Phonon-driven carrier transport caused by short excitation pulses in semiconductors

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We propose a simple model describing phonon-wind-driven transport of photoexcited carriers in bulk and in multiple-quantum-well (MQW) semiconductors. We take into account diffusion of carriers and primary acoustical phonons, generated at the final stage of the carrier's relaxation, and show that their simultaneous action gives fast (even supersonic at higher excitation intensities) transport of photoexcited carriers. The phonon retardation effects are very essential in our approach. It is shown that this model may be applicable to recent experiments of Wolfe and collaborators [D. W. Snoke *et al.*, Phys. Rev. B **41**, 11 171 (1990); **44**, 12 109(E) (1991); L. M. Smith *et al.*, *ibid.* **39**, 1862 (1989)] on fast transport of excitons in Cu_2O and $Al_xGa_{1-x}As$-GaAs MQW's.

I. INTRODUCTION

A number of interesting effects arise due to nonequilibrium phonons generated in semiconductors during the relaxation and recombination of photoexcited carriers and propagated ballistically at low temperatures. Nonequilibrium phonon-driven transport of carriers (i.e., the so-called phonon wind) was thoroughly investigated in Ge and Si, where the carriers condense into heavy macroscopic droplets of electron-hole liquid, and these phonon-wind effects are not screened by diffusion. Striking crystal anisotropy effects in propagation of ballistic phonons (phonon focusing) and the behavior of slowly decaying phonon "hot spot" in the area of photoexcitation were also investigated. Detailed reviews of this research have been given in Refs. 1–3.

The sequence of events which eventually leads to the appearance of low-energy ballistic acoustic phonons is very complicated. If the energy of the photoexcited carriers is much higher than the energy of an optical phonon, $\varepsilon \gg \hbar\omega_{\rm opt}$ (e.g., in case of surface excitation), the thermalization begins with the emission of a cascade of optical phonons which lasts 10^{-13}–10^{-11} s. The remaining energy $\varepsilon^{\rm PW} < \hbar\omega_{\rm opt}$ is emitted in the form of low-energy acoustic phonons, and this process lasts 10^{-11}–10^{-10} s. These phonons can propagate ballistically at low temperatures, they interact strongly with carriers, and they are the primary source of the phonon wind. Ballistic phonons may also arise from the down-conversion of high-energy phonons of hot spot (which we will call "secondary" to distinguish them from "primary" phonons which arise directly during the thermalization). This secondary phonon wind is much more effective than the primary one in experiments with steady-state excitation. But in the case of short excitation pulses, secondary phonons appears a long time after the primary phonons, in 10^{-7}–10^{-6} s, and only primary phonons may accelerate carriers up to sonic velocities for the time of the order of the carrier's relaxation time.

Most of the experiments with phonon wind and electron-hole droplets were explained by means of the "electrostatic" analogy, proposed by Keldysh[5] and generalized for the accounting of phonon focusing effects in Ref. 6. But for the treatment of fast sonic transport at short excitation pulses, retardation effects were important. A very simple model was proposed[7,8] by one of us (see also Ref. 3). This model agreed well with the experimental data[7] on the fast expansion of the electron-hole-droplet cloud in Ge. It also explained the behavior of nonequilibrium carriers in Ge subjected to a short microwave heating pulse.[8] But this model worked for short excitation pulses only, $\tau_i \ll l/S$ (l is the minimal initial dimension of the carriers cloud; and S, the velocity of sound), and the diffusion and lifetime of carriers were not taken into account.

Recently, two groups of experiments were reported which demonstrated rapid expansion of carriers after short excitation pulses: in Cu_2O (Refs. 9 and 10) and in $Al_xGa_{1-x}As$-GaAs multiple quantum wells (MQW's).[11] In the first group of experiments 10-ns pulses of surface excitation were used, and the authors' goal was to obtain a dense Bose gas of excitons which may undergo Bose-Einstein condensation (BEC). The conditions for equilibrium BEC were nearly met in these experiments (see also Ref. 12), and the authors attributed the fast exciton transport in their experiments to the superfluidity of the BEC. The second group of experiments deals with in-plane transport of excitons, generated in thin (1-μm) sample of $Al_xGa_{1-x}As$-GaAs MQW's by 6-ps pulses. This transport was interpreted as the phonon-wind-enhanced diffusion, but the retardation effects were not taken into account.

