Coherence Length of Excitons in a Semiconductor Quantum Well

Hui Zhao, Sebastian Moehl, and Heinz Kalt

Institut für Angewandte Physik, Universität Karlsruhe, D-76128 Karlsruhe, Germany

We report on the first experimental determination of the coherence length of excitons in semiconductors using the combination of spatially resolved photoluminescence with phonon sideband spectroscopy. The coherence length of excitons in ZnSe quantum wells is determined to be 300–400 nm, about 25–30 times the exciton de Broglie wavelength. With increasing exciton kinetic energy, the coherence length decreases slowly. The discrepancy between the coherence lengths measured and calculated by considering only the acoustic-phonon scattering suggests an important influence of static disorder.

           PACS numbers: 78.67.De, 71.35.–y, 78.55.Et

Coherence is an essential intrinsic property of quantum-mechanical particles. A particle is called coherent if it propagates like a wave packet with well-defined phases for its spectral components. Such particles have properties quite different from classical particles such as the ability to show interference. Constructive interference leads to a macroscopic coherence of an ensemble of particles. This has profound consequences as illustrated by the properties of laser radiation (coherent ensemble of photons) or by the formation of a new state of matter in a Bose-Einstein condensate (coherent ensemble of atoms). The coherence of such an ensemble is destroyed by phase-relaxing processes of individual ensemble members. The temporal and spatial scales of phase-destroying processes are given by the particle coherence time and length, respectively.

The above arguments also hold for excitons, which are the fundamental quasiparticles of optical excitation in semiconductors. Recently, coherent control of both ensemble excitons [1] and individual excitons [2] has been demonstrated, making excitons possible candidates for quantum-information processing [3]. So it is important to study the coherence time and length of excitons, which define an upper temporal and spatial limit for coherent manipulations. Even for spin manipulations, the temporal or spatial decoherence can influence spin coherence whenever spin-orbit coupling is present [4]. In this Letter we present the first experimental determination of the coherence length of excitons in semiconductors.

Excitonic coherence is temporally and spatially limited due to interactions within the exciton ensemble and its coupling to its environment. We can divide these interactions into two classes: elastic and inelastic scattering. They have essentially different influences on the excitonic coherence [5]. For elastic scattering, the direction of the wave vector changes, but the energy of the exciton keeps constant. Consequently, the elastic scattering does not destroy the phase of the exciton. The spatial distribution of the wave function remains independent of time even after several elastic-scattering events. Thus, the exciton wave function remains coherent [5]. One can find an analog of this coherent propagation in the presence of elastic scattering in Anderson localization of electrons in metals [6]. On the contrary, an inelastic-scattering event changes the exciton energy and phase and thus destroys the coherence.

It is now well established how to measure the excitonic coherence time, e.g., from the decay of the macroscopic optical polarization using ultrafast spectroscopy [7] or from the homogeneous linewidth of exciton luminescence [8]. Methods to investigate spatially coherent phenomena are much less developed. They require not only a spatial resolution on the order of the light wavelength [9,10], but also a means to simultaneously test the coherence of the excitons. First attempts towards this goal were based on time- and space-resolved pump-probe experiments [11,12]. But since the nonlinear response of the semiconductor was tested, only high-density regimes beyond the excitonic phase were accessible. We will show in this Letter that we are able to investigate the spatial coherence and measure the coherence length of excitons using spatially resolved phonon sideband spectroscopy in quantum wells based on the polar semiconductor ZnSe.

