Hot exciton transport in ZnSe quantum wells

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The in-plane transport of excitons in ZnSe quantum wells is investigated directly by microphotoluminescence in combination with a solid immersion lens. Due to the strong Fröhlich coupling, the initial kinetic energy of the excitons is well controlled by choosing the excess energy of the excitation laser. When increasing the laser excess energy, we find a general trend of increasing transport length and more importantly a pronounced periodic quenching of the transport length when the excess energy corresponds to multiples of the LO-phonon energy. Such features show the dominant role of the kinetic energy of excitons in the transport process. Together with the excitation intensity dependence of the transport length, we distinguish the phonon wind driven transport of cold excitons and defect-limited hot exciton transport. © 2002 American Institute of Physics.

The in-plane transport of excitons in semiconductor quantum wells (QWs) has attracted much interest due to both fundamental and technological reasons. Generally, there are several possible transport processes after an optical excitation. The first one is the classical diffusion of cold excitons.1 In this case, the excitons have the same temperature as the lattice. The transport can be well described by the diffusion equation, with a constant diffusivity. The second process is the transport of hot excitons,2 initially ballistic (before the first scattering event) and then diffusive. Since the excitons remain hot, the transport is coupled with the relaxation process. This kind of transport cannot be described by the diffusion equation, since the effective “diffusivity” varies in both temporal and spatial domains. Beside these “active” transport processes, which are governed by the velocity of the excitons, the excitons can also be passively driven by other factors, i.e., phonon wind.3 Due to the increasing importance of nanostructures, transport of excitons or carriers has to be understood on a length scale comparable to the mean free path of the particles. It is obvious that strong deviations from classical transport can be expected.

During the past two decades, the transport experiments have concentrated mainly on III–V semiconductor QWs. The employed optical techniques, namely transient grating,4,5 pump–probe,6,7 time-of-flight,8,9 microphotoluminescence (μ-PL),7 and near-field pump–probe,8 have achieved an increasing spatial resolution. The in-plane transport of excitons in II–VI QWs was less investigated, and the transport has been regarded as classical diffusion.9,10

In this letter, we investigate the exciton in-plane transport in ZnSe QWs on the length scale of few μm by solid immersion lens (SIL)-enhanced μ-PL. We show that the exciton transport in this regime is not classical diffusion, but dominated by hot exciton transport. In particular, we exploit the effect that, in contrast to GaAs QWs, the initial kinetic energy of the excitons in ZnSe QWs can be tuned in a well defined manner, due to LO-phonon assisted generation of the excitons.

The confocal μ-PL system consists of a microscope objective (20×, NA=0.4) and a hemisphere SIL11 of refractive index n r=2.2. The SIL is adhesively fixed to the sample surface. The spatial resolution of the whole system was determined to be about 400 nm full width at half maximum [(FWHM) of the Airy pattern]. The excitation source is a cw Ti:sapphire laser pumped by an Ar-ion laser and frequency doubled using a BBO crystal. The laser beam is focused on the sample surface by the objective. The luminescence is collected by the same objective. A 20 μm pinhole in the image plane of the objective limits the detection area to 455 nm in diameter. All measurements were performed at 7 K.

Two samples are studied in this investigation. Sample 1 is 120 periods of ZnSe(7.3 nm)/ZnS 0.1 Se 0.9(10.7 nm) multiple QW grown by MOVPE. Sample 2 is a ZnSe(5 nm) single QW with Zn 0.9 Mg 0.1 Se 0.98 barriers grown by molecular beam epitaxy (MBE). The PL spectra of the two samples (Fig. 1) are dominated by the peaks of the heavy hole excitons (hh), with similar linewidth of about 2 meV(622,869),(772,909). However, we note that the linewidth cannot be used for comparing the quality of the two samples, since the carrier conﬁnements are different due to the different barrier materials. Actually, the luminescence efficiency of sample 2 is two or-

FIG. 1. Photoluminescence of sample 1 (solid) and sample 2 (dots). Inset: An example of the spatial proﬁles of the heavy-hole exciton luminescence (upper squares) and the laser spot (lower squares). The curves represent the corresponding Gaussian ﬁts.

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We note that in the case of excitation since the LO-phonon emission path is not available. Luminescence is very weak due to the inefficient exciton formation. In this case, we can define the exciton initial kinetic energy of the slow relaxation process realized by acoustic phonons until their kinetic energy is lower than the LO-phonon energy, $E_{\text{ini}}$, in which $n$ equals to 1, 2, or 3 depending on $E_{\text{excess}}$. By tuning the $E_{\text{excess}}$, we can periodically tune the $E_{\text{ini}}$, and thus the initial in-plane group velocity of the excitons, to investigate the influence of this velocity on the transport process.