The purpose of this paper is to generalize the model of Refs. 7, 8, and 3, taking into account the diffusion of carriers, their finite lifetime, and the finite duration of excitation pulses. We neglect crystal anisotropy and take into account only one sort of phonons, e.g., LA phonons. It is shown that the simultaneous action of the diffusion and

the phonon wind may cause even supersonic expansion of the carrier's cloud at higher excitation intensities (provided that the thermal velocity of the carriers or the excitons is higher than that of sound). It is shown that fast transport of excitons[9,10] in Cu_2O may be described within this model without assuming their superfluidity. It is remarkable that the principal adjustable parameter of our theory, $\sigma_{ph}\varepsilon_{PW}$ (where σ_{ph} is the exciton-phonon scattering cross section), obtained by fitting our theory to the experiments in Cu_2O, appears to be of the same order of magnitude as in earlier experiments[7,8,3] in Ge, $\sigma_{ph}\varepsilon_{PW}\approx 2\times 10^{-15}$ meV cm^2. And this could have been expected because the electron-phonon interaction, which governs both σ_{ph} and ε_{PW}, does not change appreciably from one substance to another.

As to the experiments in MQW's, our model predicts fast diffusion-assisted expansion at excitation intensities very close to that applied in experiments,[11] if we use $\sigma_{ph}\varepsilon_{PW}\approx 10^{-15}$ meV cm^2—that is, of the same order of magnitude as in other semiconductors.

II. PHONON WIND CAUSED BY PRIMARY THERMALIZATION PHONONS

In order to describe the primary phonon flux, let us assume that the generation of phonons just follows the generation of carriers, and energy ε_{PW} per excitation quanta goes into ballistic phonons. Then we can easily write the following equation for the momentum flux of primary acoustic phonons, which propagate ballistically with sound velocity:

$$\mathbf{W}(\mathbf{r},t)=\frac{1}{4\pi}\frac{\varepsilon_{PW}}{S}\int_V \frac{\mathbf{r}-\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|^3} g\left[\mathbf{r}', t-\frac{|\mathbf{r}-\mathbf{r}'|}{S}\right] d^3r' . \tag{1}$$

Here V is the excitation volume and $g(\mathbf{r},t)$ is the carriers generation rate. Deriving Eq. (1) we assumed that there is no coherency between phonons. It was mentioned above that the risetime of primary acoustic phonons $\tau_{ph}\approx 10^{-10}$ s. As a result, Eq. (1) holds only for time and space intervals larger than τ_{ph} and $S\tau_{ph}$ ($\approx 10^{-10}$ s and 10^{-5} cm, correspondingly). But we do not need higher accuracy when describing the expansion of the carrier's cloud. And if the excitation pulse is shorter than τ_{ph}, we may use a δ-function representation for the generation rate, $g(\mathbf{r},t)=g(\mathbf{r})\delta(t)$. It greatly simplifies the calculations. For example, for surface excitation,

$$g(\mathbf{r},t)=g_s e^{-\lambda z}\delta(t) , \tag{2}$$

where z is the coordinate, normal to the excitation surface, λ is the absorption coefficient of the pumping wave,

$$g_s=\frac{\lambda E}{\hbar\omega} ,$$

E and $\hbar\omega$ are the energy of excitation pulse (per cm^2) and the energy of excitation quantum. Then $\mathbf{W}$ is normal to the excitation surface and takes the form

$$W_z(z,t)=\frac{g_s\varepsilon_{PW}}{S}\begin{cases}[\tau\cosh(\tau)-\sinh(\tau)]e^{-\zeta}, & 0<\tau<\zeta\\ 2\zeta-(\tau+1)e^{-\tau}\sinh(\zeta), & \zeta<\tau .\end{cases} \tag{3}$$

Here $\tau=\lambda St$ and $\zeta=\lambda z$ are dimensionless time and distance. We assumed here that all phonons reflect elastically from the surface $z=0$.

