Phonon wind and excitonic transport in Cu_2O semiconductors

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The excitonic transport over macroscopic distance recently detected in Cu_2O semiconductors can be accounted for in terms of exciton drag by a flux of nonequilibrium phonons generated in the initial stage of formation of the exciton cloud, without invoking the hypothesis that the exiton gas undergoes a transition to a superfluid state. © *1996 American Institute of Physics.*
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1. INTRODUCTION

Over the last fifty years, a lot of attention has been given to the problem of the Bose–Einstein condensation of weakly nonideal excitonic gas. Nonetheless, no unambiguous experimental evidence in favor of this condensation in any material has been obtained. Only recently some observations have indicated the possibility of Bose–Einstein condensation in assemblies of rubidium[1] and sodium[2] atoms cooled by laser beams and separated in magnetic field.

The list of candidates for Bose–Einstein condensation is quite long (see, for example, Ref. 3). A significant place in this list is occupied by excitons, i.e., hydrogen-like bound states of electron-hole pairs in semiconductors. The arguments in favor of searching the superfluid transition in excitonic systems are obvious: first, a small mass, hence a relatively high critical temperature for the condensation; second, the possibility of tuning the exciton density over a wide range and creating excitonic matter of a considerably high density; third, the possibility of monitoring the exciton distribution function through their photoluminescence spectra.

Presently Cu_2O is considered the best material in which to search for the Bose-Einstein condensation,[4] since the interaction among excitons is repulsive in this material owing to its electronic spectrum,[1)] direct recombination of excitons is forbidden, and the lifetime of the excitons is quite long.

Starting in 1990, Wolfe, Mysyrowicz, Snoke *et al.*[4–6] have investigated the excitonic system by recording time- and coordinate-resolved photoluminescence spectra of excitons in Cu_2O and found that they can propagate across the entire crystal over a macroscopic distance of up to 1 cm, provided that the pumping pulse energy is sufficiently high. In their experiments a sample was excited by a laser pulse with a duration of about 10 ns and a quantum energy larger than the band gap, so that the light was absorbed in a narrow surface layer. Thus an initial density of excitons sufficient for the Bose–Einstein condensation could be generated. At the maximum pumping power, the energy distribution of excitons derived from luminescence spectra was in fact essentially Bose–Einstein and very close to the degenerate one with $\mu \approx 0$.[2)]

The most interesting feature of excitons in Cu_2O is the propagation of the excitonic gas from the excited surface layer into the crystal volume. It is controlled by diffusion at low pumping powers and is ballistic (with propagation velocity equal to that of sound) at higher pumping powers. At the maximum pumping power, Snoke et al.[4] detected supersonic propagation of the exciton cloud in the initial stage of its expansion (i.e., not far from the excited surface).

This propagation of excitons into the crystal faster than the diffusion was interpreted[4] in terms of the Bose–Einstein condensation and a transition of the exciton gas to a superfluid state (see also the theoretical study by Link and Baym[7]).

After further experiments, Mysyrowicz *et al.*[5,6] claimed to have more evidence in favor of Bose-Einstein condensation of excitons in Cu_2O. For example, they detected propagation of excitons over a macroscopic distance of up to 1 cm and a sharp temperature dependence of this effect:[5] when the crystal temperature was increased from 2 to 4 K, the ballistic propagation of excitons was no longer detected.

They had also proved that the process was nonlinear, which was clearly demonstrated in two-pulse experiments.[6] If the delay between the first, more intense, and second pulse was sufficiently short (between 20 and 150 ns), only one luminescence pulse was generated on the back side of the crystal instead of two, and the total luminescence intensity was larger than the sum of two signals generated by separate pulses of the same intensities. If the delay was sufficiently long (about 300 ns), the second pulse had a symmetrical soliton-like shape for a definite ratio of intensities of the first and second pumping pulses and arrived on the back side of the crystal after a longer delay than the first pulse, whose propagation was still controlled by the sound velocity.

These features of the propagation of excitons generated by two pulses were interpreted by the authors as an unambiguous proof of a superfluid transition in the exciton gas in Cu_2O. At the same time, theoretical models were proposed to support the feasibility of soliton-like bunches of excitons, provided that the superfluid transition has taken place[9] in the exciton gas.

