Propagation of exciton pulses in semiconductors

A. D. Jackson\(^1\) and G. M. Kavoulakis\(^2,\ast\)

\(^1\)Niels Bohr Institute, Blegdamsvej 17, DK-2100 Copenhagen \(\ddot{\text{O}}\), Denmark,
\(^2\)Royal Institute of Technology, Lindstedtsvägen 24, S-10044 Stockholm, Sweden

(October 27, 2018)

Using a toy model, we examine the propagation of excitons in Cu\(_2\)O, which form localized pulses under certain experimental conditions. The formation of these waves is attributed to the effect of dispersion, non-linearity and the coupling of the excitons to phonons, which acts as a dissipative mechanism.

PACS numbers: 05.45.Y, 05.30.Jp, 71.35.-y

I. INTRODUCTION

The transport properties of excitons [1] in Cu\(_2\)O have been studied extensively in recent years. In one series of experiments [2–6], a pulse of excitons was created at one end of a crystal of Cu\(_2\)O and detected at the other following propagation inside the sample. In another series of experiments [7–9], excitons were created and imaged as they expanded inside a crystal subjected to external stress. (The stress effectively created a confining potential for the excitons.)

Two noteworthy conclusions emerged from these experiments. First, drag in the propagation of excitons due to their scattering with (longitudinal) acoustic phonons was found to decrease with decreasing lattice temperature. The second observation [3–5] followed from experiments with two laser pulses separated by a time delay of \(\sim 100\) ns. The first pulse created an asymmetric wave of excitons; the excitons created by the second pulse travelled in the trail of the first. This second wave was observed to be symmetric and to have the characteristics of a solitary-wave, specifically, those of a “bright soliton” (i.e., a local elevation in the density.)

Here, we will argue that the waves created by the second pulse are likely to resemble classical solitary waves. These waves are, however, essentially different from solitary waves in, e.g., water (as first observed by J. Scott Russell [10]) due to the presence of dissipation caused by the scattering of excitons with phonons [7–9]. We will suggest that the effects of nonlinearity and dispersion, which give rise to solitons, must be combined with dissipation in order to create generic conditions appropriate for the propagation of localized pulses of excitons in the crystal. All three mechanisms play an important role.

The studies of Refs. [11,12] attributed the observed localized waves to the formation of a Bose-Einstein condensate of excitons [13]. In Ref. [11], second-order perturbation theory was used to describe the exciton-exciton interaction [11], which gave rise to effective attraction between the condensed excitons. In Ref. [12], it was shown that an exciton condensate coupled to acoustic phonons can give rise to bright solitons. However, as pointed out in Ref. [14], solitary waves in a Bose-Einstein condensed gas have a characteristic length scale determined by its healing length \(\xi\) given by \(\hbar^2/2m\xi^2 = nU_0\), or \(\xi = (8\pi n a)^{-1/2}\), where \(m\) is the exciton mass, \(n\) is the exciton condensate density, and \(U_0 = 4\pi\hbar^2 a/m\) is the exciton-exciton scattering matrix element, with \(a\) being the scattering length. For typical liquid helium temperatures of order 2 K, the critical density for the Bose-Einstein condensation of excitons is of order \(10^{17}\) cm\(^{-3}\).

The scattering length for exciton-exciton elastic collisions in Cu\(_2\)O has been calculated to be \(a \approx 2a_B\) [15], where \(a_B\) is the exciton Bohr radius. The Bohr radius of excitons in Cu\(_2\)O is \(\approx 5\) Å [16] so that \(\xi \sim 0.01\) \(\mu\)m, which is about four orders of magnitude smaller than the width of the narrowest pulses \(l_p \approx 1\) mm. (The crystal is typically a few millimeters long.)

Finally, a model which has been proposed in connection with the ballistic propagation of excitons attributes the observed phenomenon to a classical effect due to a “phonon wind” [17]. Many of the observations have been explained in the context of this model and our study is consistent with the claim that the excitons are described by classical equations. On the other hand, the two studies examine effects which are essentially decoupled.