For experiments with thin MQW samples we may assume

$$g(\mathbf{r},t)=g_{MQW}e^{-r^2/R^2}\delta(t) , \tag{4}$$

where r is the in-plane distance from the pumping beam axis,

$$g_{MQW}=\frac{E_{abs}}{d\hbar\omega} , \tag{5}$$

d is the width of the sample, and E_{abs} is the absorbed energy (per cm^2, the spacial profile of the beam assumed to be Gaussian). Then $\mathbf{W}$ is normal to the beam axis and

$$W_r(r,t)=g_{MQW}\frac{g_s\varepsilon_{PW}}{S}e^{-\rho^2}\int_0^1 dx\, e^{-x\tau^2}\frac{x^2}{\sqrt{1-x^2}}I_1(2\rho\tau x) , \tag{6}$$

where $\rho=r/R$, $\tau=St/R$, and I_1 is the Bessel function.

Using Eqs. (3) and (6) as retarded Green's functions, we can easily obtain $\mathbf{W}$ for finite excitation pulses. For example, if

$$g(\mathbf{r},t)=\frac{g_s}{\tau_i}e^{-\lambda z}e^{-t^2/\tau_i^2} , \tag{7}$$

and the duration of pulse is large, $A=\tau_i\lambda S\gg 1$, we get the following expression for phonon flux in the region ζ, $\tau\ll A^2/2$:

$$W(z,t)=\frac{g_s\varepsilon_{PW}}{\sqrt{\pi}\lambda S^2\tau_i}\left\{\frac{1-e^{-\zeta^2}}{\zeta}e^{-(\zeta-\tau)^2/A^2}-2e^{-\tau^2/A^2}+2\zeta A\int_{(\zeta-\tau)/A}^{\infty}dx\frac{e^{-x^2}}{(Ax+\tau)^2}\right\} . \tag{8}$$

III. PHONON-DRIVEN TRANSPORT OF CARRIERS

The expansion of carriers cloud under the influence of diffusion and phonon wind is described by the following transport equation:

$$\frac{\partial n}{\partial t}+\mathrm{div}\mathbf{V}n=D\Delta n-\frac{n}{\tau_c} . \tag{9}$$

Here τ_c and D are the lifetime and the diffusion constant of carriers, $\mathbf{V}(\mathbf{r},t)$ is the drift velocity of carriers caused by the phonon wind. It may be defined from the equation

$$\sigma_{ph}\left[1-\frac{V}{S}\right]\mathbf{W}=\frac{m}{\tau_r}\mathbf{V} , \tag{10}$$

which is equivalent to the following one:

$$\mathbf{V}(\mathbf{r},t)=S\frac{\alpha\mathbf{W}}{1+\alpha W} , \tag{11}$$

where $\alpha=\tau_r\sigma_{ph}/Sm$ is the carrier–phonon-wind coupling constant at $V\ll S$. Here m and τ_r are the mass and momentum relaxation time of e-h pair, the right-hand side of Eq. (10) contains the force influenced by each pair, and the left-hand side takes into account the fact that the phonon drag velocity may not exceed the sound velocity. We are using this approximation following Ref. 11. Of course, the explicit dependence of σ_{ph} and τ_r on V is much more complex. Equation (10) gives a correct trend for dependence of V on W [see Eq. (11)]: a linear one at small W and a saturation at $V\approx S$ at large W. We would like to add that numerical calculations show that the solutions of the transport equation (9) do not depend significantly on the precise form of $V(W)$.