Some questions, however, have not been answered by the concept of superfluid excitons in Cu_2O. First, why is the ballistic propagation velocity in most cases less than or equal to the sound velocity? Second, why is the nondissipative propagation of the exciton cloud possible, when in any case its shell is in the normal, not the superfluid state?

The experimental conditions[4–6] were such that the exciton gas expanded into vacuum. In this case, it is far from obvious that a cloud, even if its initial state was superfluid,

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should propagate across the crystal as a whole at a definite velocity, rather than spread over the crystal and go over to the normal state with a lower density. It would be more plausible if the liquid, whose density is fixed by attraction among particles, propagated across the crystal, rather than the gas, in which the interaction among the excitons is repulsive.

In our previous publication[8] we demonstrated that these issues can be resolved in terms of the phonon wind model in which excitons are dragged into the crystal by a flow of nonequilibrium ballistic phonons generated due to the relaxation of nonequilibrium carriers. At a low pumping power, the phonon flow density is low, and propagation of excitons is controlled by diffusion. But if the pumping power is sufficiently high, the phonon flow density, which is proportional to the pumping power, is high enough to drive all excitons from the surface layer into the crystal volume. In this case, the propagation of excitons is purely ballistic, and their velocity is, naturally, equal to the longitudinal sound velocity.[3)] Numerical calculations based on the phonon wind model are in good agreement with experimental data[4] in the initial stage of the excitonic expansion, i.e., at a small distance from the excited surface.

In this paper we demonstrate the possibility of interpreting the propagation of excitons over a macroscopic distance in terms of the phonon wind. In Sec. 2 the phonon wind model is formulated in mathematical terms, and calculations for the excitonic propagation over a macroscopic distance are given in Sec. 3. In Sec. 4 we compare calculations to experimental data[5,6] and discuss the adequacy of our model for the measured parameters of the excitonic propagation in Cu_2O.

2. PHONON WIND

Earlier[8] we proposed a simplified model of exciton drag by a flux of nonequilibrium acoustic phonons, which are inevitably generated in the final stage of thermalization of nonequilibrium carriers created by a laser pulse. This model was applied after minor modifications to the electron-hole liquid in semiconductors[10] (see also the review by Tikhodeev[11]).

In the Cu_2O experiments[4–6] the laser quantum energy was higher than the band gap, so the generated carriers were overheated. The thermalization starts with the cascade emission of optical phonons (the typical time is 10^{-12} s) and ends with the emission of long-wave acoustic phonons after a typical time of 10^{-10}–10^{-9} s. At low temperatures acoustic phonons propagate ballistically.[12] Our model[8] takes into account only the effect of these ballistic phonons on the excitonic system. The point is that the emission time of these phonons is shorter than the pulse width ($\tau_i \approx 10^{-8}$ s) and the typical detection time (several microseconds), so their delay with respect to the laser pulse is negligible, and the profile of the generated phonon flux versus time is almost identical to that of the laser pulse, whereas secondary ballistic phonons due to decay of optical phonons are generated after a considerable delay of about 10^{-6} s and have little effect on the process.

Assuming that, first, the generation rate of ballistic phonons follows closely the laser pulse intensity, second, only a small fraction ε_{PW} of the energy of each photon energy is converted to ballistic phonons, and third, the generated phonons are incoherent, we can write the following equation for the flow of ballistic acoustic phonons:

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

where V is the excited volume, g is the carrier generation rate per unit volume,

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

$g_s=\lambda I/\hbar\omega$, I and $\hbar\omega$ are the laser pulse intensity per square centimeter and the quantum energy, λ is the light absorption index, and S is the velocity of longitudinal sound in Cu_2O ($4.5\cdot 10^5$ cm/s).

With due account of the diffusion and phonon wind, the expansion of the carrier cloud can be described by the following transport equation for the gas density $n(\mathbf{r},t)$:

$$\frac{\partial n}{\partial t}+\operatorname{div} n\mathbf{V}=D\Delta n-\frac{n}{\tau_c}+g. \qquad (3)$$

Here $\mathbf{V}(\mathbf{r},t)$ is the carrier drift velocity due to the phonon wind. We use a simple relation between the phonon flux density and this velocity:

$$\mathbf{V}(\mathbf{r},t)=S\frac{\alpha\mathbf{W}}{1+\alpha W}, \qquad (4)$$

where $\alpha=\tau_r\sigma_{ph}/Sm$ is the coupling constant between the phonon wind and driver carriers at $V\ll S$, m and τ_r are the mass and momentum relaxation time of an electron–hole pair, and σ_{ph} is the cross section of the exciton–phonon interaction. Equation (4) takes into account that the phonon wind cannot accelerate carriers to velocities beyond that of sound.[4)]