More specifically, while both models include dissipation, the phonon-wind model focuses on the spatial and temporal effects of laser excitation, whereas our model concentrates on the effects of non-linearity and dispersion. In the present study we make the implicit simplified assumption of a delta-like form of laser excitation in space and time. In such a case, the phonon-wind model essentially reduces to a diffusion-like equation, neglecting the effects of non-linearity and dispersion. Of course, one should worry about more complicated forms of the laser excitation, however the problem we have studied is quite complicated and we have chosen to overlook these effects in order to demonstrate clearly the combined effect of dissipation, non-linearity and dispersion; we believe all three effects are very crucial in these experiments. Finally, from an experimental point of view, our study would be mostly suitable for short (in space and time) laser pulses.
II. HYDRODYNAMIC DESCRIPTION

Following Ref. [18], we start with the hydrodynamic equations describing the system,

$$\frac{\partial n}{\partial t} + \nabla \cdot (n \mathbf{v}) = 0,$$

$$\left[ \frac{\partial}{\partial t} + \mathbf{v} \cdot \nabla \right] \mathbf{v} = -\frac{\nabla p}{n} - \frac{\mathbf{v}}{\tau},$$

where \( \mathbf{v} \) is the velocity field, \( p \) is the pressure, and \( \tau \) is the exciton-acoustic phonon scattering time. The neglect of viscosity in these equations can be justified by looking at the characteristic scales: The viscous force \( \eta \nabla^2 v \) is of order \( \eta v/l_p^2 \sim n\tau_s c^3/l_p^2 \), where \( \eta \) is the viscosity, \( \tau_s \) is the scattering time for elastic exciton-exciton collisions, and \( c^2 = \partial p/\partial n \) is the adiabatic sound velocity. Therefore, \( |\eta \nabla^2 v/\nabla p| \sim \tau_s/c \). Typical values of \( c \sim 5 \times 10^5 \text{ cm/s} \), \( l_p \sim 1 \text{ mm} \), and \( \tau_s = (n\sigma v)^{-1} \) with \( \sigma = 8\pi a^2 \) imply that \( \tau_s \sim 1 \text{ ns} \) for \( n \sim 10^{17} \text{ cm}^{-3} \), \( a \sim 10 \text{ Å} \), and a temperature \( T \sim 1 \text{ K} \). Therefore, \( \tau_s/l_p \sim 10^{-2} \ll 1 \).

Linearizing the above equations and considering small variations in the density and the pressure, \( n = n_0 + \delta n \) and \( p = p_0 + \delta p \) (where \( n_0 \) and \( p_0 \) are the “background” density and pressure of excitons created by the first laser pulse), one can derive the equation:

$$\frac{\partial^2 \delta n}{\partial t^2} - c^2 \nabla^2 \delta n + \frac{1}{\tau} \frac{\partial \delta n}{\partial t} = 0.$$ \hspace{1cm} (2)

This equation leads to the dispersion relation \[18\]

$$\omega^2 - c^2 k^2 + i\omega/\tau = 0,$$

which interpolates between two limits. For \( kc\tau \ll 1 \), the motion of the exciton pulse is diffusive, and

$$\partial_t \delta n = c^2 \tau \nabla^2 \delta n,$$ \hspace{1cm} (4)

with \( \omega = -ic^2 \tau k^2 \). In the opposite limit \( kc\tau \gg 1 \),

$$\partial_t \delta n - c^2 \nabla^2 \delta n = 0,$$ \hspace{1cm} (5)

with \( \omega = ck \), corresponding to sound propagation.