Typical solutions of Eq. (9), calculated for surface excitation with **W** given by Eq. (8), are shown in Fig. 1. In order to compare our model with experimental data[9,10] for Cu_2O, we take $D=600$ cm^2/s, $\lambda=1.6\times10^4$ cm^{-1}, $\tau_i=6$ ns, $S=4.5\times10^5$ cm/s ($A\approx40$ for these values of parameters). Figures 1(a)–1(c) were calculated with different dimensionless excitation energy $a=E/E_s$, where

$$E_s=\frac{2\sqrt{\pi}m\hbar\omega S^2\tau_i}{\tau_r\sigma_{ph}\varepsilon_{PW}}. \tag{12}$$

For lower excitation energy the expansion is purely diffusive, but a rapidly moving bump appears with increasing excitation energy. In the limit of $E/E_s\gg1$, we get from Eq. (11) that

$$V(\mathbf{r},t)=S \tag{13}$$

and then the solution of Eq. (9) takes the form

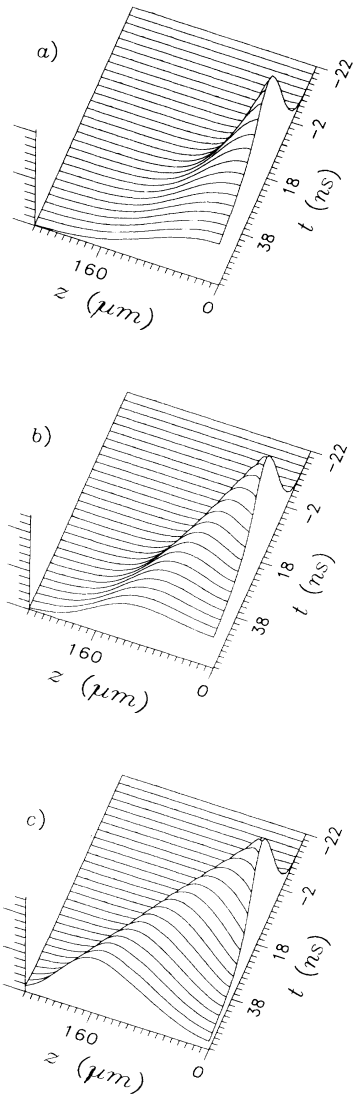

FIG. 1. The concentration of carriers $n(z,t)$, for $D=600$ cm^2/s and for different values of the dimensionless excitation energy $a=E/E_s$: (a) 0.1, (b) 1.0, and (c) 10.0.