Numerical solutions of Eq. (3) with Cu_2O parameters are in good agreement[8] with experimental data[4] in the initial stage of propagation of the carrier cloud within a time interval of 10^{-7} s and a range of 10^{-2} cm. Note that this simple model includes only two fitting parameters, namely the diffusion coefficient D and the characteristic laser pulse intensity[5)]

$$I_{PW}=\frac{mS\hbar\omega}{\sigma_{ph}\varepsilon_{PW}\lambda\tau_r\tau_i}. \qquad (5)$$

Thus all the parameters of the model (except the diffusion coefficient), whose uncertainties usually are relatively large, are included in Eq. (5) as multipliers and dividers, and the optical energy density is measured in dimensionless units: $A=I/I_{PW}$. It follows from numerical calculations (see below) that changes in the diffusion factor (between 200 and 600 cm^2/s) have little effect on the excitonic transport over a macroscopic distance. Therefore we can easily derive the parameter I_{PW} by comparing our calculations to experimental data and try to determine its temperature dependence.

Among the parameters in Eq. (5), the cross section σ_{ph} of the exciton–phonon interaction and the fraction of the photon ε_{PW} energy converted to ballistic phonons are the hardest to derive from first principles. Both these parameters are interdependent functions not only of the exciton–phonon

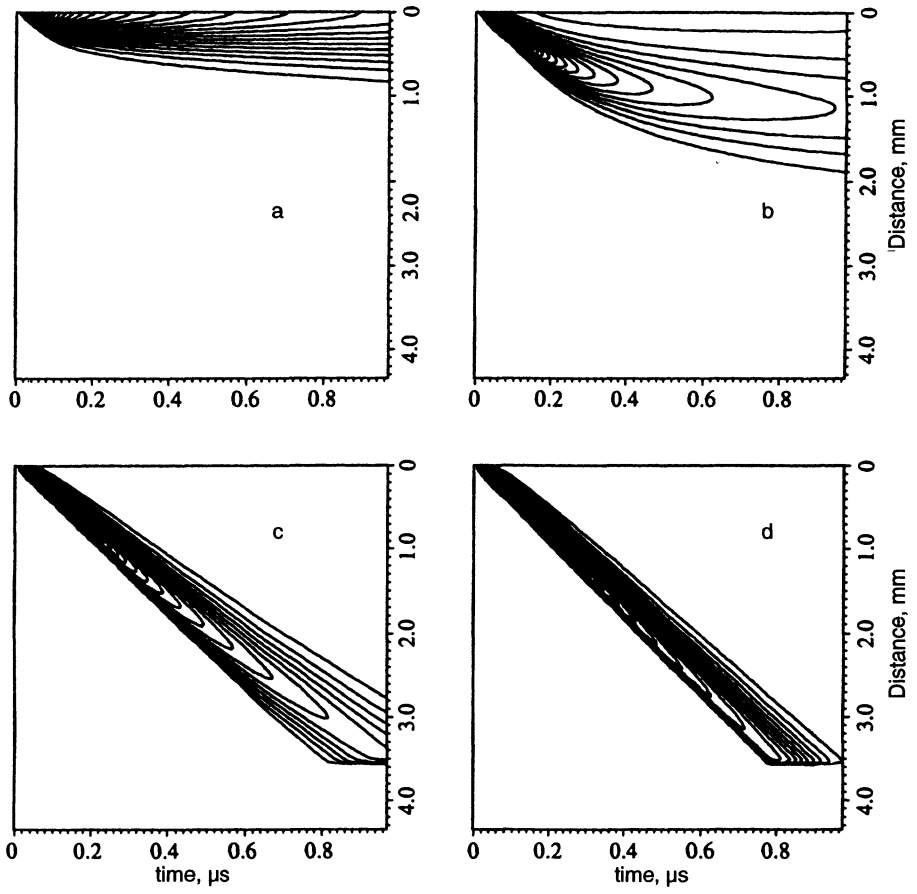

FIG. 1. Lines of equal exciton density $n(z,t)$ calculated by the phonon-wind model at four pumping power densities. The back surface of the sample at 3.56 mm from the front surface is modelled by a sharp drop in the exciton lifetime for $z \geqslant 3.56$ mm. The difference between the densities on neighboring lines is 0.01. (a) $I/I_{PW}=10^2$; (b) 10^3; (c) 10^4; (d) 10^5.