III. NON-LINEARITY VERSUS DISPERSION

The analysis so far applies to small variations around equilibrium. For larger excitations it is also necessary to account for the effects of non-linearity and dispersion. For simplicity, we start with a (one-dimensional) wave propagating with velocity \( c \) along the positive \( x \) axis,

$$\partial_t \delta n + c \partial_x \delta n = 0.$$ \hspace{1cm} (6)

Any function of the form \( \delta n = \delta n(x - ct) \) is a solution of Eq. (6). Allowing for dispersion and non-linearity, this becomes

$$\frac{\partial \delta n}{\partial t} + c(1 + \alpha \frac{\delta n}{n_0}) \frac{\partial \delta n}{\partial x} + \beta c^3 \frac{\partial^3 \delta n}{\partial x^3} = 0.$$ \hspace{1cm} (7)

Here \( \alpha \) and \( \beta \) are small (positive) dimensionless quantities, which can be calculated from microscopic theories. It is not our goal to calculate them here, and we treat them as phenomenological parameters. Also \( \tau_p \) is the time for the pulse to travel a length \( l_p \) (i.e., \( l_p = c\tau_p \)).

Typically, \( \tau_p \sim 100 \text{ ns} \) for relatively narrow pulses. Equation (7) leads to the dispersion relation

$$\omega = ck \left[ 1 + \alpha \frac{\delta n}{n_0} - \beta (k l_p)^2 \right],$$ \hspace{1cm} (8)

which provides a reasonable description of non-linearity and dispersion. For \( k \to 0 \), the sound velocity is \( c(1 + \alpha \delta n/n_0) \), and the local sound velocity is greater in regions of higher density. For \( \alpha = 0 \), \( \omega/k = c[1 - \beta (k l_p)^2] \), which implies that longer wavelengths travel faster. Equation (7) is of standard Korteweg–de Vries form, and the competition between the last two terms gives rise to soliton solutions [19].

We seek solutions of Eq. (7) which preserve their shape, i.e., which have the form \( \delta n(x - ut) \), and which satisfy the boundary condition that \( \delta n \to 0 \) as \( |z| \to \infty \). Such solutions have the form

$$\delta n = C \text{sech}^2 \left( \frac{(\alpha C)^{1/2}}{12\beta} \frac{x - ut}{l_p} \right),$$ \hspace{1cm} (9)

with \( u = c(1 + \alpha C/3) \). For fixed \( \alpha \) and \( \beta \), there is thus a family of solutions characterized by their maximum amplitude, \( C \). For \( \alpha C \sim 12\beta \) the size of the travelling wave is of order \( l_p \). This result reflects the expected competition between non-linearity and dispersion that gives rise to this localized excitation. In addition, the velocity of this pulse is of order \( c \), which can approach the speed of sound \( c_l \) of \( \text{Cu}_2\text{O} \). For example, for a classical gas \( p = nk_BT \), and thus \( c^2 = k_BT/m \), with \( m = 3m_e \) being the total exciton mass density (\( m_e \) is the electron mass.) For \( T \approx 2 \text{ K} \), \( c \approx 3.2 \times 10^{5} \text{ cm/s} \), in good agreement with experiment.

IV. SCATTERING OF EXCITONS WITH THE CRYSTAL

Equation (7) does not include the effects of the scattering of excitons with the crystal. As noted above, this mechanism leads to the diffusive motion of excitons. It is thus worth considering what is known about exciton-phonon interactions in \( \text{Cu}_2\text{O} \).

In Refs. [7,8], paraexcitons under low excitation conditions were observed to be highly mobile, with a diffusion constant \( D \sim 10^3 \text{ cm}^2/\text{s} \). For \( D = c^2\tau \), with \( c^2 = k_BT/m \), \( \tau \) is seen to be \( \approx 20 \text{ ns} \). For typical thermal velocities of order \( 3 \times 10^5 \text{ cm/s} \), the mean-free path of excitons is \( \approx 50 \mu \text{m} \). This corresponds to \( \approx 10^5 \) lattice sites and...
is thus quite large. In addition, $D$ showed a rapid increase for temperatures below $\approx 6$ K. These observations were explained in Ref. [8] using a simple classical model which showed that excitons with wavenumbers smaller than $k_{\text{f}} = mc_{\text{f}}/\hbar$, where $c_{\text{f}}$ is the longitudinal speed of sound in Cu$_2$O ($c_{\text{f}} \approx 4.5 \times 10^5$ cm/s), cannot emit acoustic phonons and thus cannot scatter with the lattice. For exciton temperatures below $T_{\text{f}}$, where $k_{\text{B}}T_{\text{f}} \approx mc_{\text{f}}^2$, exciton-acoustic phonon scattering is suppressed. This freeze-out temperature is $T_{\text{f}} \approx 4$ K for longitudinal acoustic phonons in Cu$_2$O.