Figure 3 shows the excess energy dependence of the transport length for both samples. When $E_{\text{excess}}<E_{\text{LO}}$, the luminescence is very weak due to the inefficient exciton formation since the LO-phonon emission path is not available. We note that in the case of $E_{\text{excess}} \approx E_{\text{LO}}$, the hh peak is superimposed by a strong resonant Raman scattering peak, so the spatial profile gained in this case is not influenced by the Raman scattering. In Fig. 3, we observe two features: (1) a general trend of increasing transport length with increasing $E_{\text{excess}}$ and more importantly (2) a pronounced periodic quenching of the transport length when $E_{\text{excess}} \approx nE_{\text{LO}}$.

Figure 4 shows the excitation intensity dependence of the transport length of sample 1. Several values of $E_{\text{excess}}$ are chosen in the range of $1E_{\text{LO}}-2E_{\text{LO}}$, and the corresponding values of the $E_{\text{ini}}$ are shown in Fig. 4. We estimate the excitation density from the excitation intensity, as shown in Fig. 4 as the top axis. An absorption coefficient of $6.5 \times 10^3 /\text{cm}$ (measured by absorption spectroscopy) and a decay time of 300 ps (measured by time-resolved photoluminescence) are used for this estimation. In Fig. 4 we find that in the cases of small $E_{\text{ini}}$ (1 and 7 meV), the transport length increases sub-linearly with the excitation intensity. For higher $E_{\text{ini}}$, the transport length is independent of the excitation intensity.

In the case of phonon wind driven transport, the transport length increases with the excitation intensity, while for the classical diffusion of cold excitons and the hot exciton transport, the transport lengths are independent of the excitation intensity. In our experiment, when the $E_{\text{excess}}$ is slightly larger than $nE_{\text{LO}}$, cold excitons are generated with small $E_{\text{ini}}$. According to the excitation intensity dependence behavior, these cold excitons are driven by phonon wind. Increasing the $E_{\text{excess}}$ within one period, we observe the increase of the transport length in Fig. 3. This behavior can be
explained by either the hot exciton transport or the phonon wind driven model. In the former case the \( E_{\text{ini}} \) increases with \( E_{\text{exc}} \), while in the latter case, the force of the phonon wind also increases with the \( E_{\text{exc}} \) due to the increasing of the number of phonons emitted during the relaxation of the excitons. However, the latter possibility can be ruled out by the strong periodic feature observed in Fig. 3. As mentioned above, the \( E_{\text{ini}} \) is a periodic function of the \( E_{\text{exc}} \) with a period of \( E_{\text{LO}} \) due to the fast LO-phonon emission. Thus, the periodic feature can be well explained by the hot exciton transport. In the phonon wind driven model, the wind force is anticipated to be increase monotonously with \( E_{\text{exc}} \), since the LO phonons emitted during the relaxation decay into acoustic phonons within few ps.\(^{13}\) For these reasons, we attribute the transport of the excitons with high \( E_{\text{ini}} \) to the hot exciton transport. Actually, since the phonon wind cannot drive the excitons at drift velocities exceeding the sound velocity,\(^{2}\) the influence of the phonon wind on the hot excitons should be weak. The above explanations are confirmed by the independence of transport length on the excitation intensity for hot excitons, as observed in Fig. 4.

Finally, we discuss briefly the influence of defects on the transport properties. In Fig. 3, we find that the transport length of sample 2 is larger than that of sample 1. The dip around \( E_{\text{LO}} \) is more pronounced in sample 1 than in sample 2, comparing the dips around \( 2E_{\text{LO}} \). In the case of \( E_{\text{exc}} \approx E_{\text{LO}} \) (see \( E_{\text{exc}} \) in Fig. 2), the exciton formation is inefficient due to the small momentum difference. However, the formation can be assisted by the relaxation of momentum conservation due to defects (shown as the dashed line after the \( E_{\text{exc}} \) in Fig. 2). Thus, the dip around \( E_{\text{LO}} \) is anticipated to be more pronounced in the sample containing more defects (sample 1, due to the smaller transport length and the lower luminescence efficiency). Furthermore, the periodic feature of the transport length is more pronounced in sample 1 than in sample 2 (see the dips around \( 3E_{\text{LO}} \)). This coincides with the fact that normally in PLE, the LO-phonon cascades are easier to be observed in the samples containing more defects, and can be attributed to the loss of excitons during the slow relaxation process. The difference of the two samples implies that the hot exciton transport in these samples is limited by defects. This statement is also consistent with calculations of the transport length from exciton acoustic–phonon interaction neglecting defects, which yields expected values of about 10 \( \mu m \).

In summary, we measured directly the in-plane transport of excitons in ZnSe QWs on the length scale of few \( \mu m \) by SIL-enhanced \( \mu-\)PL. Due to the strong Fröhlich coupling, the initial kinetic energy of the excitons is well controlled by tuning the laser excess energy. We find the dominant role of the kinetic energy of excitons in the transport process. The cold excitons are driven by the phonon wind. For the excitons with high initial kinetic energies, the process is dominated by the hot exciton transport. Furthermore, the hot exciton transport is limited by defects.

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