interaction in the semiconductor, but also of the parameters of the interaction among phonons. Simple estimates given in Ref. 8 indicate that, from the viewpoint of the microscopic theory, it is reasonable to consider the product of these parameters, $P=\sigma_{\rm ph}\varepsilon_{PW}$, as an average characteristic of the phonon-wind intensity. It is remarkable that the value of P obtained by comparing phonon-wind calculations to experimental data on Cu_2O[4] is of the same order of magnitude as for the electron-hole liquid in Ge and Si.[11]

3. NUMERICAL CALCULATIONS OF MACROSCOPIC TRANSPORT

The phonon wind model allows us not only to simulate the exciton transport over a macroscopic distance without any assumption about superfluidity, but also to model the most interesting effects observed in experiments, such as nonlinearity and formation of soliton-like pulses. Let us start with the transport over a large distance. Figure 1 shows typical numerical solutions of Eq. (3) which describe the exciton density as a function of time and position, $n(z,t)$, where z is the distance from the excited surface and t is the time with respect to the laser pulse. The model parameters used in this calculations are the same as in our previous publication:[8] $\tau_i=6$ ns, $D=600$ cm^2/s, $m=2.7m_0$, $\hbar\omega=2.41$ eV, $\lambda=1.6\cdot10^4$ cm^{-1}, $S=4.5\cdot10^5$ cm/s. The back surface of the sample, whose coordinate was $z=3.56$ mm in this case, was simulated by a sharp drop in the exciton lifetime from 10^{-5} s to 10^{-12} s.

Figure 1 clearly shows how the numerical solution changes with the phonon wind intensity. At a low pumping power it is totally controlled by the diffusion, and at higher powers it is purely ballistic. Note that at a given intensity of the driving pulse, irrespective of its amplitude, the phonon wave, in the long run, leaves behind the cloud of excitons at some distance $L_*(I)$ from the excited surface. Then the exciton cloud lagging behind the acoustic wave spreads in the sample due to diffusion. The higher the laser pulse intensity I, the larger the distance at which the excitons fall behind the phonons.

Therefore excitons cannot be detected at a large distance z_0 from the excited surface if the pumping power is not sufficiently large, i.e., if the distance is larger than the diffusion length corresponding to the detection time. But the signal emerges suddenly when the pumping power exceeds a certain threshold. It is important in what follows that at a pumping power much higher than the threshold, the bunch of excitons travels to the detection site with the sound velocity. But at an intermediate pumping power, when the distance to the excited surface is comparable to that at which excitons fall behind the phonon wave, $z_0 \approx L_*(I)$, the average velocity of the excitons is less than that of sound.

This is illustrated by Fig. 2, which shows calculations of the exciton flux at a distance of 3.56 mm from the excited surface versus time at several pumping powers. The curves conform to the experimental data given in Fig. 2 of Ref. 5. Figure 3 demonstrate good agreement between our calcula-

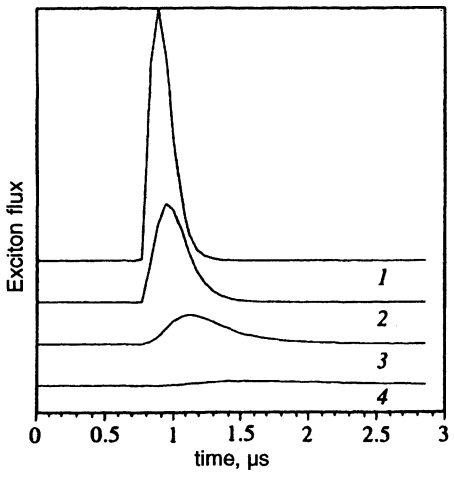

FIG. 2. Flow of excitons on the back surface of the 3.56-mm sample calculated by the phonon-wind model at four pumping power densities: *1*) $I/I_{PW}=10^{4.25}$; *2*) 10^4; *3*) $10^{3.75}$; *4*) $10^{3.5}$; $D=600$ cm^2/s.