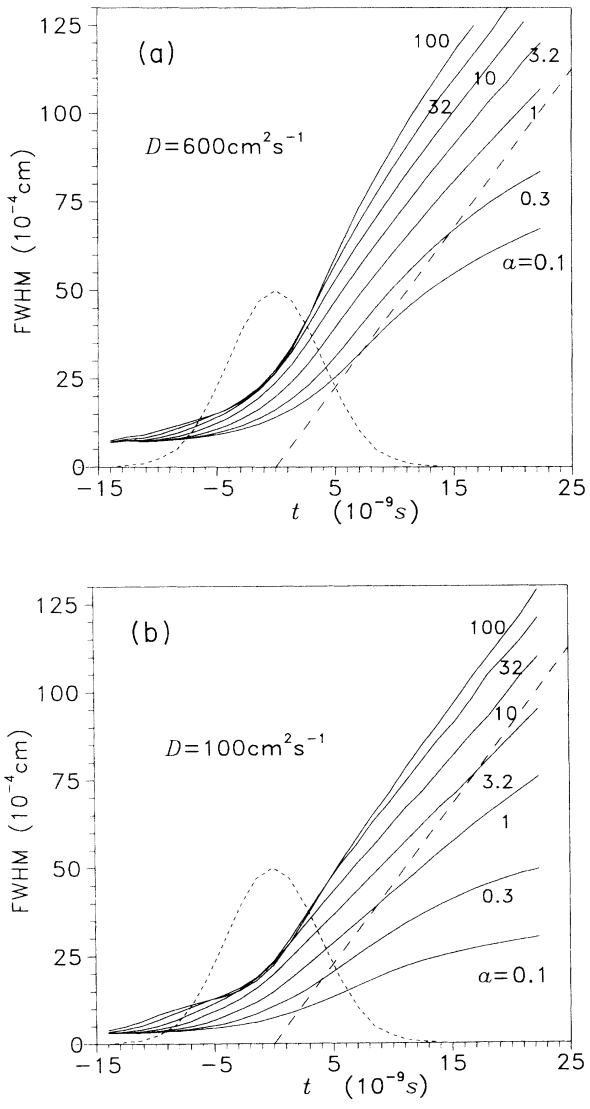

FIG. 2. The time dependence of the FWHM of the carrier concentration $\Delta(t)$ for different values of dimensionless excitation energy $a=E/E_s$ in case of surface excitation. (a) $D=600$; (b) $D=100$ cm^2/s; dashed line has slope given by S.

$$n(z,t)=\frac{g_s}{\tau_i}e^{D\lambda^2 t}\left[e^{-\lambda z_0}\mathrm{erfc}\left[\frac{2D\lambda t-z_0}{\sqrt{4Dt}}\right]+e^{\lambda z_0}\mathrm{erfc}\left[\frac{2D\lambda t+z_0}{\sqrt{4Dt}}\right]\right], \quad (14)$$

with $z_0=z-St$. One can easily show that the width of the cloud,

$$Z=\sqrt{\langle z^2\rangle}=\sqrt{S^2t^2+2Dt+4/\lambda^2}\ ,$$

expands faster than sound.

In order to fit our model to the experiments[9,10] in Cu_2O more quantitatively, we show in Fig. 2 the dependence of the full width at half maximum (FWHM) of the cloud density on the time delay for different excitation energies and $D=600$ [Fig. 2(a)] and 100 cm^2/s [Fig. 2(b)]. The dependence of the maximum velocity during the expansion on E/E_s is shown in Fig. 3. Comparing these results with the experimental data,[9,10] we may conclude that this model is in agreement with experiments in the case of larger diffusion only. We can also obtain

$$E_s\approx 1.5\times 10^4\ \mathrm{erg/cm^2}\ .$$

With $m=2.7m_0$ and $\hbar\omega=2.41$ meV (see Ref. 9), $\tau_r=1$ ns (we can estimate τ_r, using Einstein's formulas and $D=600$ cm^2/s), it follows that

$$\sigma_{\mathrm{ph}}\varepsilon_{\mathrm{PW}}\approx 2\times 10^{-15}\ \mathrm{meV\,cm^2}\ .$$

As we have already mentioned, this value is of the same order of magnitude as in experiments[7,8] in Ge.

As to the transport in MQW's, our model produces results for the in-plane expansion of the carrier cloud which are very similar to that discussed above. In order to compare these results with experiments,[11] we may use the δ-function approximation of Eq. (6) for W_r, because $\tau_i\approx 6$ ps $\ll\tau_{\mathrm{ph}}$ in these experiments. Figure 4 shows the dependence of FWHM on the time delay after a short excitation pulse, for $S=5\times 10^5$ $cm\,s^{-1}$, $R=1.4\times 10^{-4}$ cm, $D=20$ cm^2/s, $d=10^{-4}$ cm (see Ref. 11) and for different dimensionless excitation energies $a=E/E_{\mathrm{MQW}}$, where

$$E_{\mathrm{MQW}}=\frac{2m\hbar\omega Sd}{\tau_r\sigma_{\mathrm{ph}}\varepsilon_{\mathrm{PW}}}\ . \quad (15)$$