In the experiments of Refs. [2–5] with an excitation of $\approx 1.2 \times 10^6$ W/cm$^2$, the temperature varied between 1.9 K and 4.1 K. For $T \approx 2.5$ K, which is close to the freeze-out temperature $T_{\text{f}}$, the propagation of the exciton pulse showed a transition from diffusive to ballistic motion. In Ref. [4] for conditions where the exciton propagation was diffusive, the diffusion constant was reported to be even higher, $D \approx 2 \times 10^3$ cm$^2$/s, corresponding to $\tau \approx 40$ ns.

It is worth pointing out that in the experiments where the sample was illuminated with two pulses [3–5], these were delayed by $\approx 100$ ns. This time delay is comparable to $\tau_{\text{p}}$. It is thus reasonable to expect that the first pulse diffuses inside the crystal and creates a background density of excitons through which the second pulse must pass.

V. A MORE REALISTIC PROBLEM

In view of these considerations, we have included an additional term in Eq. (7) to describe the effects of exciton-phonon scattering:

$$\frac{\partial \delta n}{\partial t} + c(1 + \frac{\delta n}{n_0}) \frac{\partial \delta n}{\partial x} + \frac{\beta \delta n}{\tau_{\text{p}}} \frac{\partial^2 \delta n}{\partial x^2} = \frac{\gamma l_p^2 \partial^2 \delta n}{\tau_{\text{p}}} \frac{\partial^2 \delta n}{\partial x^2}, \quad (10)$$

with $\gamma$ being a positive parameter. Here $\tau_{\text{p}}/\gamma$ can be identified as the exciton-acoustic phonon scattering time. Since $\tau_{\text{p}} \sim 100$ ns and $\tau \sim 30$ ns, $\gamma$ is expected to be greater than, but on the order of unity. The term on the right side of Eq. (10) gives an imaginary part to the dispersion relation, modifying Eq. (8) to

$$\omega = ck \left[ 1 + \frac{\delta n}{n_0} - \beta (kl_{\text{p}})^2 \right] + (kl_{\text{p}})^2 \frac{2\gamma}{\tau_{\text{p}}}. \quad (11)$$

With such a dispersion relation the wave is damped. For example for $k \sim 1/l_{\text{p}}$, $\delta n \sim e^{-\gamma t/\tau_{\text{p}}}$. 

VI. EFFECT OF DISSIPATION ON THE PULSE PROPAGATION

Equation (10) interpolates between two extreme modes of propagation. For $\gamma = 0$ the solution is a travelling wave, which propagates without change in shape with a velocity of order $c$. When $\gamma$ is nonzero, the width of the pulse increases with time and its shape changes. As $\gamma$ is increased, this spreading velocity increases and eventually equals $c$ at some $\gamma_{\text{max}}$. For $\gamma \ll \gamma_{\text{max}}$, the pulse still propagates slowly relative to its propagation velocity of $\approx c$.

To determine $\gamma_{\text{max}}$, it is convenient to use the notation $\phi = \delta n/n_0$ and the quantities

$$\langle X \rangle = \int \frac{x \phi \, dx}{\int \phi \, dx} \quad \text{and} \quad \langle X^2 \rangle = \int \frac{x^2 \phi \, dx}{\int \phi \, dx}, \quad (12)$$

and the associated width $\Delta X = \sqrt{\langle X^2 \rangle - \langle X \rangle^2}$. The time derivative $\partial_t \langle X \rangle$ can be regarded as the velocity of propagation of the pulse; $\partial_t \Delta X$ describes the rate of increase of the width. The desired average values follow immediately from appropriate integrals of Eq. (10). We find that

$$\frac{\partial}{\partial t} \langle X \rangle = c \left( 1 + \frac{\alpha}{2} \int \frac{\phi^2 \, dx}{\phi} \right). \quad (13)$$