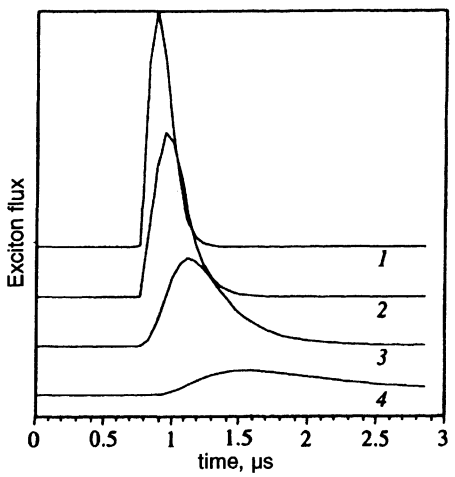

FIG. 4. Flow of excitons on the back surface of the 3.56-mm sample calculated by the phonon-wind model at $I=3.6\cdot10^5$ W/cm^2 and (*1*) $I_{PW}=20$ W/cm^2; (*2*) 36; (*3*) 63; (*4*) 112 W/cm^2; $D=600$ cm^2/s.

tions and experimental data[5] measured in two samples with thicknesses of 3.56 mm (dots) and 8.3 mm (diamonds) at $T=1.85$ K. The arrival time of the exciton-density maximum versus the pumping power density has been calculated at $D=200$ and 600 cm^2/s. By comparing our calculations to the measurements, we have obtained $I_{PW}\approx20$ W/cm^2 at $T=1.85$ K.

Let us recall that in our model the critical pumping power of the ballistic propagation of excitons should increase with the distance to the excited surface. This conclusion is illustrated by the curves in Fig. 3.

As was noted above, it follows from the definition of the dimensionless optical power density that the critical power

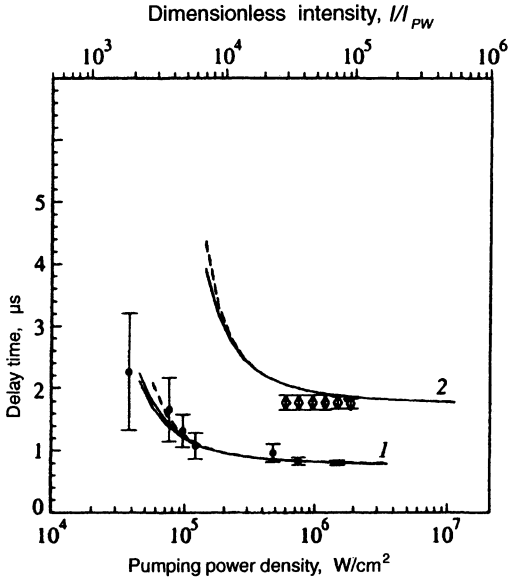

FIG. 3. Delay of the excitonic pulse as a function of the pumping power density for samples with thicknesses of (*1*) 3.56 and (*2*) 8.3 mm. Experimental data from Ref. 5 are shown by dots and diamonds, calculations by solid lines ($D=600$ cm^2/s) and dashed lines ($D=200$ cm^2/s); $I_{PW}=20$ W/cm^2.

of the ballistic propagation should change with system parameters (such as temperature). This statement is illustrated in Fig. 4 by a set of curves calculated at a fixed distance of 3.56 mm and pumping power density of $I=3.6\cdot10^5$ W/cm^2, but at different values of I_{PW}. The exciton propagation mode changes from diffusion to ballistic when the parameter I_{PW} decreases, as in the case of increasing power density (cf. Fig. 2). Hence our model can also describe the temperature dependence of the exciton transport over a microscopic distance if I_{PW} is a function of temperature. By comparing our results with the experimental data from Ref. 5, we have concluded that in the temperature range of 2 to 4 K, I_{PW} should be proportional to T^4. A tentative interpretation of this temperature dependence will be given in the next section.

Now let us consider calculations for the two-pulse excitation and compare our results to the experimental data from Ref. 6. These experiments demonstrated that, in the two-pulse experiments, the exciton propagation was nonlinear in the pumping power, which was interpreted by the authors as crucial evidence in favor of the Bose condensation and superfluidity of excitons. But we can demonstrate that this effect can be ascribed to the phonon wind and interpreted without the hypothesis about a superfluid exciton flux.