The investigations require both specific material properties and a novel experimental design. The material of choice is a ZnSe/ZnSSe multiple quantum well for two reasons. First, due to the strong Fröhlich coupling in polar II-VI quantum structures, one can optically generate well-defined hot excitons assisted by the emission of longitudinal optical (LO) phonons within some 100 fs [13], as shown in Fig. 1. The initial kinetic energy of the exciton can be well controlled over a range of 30 meV simply by choosing the laser excitation energy, E_{exc}. This exciton formation mechanism is drastically different from that of III-V structures [14], in which most of the optically generated electrons and holes relax individually toward their respective band extrema and bind to excitons on a time scale of 10 ps [15]. Second, the strong Fröhlich coupling induces a pronounced LO-phonon sideband (PSB) beside the zero-phonon line (ZPL) in the photoluminescence (PL) spectrum. After their formation, the hot excitons relax to the band minimum by phonon emission. The relaxed excitons with nearly zero kinetic energy can radiatively recombine, resulting in the ZPL. Since the photon momentum is very small, this direct coupling to photons is forbidden for hot excitons due to momentum conservation.
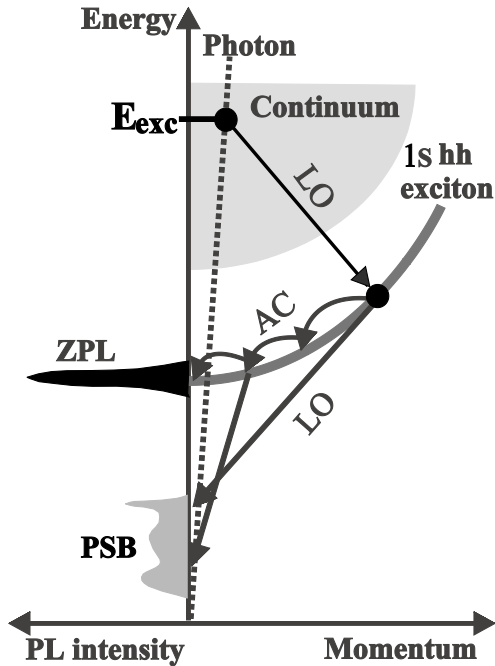
FIG. 1. Schematic drawing of the exciton formation assisted by LO-phonon emission, the subsequent relaxation by acoustic-phonon (AC) emission, and the resulting zero-phonon line (ZPL) and phonon sideband (PSB).

This is the main obstacle of hot-exciton studies in conventional PL spectroscopy, which exploits the ZPL. For the same reason, the hot excitons are not observable in pump-probe experiments. However, the LO phonon can assist the hot exciton in coupling with a photon by taking away its momentum (see Fig. 1). The simultaneous well-defined energy loss to the LO phonon leads to the appearance of the PSB. This process is an ideal tool for the *direct* investigation of hot excitons [13,16].

Figure 2 shows three spectra excited by a continuous-wave laser at various photon energies. The PSB is found in the photon-energy range between one LO-phonon energy ($E_{LO} = 31.8$ meV) below the ZPL and $2E_{LO}$ below the E_{exc} (see Fig. 2). The latter energy defines the upper limit of the PSB, which stays a strict limit when E_{exc} is varied. This fact confirms that the PSB really reflects the hot-exciton distribution and that no additional luminescence, e.g., related to defects, occurs in this spectral range. In Fig. 2, a sharp peak (designated HL) is observed at $2E_{LO}$ below E_{exc}. This peak is the main subject of the present investigation. In the following we will identify its origin and discuss what we can learn from this peak.

Generally, the peak observed at $E_{exc} - nE_{LO}$ can be induced by hot-exciton luminescence (HL) and/or resonant Raman scattering (RRS) [17]. There has been a debate in the 1970s on how to distinguish these two processes [18,19]. The actual difference between them is whether a real excitonic population is involved or not [18]. HL is composed of two distinct one-photon processes, i.e., ab-
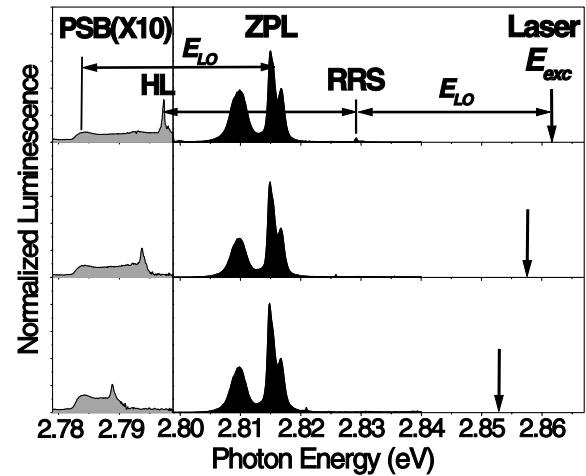

FIG. 2. Photoluminescence spectra measured at 7 K. The excitation intensity is 1 kW/cm^2.

sorption followed by emission, while RRS is a single two-photon process [20]. In the case of $n = 1$, HL is not possible [17], thus the peak can be easily identified as a first order RRS. Indeed, this peak is observed in Fig. 2 on the high-energy side of the ZPL. In the cases of $n \geq 2$, both HL and RRS are possible. In experiments, one can distinguish them according to their different features [17]. Recently, it was proved that HL can be the dominant process at low temperature in several systems, e.g., II-VI [20,21], III-V [22], and IV-IV [23] semiconductors. In our experiments, we prove that the sharp peak observed at the upper limit of PSB is dominated by HL by the following experimental facts. At first, raising the temperature we observe a pronounced thermal quenching of this peak (from 800 counts at 10 K to less than 100 counts at 70 K), while the RRS peak keeps unchanged in this temperature range (about 160 counts). Such different temperature behaviors clearly distinguish HL and RRS [17,23]. Secondly, increasing the photon energy of excitation by E_{LO}, the spectral linewidth of the HL peak becomes 2–3 times wider. This is also a signature of an HL process [21]. Beside these facts, the different spatial profiles of the HL and RRS peaks (see below) also provide evidence of their origins.