For $\gamma = 0$ and $\phi$ given by Eq. (9), Eq. (13) gives the expected result that $\partial_t \langle X \rangle = c(1 + \alpha C/3)$, i.e., the velocity of propagation $u$ given above. Similarly, we find that

$$\frac{\partial}{\partial t} \Delta X^2 = \alpha C \left( \int \frac{x^2 \phi^2 \, dx}{\phi} - \int \frac{x \phi \, dx}{\phi} \int \frac{\phi^2 \, dx}{\phi} \right) + 2\gamma c l_{\text{p}}. \quad (14)$$

The $\alpha$-dependent term in this equation evidently vanishes for any symmetric pulse shape. For such pulses we see that

$$\frac{\partial_t \Delta X}{\Delta X} = \frac{c l_{\text{p}}}{\Delta X}. \quad (15)$$

For pulses of typical width of order $l_{\text{p}}$, this suggests that $\gamma_{\text{max}}$ is of order 1. The characteristic scattering time of $\approx \tau_{\text{p}}$, which establishes the crossover from the motion of a localized pulse to diffusive motion, is thus of the same order as the characteristic scattering time $\tau$ for exciton-phonon collisions that has been determined experimentally as described above. As we discuss below, the approximately linear growth of $\Delta X^2(t)$ indicated by Eq. (14) provides a definite test for the validity of our description.

VII. THE BENEFITS OF DISSIPATION

While dissipation can obscure the solitonic propagation, it also plays a far more positive role. In its absence, an initially localized pulse will often break up into a discrete number of solitons (plus small amplitude, non-solitonic motion.) This is illustrated in Fig. 1(a) where we show the evolution of an initial pulse.
with $\lambda = 3$, which leads to the formation of three solitons. (The calculation was performed for $\alpha = 0.24$, $\beta = 0.02$, and $\gamma = 0$.) Once formed, the distance between the individual solitons grows linearly with $t$. The generic breakup of localized peaks into many solitons is in marked contrast to the experiments considered here, which see only single, localized peaks. Dissipation of sufficient strength can provide an effective mechanism for preventing the formation of unwanted local maxima. This is illustrated in Figs. 1(b)–(d) obtained numerically for $\gamma = 0.01, 0.03$, and 0.2, respectively. Once new local extrema have been formed, dissipation will tend to reduce the local maximum (where the curvature is negative) and enhance the local minimum (where the curvature is positive.) For all values of $\gamma$, the local maximum and minimum slowly merge and ultimately disappear. Given the linear growth of the separation between local maxima and the much longer time scale required for dissipation to eliminate them, it is clear that $\gamma$ should exceed some minimum value, $\gamma_{\text{min}}$, chosen to ensure that secondary maxima never form. The value of $\gamma_{\text{min}}$ is set by a complicated interplay between the initial pulse shape and the parameters appearing in Eq. (10). For the present case of $\lambda = 3$, numerical simulations indicate that $\gamma_{\text{min}} = 0.024$. The initial pulse width considered here is larger than that appropriate to the experimental initial conditions of Refs. [2–5]. A more realistic calculation would involve a value of $\lambda$ that is smaller than (but on the order of) unity. We have used here the value $\lambda = 3$ for convenience. In addition, numerical evidence indicates that $\gamma_{\text{min}}$ grows slowly as the width $\lambda$ decreases.

An interesting feature of our results is that $\gamma_{\text{max}}$ and $\gamma_{\text{min}}$ differ by a large factor (i.e., 40 in the present case.) This suggests that it is not necessary to fine tune the dissipative term in Eq. (10) in order to obtain localized pulse propagation. Indeed, the qualitative impression of solitary pulse propagation seems to be a particularly robust phenomenon, and its observation cannot be regarded as a demonstration of true solitonic propagation.