Calculations for a sample with a thickness of 2.65 mm and a sequence of two laser pulses are summarized in Fig. 5 (curves a and b). The curve a shows the signal due to the single first pulse, and the curve b is the signal due to the second pulse. The curve c shows the signal due to the two pulses with a delay of 20 ns between them. The curve d, like the similar curve in Fig. 1 of Ref. 6, is the algebraic difference between the curves c and a, and, according to Mysyrovicz *et al.*,[6] it illustrates attraction between two sequential excitonic pulses with a small delay between them. But in the phonon wind model, this is caused by the overlap between two phonon pulses. Since phonons generated on the periphery of the laser spot arrive after a considerable delay, the phonon pulse generated by laser light is fairly long, and if

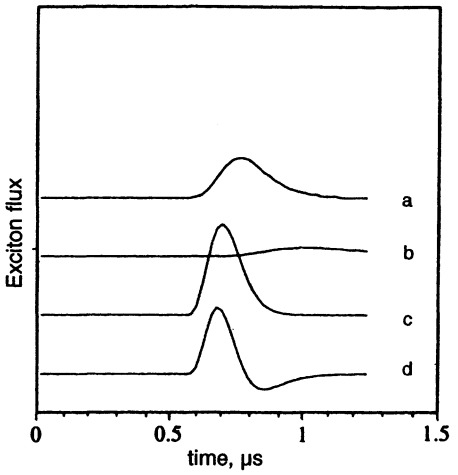

FIG. 5. Flux of excitons on the back surface of the 2.65-mm sample calculated by the phonon-wind model: (a) excitation by a single pulse; (b) excitation by a weaker single pulse; (c) excitation by two pulses with a delay of 20 ns between them; (d) algebraic difference between the signals shown by curves c and a. $I_a = 10^{3.75} I_{PW}$; $I_b/I_a = 0.562$; $D = 200$ cm^2/s.

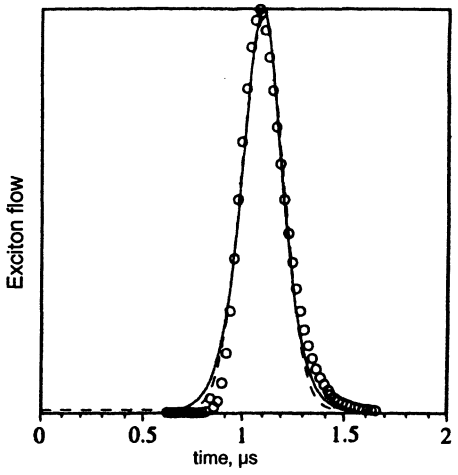

FIG. 7. Second exciton pulse on the back surface of the 2.5-mm sample due to two laser pulses with a delay of 278 ns minus the signal due to the isolated first pulse calculated by the phonon-wind model (circles). The solid line shows the soliton-like pulse shape and the dashed line the Gaussian shape. $I_a = 10^4 I_{PW}$; $I_b/I_a = 0.1$; $D = 200$ cm^2/s.

the delay between pulses generating excitons is sufficiently short, the excitons generated by the second pulse can be driven by the phonons due to the first pulse. Since its intensity is higher than that of the second pulse, these excitons can be driven farther into the sample than in the case of an isolated weaker optical pulse generating nonequilibrium phonons.

Figure 6 shows calculations for a larger delay between laser pulses and simulates another effect illustrated by Fig. 2 in Ref. 6. Namely, the signal due to two sequential pulses is considerably larger than the algebraic sum of two signals due to isolated pulses. Mysyrowicz *et al.*[6] interpreted this nonlinearity in terms of interaction between superfluid excitonic systems. But in our phonon wind model, this nonlinearity means that the excitons generated by the first pulse and not entrained by the first phonon wave are driven by the second phonon pulse.

When the delay between two pulses is larger, the following effect was observed:[6] at a certain ratio of the pulse intensities (about 0.1) and a delay of about 300 ns, the signal due to the second pulse, which could not be detected if its laser pulse was isolated, was detected after a considerable delay, i.e., propagated at a velocity much lower than that of sound, and had a symmetrical soliton-like shape described by the function $\mathrm{sech}^2\xi$, where $\xi = (t - t_0)/\tau_s$. We could simulate delayed excitonic pulses within the framework of our phonon wind model (Fig. 7). But we were unable to adjust the model parameters so as to reproduce the symmetrical soliton-like shape of pulses.