Having excluded a significant Raman contribution to the HL peak, we will see that the peak can be exploited to study the individual coherence of excitons. The peak is spectrally located at $2E_{LO}$ below E_{exc}. So it actually monitors the population of excitons right after their LO-phonon assisted formation and before their first inelastic-scattering event. An exciton that has undergone the first inelastic-scattering event thus has a different energy and cannot contribute to the peak. Since an exciton remains coherent between two inelastic-scattering events, *the HL peak at $2E_{LO}$ below E_{exc} monitors the presence of individual coherent excitons* with a well-defined kinetic energy $E_{exc} - E_{LO}$.

Our experimental setup is a solid immersion lens enhanced confocal microphotoluminescence system with a resolution of 450 nm (FWHM). The details of the system have been described previously [24]. By moving a pinhole in the image plane of the microscope, we can scan the detection spot with respect to the excitation spot, thus studying the spatial coherence in a rather direct way. Also, the high collection efficiency of the setup enables us to access the low-density regime where exciton-exciton interactions are negligible.

By scanning the pinhole, we measure the spectra of the PSB at different detection positions with respect to the excitation spot, as shown in Fig. 3(a). We find that the HL peak drops with increasing the excitation-detection distance, but is still visible at a distance as large as 1380 nm. We also find a systematic change of the PSB spectral shape, which reflects the relaxation of the hot excitons during transport. These spectra allow us to obtain the spatial profile of the HL peak, as shown by the squares in Fig. 3(b). We also plot in the same figure the spatial profiles of the laser spot (circles), the RRS peak (stars), and the ZPL (down triangles) measured in the same pinhole scanning. The profile of the RRS peak is similar to that of the laser spot, while the HL peak distribution is significantly wider. We note that the difference in spatial extension can also be used as a method to distinguish the HL and RRS processes. We also check the spatial profile of a spectral component in the middle part of the PSB, as indicated by the arrow in Fig. 3(a). The profile (up triangles) locates between the HL and ZPL profiles, showing the undergoing relaxation. Such a feature is shown quantitatively in Fig. 3(c). Decreasing the detection energy from HL peak to $E_{ZPL} - E_{LO}$, i.e., scanning the arrow in Fig. 3(a) from right to left within the PSB, we find an increase of the HWHM (half width at half maxima, obtained by Gaussian fits) approaching that of ZPL. This confirms that the PSB really monitors the exciton population as well as its spatial extension.

As discussed before, the HL peak monitors the individual coherence of the excitons. Thus the spatial profile of HL peak reflects the coherent propagation of excitons in space. We note that the HWHM of the HL peak is related, but not equal, to the coherence length of the excitons, since both the excitation spot and the detection spot in our measurement are not given by delta functions in space. Using Monte Carlo simulations for deconvolution, we deduce that in our experimental setup the measured HWHM of the HL peak profile is actually 1.35 times the coherence length.