We can easily see that if we take for $\sigma_{\mathrm{ph}}\varepsilon_{\mathrm{PW}}=2\times 10^{-15}$ meV cm^2, i.e., the same value as in Cu_2O; $m=m_{\mathrm{h.h.}}=0.45m_0$, $\hbar\omega=1.72$ meV (see Ref. 11), and $\tau_r=6$ ps (again, we are estimating τ_r, using Einstein's formulas and $D=20$ cm^2/s), we obtain

$$E_{\mathrm{MQW}}\approx 2\times 10^3\ \mathrm{erg/cm^2}\ .$$

If $E_{\mathrm{abs}}>E_{\mathrm{MQW}}$, our model predicts a large contribution of the phonon wind to the expansion of the carrier cloud in the MQW, and the expansion becomes supersonic. This value of E_{MQW} is in agreement with the experiments.[11]

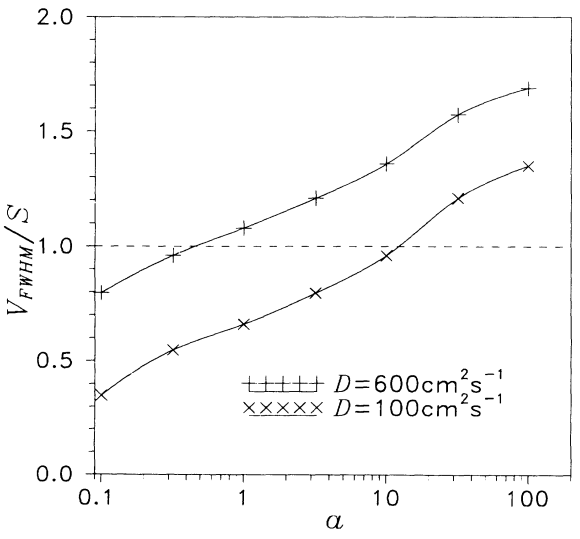

FIG. 3. The dependence of the maximum velocity of FWHM expansion after the excitation, V_{FWHM}, on dimensionless excitation energy in the case of surface excitation (see the text).

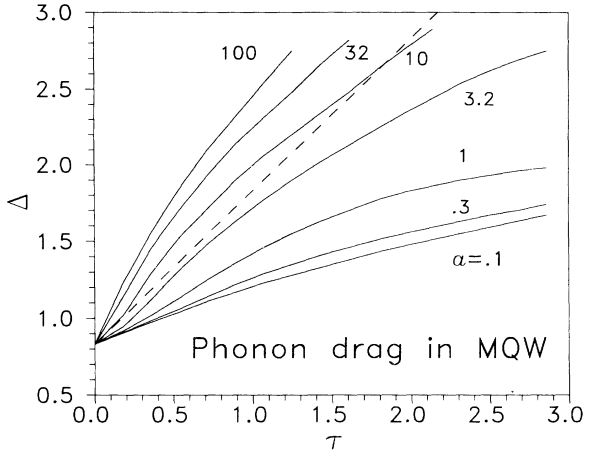

FIG. 4. The time dependence of the FWHM of the carrier concentration $\Delta(\tau)$ for different values of dimensionless excitation energy $a=E/E_{\mathrm{MQW}}$ in the case of experiments with MQW's (see the text). Unit of distance is 1.7 μm; and unit of time, 340 ps; dashed line has slope given by S.

IV. DISCUSSION

In this paper we developed a very simple model of phonon-wind-driven expansion of carriers in bulk semiconductors and in MQW's, which takes into account primary ballistic acoustic phonons, retardation in their traveling and diffusion of carriers. The principal adjustable parameter of our theory to be found from experiment (and from microscopic consideration) is $\sigma_{\mathrm{ph}}\varepsilon_{\mathrm{PW}}$, the product of the phonon-carrier scattering cross section and energy per excitation which is left after the emission of the optical-phonon cascade. The most surprising point is that, after fitting our theory to the experiments,[9] we obtained a value of $\sigma_{\mathrm{ph}}\varepsilon_{\mathrm{PW}}$ of the same order of magnitude as in experiments conducted in the past[7,8] in Ge. And using $\sigma_{\mathrm{ph}}\varepsilon_{\mathrm{PW}}$ of the same order of magnitude for experiments[11] in a GaAs-$Al_xGa_{1-x}As$ MQW we got a reason-