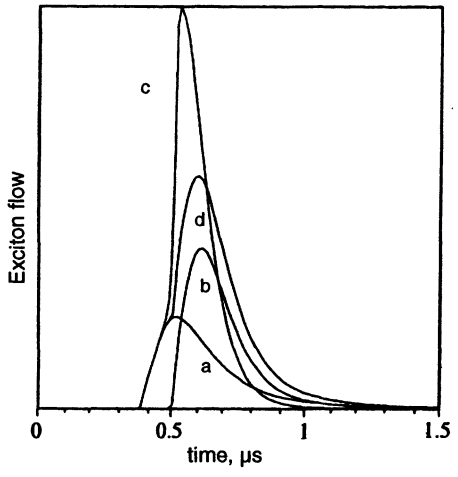

FIG. 6. Flux of excitons on the back surface of the 2.5-mm sample calculated by the phonon-wind model: (a) excitation by a single pulse; (b) excitation by a stronger pulse; (c) by two pulses with a delay of 125 ns between them; (d) algebraic sum of the signals shown by curves a and b; $I_a = 10^{3.5} I_{PW}$; $I_b/I_a = 1.3$; $D = 600$ cm^2/s.

4. DISCUSSION

We have demonstrated that, although our phonon wind model is very approximate, it has allowed us to interpret most experimental results concerning exciton transport in Cu_2O[4–6] with no hypothesis about a superfluid transition in the excitonic system.

In conclusion, we would like to add the following statements:

1. Our model does not rule out that the excitonic gas driven by nonequilibrium phonons may be superfluid, provided that the Bogolyubov sound velocity is smaller than the sound velocity S in the semiconductor.

2. The temperature dependence of I_{PW} may be controlled by the temperature dependence of the momentum relaxation time $\tau_r(T)$ in the excitonic system and by the decrease in the fraction of ballistic phonons with the temperature, which seems well justified.

3. The only effect that we could not simulate was the symmetrical soliton-like shape of the second pulse excited after a delay of about 300 ns. However, given that our model

can be easily modified in order to obtain a symmetrical shape of the second excitonic pulse at certain delays and ratios of pulse amplitudes, we do not consider this fact as a serious argument against the phonon wind model. For example, we may take into account the phonons due to the first pulse reflected from the back surface of the sample. The oppositely directed phonon wave may delay the second exciton pulse and make its shape more symmetrical. Another possibility is to include, in addition to the ballistic phonons, the phonons generated from the hot-spot evolution. Recent experimental results[6)] indicate that such phonons may be important for the excitonic transport in Cu_2O. Unfortunately, after such modifications the number of adjustable parameters in the model will increase, making it less convincing.

To sum up, our phonon wind model provides a satisfactory interpretation of the excitonic transport over macroscopic distances in Cu_2O. By introducing additional fitting parameters, we can improve the agreement between calculations and experimental data. But this is hardly necessary because one should not expect to obtain too much from the simplified model, which demonstrates qualitative agreement with the experimental data.

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[1)]Usually excitons are attracted to each other due to the exchange interaction, so the Bose–Einstein condensation is possible only in a system of excitonic molecules, i.e., biexcitons.

[2)]Here we do not discuss the energy spectrum of excitons in Cu_2O in detail since it is fairly complex: the excitonic level is split into the states of the nondegenerate ortho-exciton and the three-fold degenerate para-exciton. The difference between the two components of the excitonic system is not essential for our purpose.

[3)]The propagation velocity near the excited surface may be higher than that of sound owing to diffusion.[8] Besides, plasma expansion, which is not included in our calculations, is also possible.

[4)]In reality, one can conclude from a simple kinematic diagram that even a pair moving at a supersonic speed can absorb a phonon and gain an additional acceleration if the velocity projection on the phonon velocity is smaller than the speed of sound. But this process considerably complicates our model and has little effect on the final results.

[5)]In Ref. 8 we used the dimensionless energy density instead of intensity, $E_s \propto I_{PW}\lambda\tau_i^2 S$. In this calculation we use the intensity to make easier the comparison to experimental data.[4–6]

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Note added in proof (10 April 1996). In the preliminary version of this paper [G. A. Kopelevich and S. G. Tikhodeev, in: *Proceedings of the 22nd Conf. on the Physics of Semiconductors*, ed. by D. J. Lockwood, World Scientific, Singapore (1995), p. 61.], a computational mistake was made, which resulted in too short ballistic pulses.

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