Figure 4 shows the coherence lengths measured in this way for several exciton kinetic energies by tuning the photon energy of the excitation laser. Increasing the exciton kinetic energy, we find a slow drop of the coherence length. The decrease is less than 20% as the kinetic energy is increased from 1.6 to 20.7 meV. We also show in the same figure (dotted line) the coherence length calculated
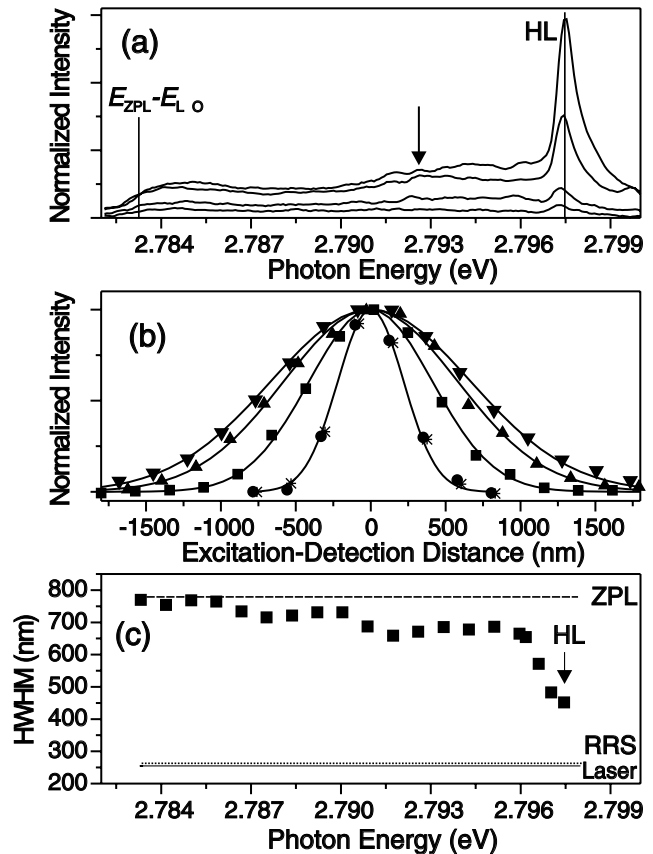

FIG. 3. (a) Spectra of PSB measured at different positions with respect to the excitation-laser spot. The distances from the excitation spot to the detection spot are (from top to bottom) 0, 460, 920, and 1380 nm, respectively. The excitation-laser energy is 2.8611 eV, with an intensity of 1 kW/cm². (b) Spatial profiles of the excitation-laser spot (circles), RRS (stars), HL (squares), and ZPL (down triangles). The spatial profile of one spectral component in the PSB, indicated as the arrow in (a), is also shown as the up triangles. The solid lines represent the corresponding Gaussian fits to the data. (c) HWHM of the spectral components in the PSB as function of the photon energy. The HWHMs are obtained by Gaussian fits of the spatial profiles of the selected components. The HWHMs of the ZPL (dashed line), RRS (dotted line), and laser spot (solid line) are also shown for comparison.

by only considering the acoustic-phonon scattering. For this estimation, the influence of static disorder is not considered; thus, the exciton is assumed to propagate ballistically before the first acoustic-phonon scattering event. The coherence length is thus calculated by simply multiplying the group velocity obtained from a parabolic dispersion and the acoustic-phonon scattering time [25]. The discrepancy observed in Fig. 4 suggests that the excitonic propagation is limited by not only the acoustic-phonon scattering but also by other mechanisms such as static disorder. The importance of the disorder on excitonic properties of semiconductor quantum wells at low temperature has been proven (see, e.g., Ref. [26]). Recently,
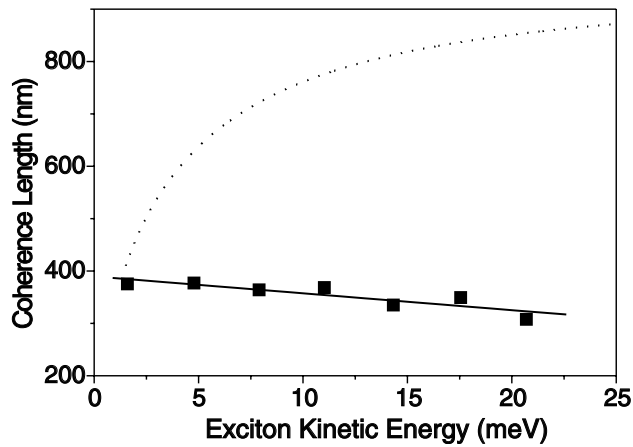
FIG. 4.   Exciton coherence lengths (squares) deduced from the spatial profiles of the HL peaks for several exciton kinetic energies. The solid line is a guide for the eyes. Also shown is the calculated coherence length (dotted line) limited by acoustic-phonon scattering without considering the influence of disorder.

theoretical investigation [27] suggested a strong influence of disorder on the spatiotemporal dynamics of excitons in quantum wells under near-field pulsed excitation. Strong disorder can eventually result in exciton localization. Here we are dealing with excitons with rather high kinetic energy above the effective mobility edge. A detailed theoretical study on the influence of static disorder on the coherence length of excitons is beyond the scope of the present investigation. However, Fig. 4 provides important input for this kind of study. We also note that the coherence length determined from the present investigation (300–400 nm) is about 25–30 times the de Broglie wavelength of the exciton in ZnSe quantum wells.

