to a. Since the dispersion relations for the interface phonons were derived on the assumption of a static electric field,[2] our consideration is limited by phonon speed much less than the speed of the electromagnetic wave in the material. Since the scattering of the long-wavelength phonons by the uncontrollable short-scale lattice imperfections is small, the broadening of the phonon dispersion curve is of the order of the inverse decay time $\tau \sim 10$ ps and the group velocity $d\omega/dk$ remains well defined in the limit of small k. We note, however, that with an increase of the separation distance a, the fraction of the propagating interface phonons will decrease as $1/a$ in comparison with the nonpropagating confined and half space optical phonons.

In conclusion, we have theoretically considered the effect of the propagation of dispersive interface optical phonons in inhomogeneous structures. Our estimations show that the long-wavelength interface phonons can propagate a distance of several micrometers before the decay and the effect can lead to a modification of the temporal behavior of an electron system.

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