VIII. TWO EXPERIMENTAL CHALLENGES

The model we have presented can be checked easily since it makes the following two definite predictions:

1) We have noted that, in the presence of dissipation, $\Delta X^2(t)$ grows linearly with $t$. Since $t$ is the time for the pulse to propagate inside the crystal, $\Delta X^2$ should also grow linearly with the size $L$ of the crystal along the direction of pulse propagation (keeping all other conditions fixed.) 2) As we have seen, the propagation of excitons is sensitive to the value of $\gamma$, and thus to their coupling with phonons. If an external stress (of a few kbar) is applied, excitons can couple to transverse acoustic phonons, as discussed in Ref. [8]. The transverse speed of sound $c_t$ is lower than $c_l$, and the corresponding freeze-out temperature is consequently some ten times smaller than $T_l$, i.e., $\approx 0.5$ K [8]. If the same experiments were performed in the presence of stress, the term on the right of Eq. (10) would be enhanced and the motion would become more diffusive with increasing stress. (The conclusions of the other theories [11,12,17] are expected to be less sensitive to external stress.) These tests are likely to provide compelling evidence for or against our picture since the

FIG. 1. Solutions of Eq. (10). We have plotted $\delta n/n_0$ (y axis) as function of distance (x axis), measured in units of $l_p$. The initial disturbance is given by Eq. (16) with $\lambda = 3$. We have used the values $\alpha = 0.24$ and $\beta = 0.02$. The figures show snapshots of the solution for $\gamma = 0$ (a), 0.01 (b), 0.03 (c), and 0.2 (d), at times $t/\tau_p = 10.0, 25.0$, and 50.0.
empirical value of $\gamma$, estimated above, is relatively close to $\gamma_{\text{max}}$.

**IX. SUMMARY**

We have presented a phenomenological theory attributing the formation of localized waves of excitons in Cu$_2$O under certain experimental conditions to the combined effects of dispersion, non-linearity and dissipation. Both analytical estimates and numerical simulations suggest that the characteristic time scale for the crossover between the regimes of localized propagation of the pulse and diffusive motion is comparable to the scattering time for exciton-phonon collisions.

**ACKNOWLEDGMENTS**

G.M.K. is grateful to G. Baym, A. Jolk, M. Jörger, and C. Klingshirn for useful discussions. G.M.K. would also like to thank the Physics Department of the Univ. of Crete, Greece for its hospitality.

* Present address: Mathematical Physics, Lund Institute of Technology, P.O. Box 118, S-22100 Lund, Sweden

[1] D. L. Dextler and R. S. Knox, “Excitons” (John Wiley and Sons, New York, 1965).
[2] E. Fortin, S. Fafard, and A. Mysyrowicz, Phys. Rev. Lett. 70, 3951 (1993).
[3] A. Mysyrowicz, E. Fortin, E. Benson, S. Fafard, and E. Hanamura, Solid State Commun. 92, 957 (1994).
[4] E. Benson, E. Fortin, and A. Mysyrowicz, Phys. Stat. Sol. B 191, 345 (1995).
[5] E. Benson, E. Fortin, B. Prade, and A. Mysyrowicz, Europhys. Lett. 40, 311 (1997).
[6] A. Jolk, M. Jörger, and C. Klingshirn, to be submitted.
[7] D. P. Trauernicht, J. P. Wolfe, and A. Mysyrowicz, Phys. Rev. Lett. 52, 855 (1984).
[8] D. P. Trauernicht and J. P. Wolfe, Phys. Rev. B 33, 8506 (1986).
[9] D. W. Snoke, J. P. Wolfe, and A. Mysyrowicz, Phys. Rev. B 41, 11171 (1990).
[10] J. Scott Russell, Edinb. Roy. Soc. Trans. 14, 47 (1840).
[11] E. Hanamura, Solid State Commun. 91, 889 (1994).
[12] I. Loutsenko and D. Roubtsov, Phys. Rev. Lett. 78, 3011 (1997).
[13] S. A. Moskalenko and D. W. Snoke, “Bose-Einstein Condensation of Excitons” (Cambridge University Press, Cambridge, England, 2000).
[14] S. G. Tikhodeev, Phys. Rev. Lett. 84, 3502 (2000).
[15] J. Shumway and D. Ceperley, Phys. Rev. B 63, 165209 (2001).