In summary, we show that the combination of a highly efficient, sub-μm spatially resolved photoluminescence system with phonon sideband spectroscopy enables one to investigate coherence transport process in semiconductors. By monitoring the spatial evolution of the exciton coherence, we directly determine the exciton coherence length of 300–400 nm, which decreases slowly when increasing the exciton kinetic energy.

[1] A. P. Heberle, J. J. Baumberg, and K. Köhler, Phys. Rev. Lett. 75, 2598 (1995).

[2] N. H. Bonadeo, J. Erland, D. Gammon, D. Park, D. S. Katzer, and D. G. Steel, Science 282, 1473 (1998).

[3] G. Chen, N. H. Bonadeo, D. G. Steel, D. Gammon, D. S. Katzer, D. Park, and L. J. Sham, Science 289, 1906 (2000).

[4] D. D. Awschalom and J. M. Kikkawa, Phys. Today 52, No. 6, 33 (1999).

[5] V. V. Mitin, V. A. Kochelap, and M. A. Stroscio, *Quantum Heterostructures* (Cambridge University, Cambridge, 1999), p. 18.

[6] P. A. Lee and T. V. Ramakrishnan, Rev. Mod. Phys. 57, 287 (1985).

[7] J. Shah, *Ultrafast Spectroscopy of Semiconductors and Semiconductor Nanostructures* (Springer-Verlag, Berlin, 1996), Chap. 2.

[8] D. Gammon, E. S. Snow, B. V. Shanabrook, D. S. Katzer, and D. Park, Science 273, 87 (1996).

[9] J. R. Guest, T. H. Stievater, G. Chen, E. A. Tabak, B. G. Orr, D. G. Steel, D. Gammon, and D. S. Katzer, Science 293, 2224 (2001).

[10] D. Sanvitto, F. Pulizzi, A. J. Shields, P. C. M. Christianen, S. N. Holmes, M. Y. Simmons, D. A. Ritchie, J. C. Maan, and M. Pepperd, Science 294, 837 (2001).

[11] M. Vollmer, H. Giessen, W. Stolz, W. W. Rühle, L. Ghislain, and V. Elings, Appl. Phys. Lett. 74, 1791 (1999).

[12] J. Hetzler, A. Brunner, M. Wegener, S. Leu, S. Nau, and W. Stolz, Phys. Status Solidi B 221, 425 (2000).

[13] M. Umlauff, J. Hoffmann, H. Kalt, W. Langbein, J. M. Hvam, M. Scholl, J. Söllner, M. Heuken, B. Jobst, and D. Hommel, Phys. Rev. B 57, 1390 (1998).

[14] K. Siantidis, V. M. Axt, and T. Kuhn, Phys. Rev. B 65, 035303 (2002).

[15] P. W. M. Blom, P. J. van Hall, C. Smit, J. P. Cuypers, and J. H. Wolter, Phys. Rev. Lett. 71, 3878 (1993).

[16] S. Permogorov, in *Excitons*, edited by E. I. Rashba and M. D. Sturge (North-Holland, Amsterdam, 1982), Chap. 5.

[17] S. Permogorov, Phys. Status Solidi B 68, 9 (1975).

[18] Y. R. Shen, Phys. Rev. B 9, 622 (1974).

[19] J. R. Solin and H. Merkelo, Phys. Rev. B 12, 624 (1975).

[20] R. P. Stanley, J. Hegarty, R. Fischer, J. Feldmann, E. O. Göbel, R. D. Feldman, and R. F. Austin, Phys. Rev. Lett. 67, 128 (1991).

[21] N. Pelekanos, J. Ding, Q. Fu, A. V. Nurmikko, S. M. Durbin, M. Kobayashi, and R. L. Gunshor, Phys. Rev. B 43, 9354 (1991).

[22] R. Nötzel, L. Däweritz, N. N. Ledentsov, and K. Ploog, Appl. Phys. Lett. 60, 1615 (1992).

[23] I. G. Ivanov, T. Egilsson, A. Henry, B. Monemar, and E. Janzén, Phys. Rev. B 64, 085203 (2001).

[24] H. Zhao, S. Moehl, S. Wachter, and H. Kalt, Appl. Phys. Lett. 80, 1391 (2002).

[25] T. Takagahara, Phys. Rev. B 31, 6552 (1985).

[26] S. D. Baranovskii, U. Doerr, P. Thomas, A. Naumov, and W. Gebhardt, Phys. Rev. B 48, 17 149 (1993).

[27] B. Hanewinkel, A. Knorr, P. Thomas, and S. W. Koch, Phys. Rev. B 60, 8975 (1999).