able estimate for excitation intensity of sonic drift. We would like to stress that all our estimates are very crude, they may guarantee only the order of magnitude of $\sigma_{ph}\varepsilon_{PW}$. So we cannot assert any actual universality of this value. But universality of its order of magnitude may make sense, because $\sigma_{ph}\varepsilon_{PW}$ is governed by mean interaction of carriers with low energy acoustic phonons, which should not vary appreciably from one semiconductor to another.

One can develop[13] a very simple model to estimate $\sigma_{ph}\varepsilon_{PW}$ at low temperatures $k_BT \ll \hbar\omega_{opt}$. We use in this model the fact that the characteristic phonon traveling time l/S is long on the scale of phonon emission times. It means just that phonons are emitted by hot carriers, whereas they are absorbed by cool carriers. Let us assume the initial energy of hot (i.e., just generated) carriers to be $\mathcal{E}_i$, and the final averaged energy and momentum of cooled carriers to be $\bar{\mathcal{E}}_f$ and $\bar{p}_f$ [e.g., $\bar{\mathcal{E}}_f \sim k_BT$, $\bar{p}_f \sim (2mk_BT/\pi)^{1/2}$ in the case of the nondegenerate final distribution of cool carriers]. Optical-phonon emission cascade is very fast, and the energy which is left after it, ε_{PW}, can be varied between 0 and $\hbar\omega_{opt}$ by tuning the excitation laser frequency: $\varepsilon_{PW} \sim \mathcal{E}_i - n\hbar\omega_{opt}$, $n = 1,2,\ldots$. Due to the energy and momentum conservation the averaged momentum of the emitted acoustic phonons is $\sim\sqrt{m\varepsilon_{PW}}$, whereas that of absorbed phonons cannot exceed $\sim\sqrt{m\bar{\mathcal{E}}_f}$. As a result the dependence of $\sigma_{ph}\varepsilon_{PW}$ on $\mathcal{E}_i$ is smoothed. The finite-energy width of excitation and carrier-carrier collisions in multicomponent system of carriers will cause the further weakening of this dependence. Finally we can obtain[13] the following estimate for our parameter:

$$\sigma_{ph}\varepsilon_{PW} = \frac{\pi E_1^2 \bar{p}_f}{\rho S^2 \hbar} , \tag{16}$$

where ρ and E_1 are the density and the deformation-potential constant of the semiconductor.

It is interesting to note that if $m \sim 10^{-27}$ g, $E_1 \sim 1$ eV, $T \sim 10$ K, $\rho \sim 5$ g cm^{-3}, and $S \sim 5\times 10^5$ cm/s, then Eq. (16) gives

$$\sigma_{ph}\varepsilon_{PW} \sim 4\times 10^{-15} \text{ meV cm}^2 .$$

Of course this value increases rapidly with the increase of E_1, but its order of magnitude coincides with that obtained from the experiments. Thus Eq. (16) justifies to some extent our conclusion that $\sigma_{ph}\varepsilon_{PW}$ should not change considerably from one semiconductor to another.

To conclude, our model may explain fast supersonic transport of excitons in experiments[9,10] with Cu_2O without assuming their superfluidity. However, it should be mentioned that, in case of superfluidity of excitons, not only their momentum relaxation time $\tau_r \to \infty$, but also exciton-phonon scattering cross section $\sigma_{ph} \to 0$ (due to spectra renormalization of superfluid excitons). It may even happen that $\tau_r\sigma_{ph}$ does not change significantly after the superfluid transition. Then our phonon-wind model does not exclude the superfluidity of excitons in experiments.[9,10] This model may be also applicable to experiment[11] on exciton transport in MQW's.

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