Radiative lifetime and dephasing of excitons studied by femtosecond time resolved intersubband spectroscopy

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Abstract. We used time resolved photoinduced intersubband absorption excitation spectroscopy in order to measure the dynamics of resonantly photoexcited excitons in multi-quantum well and superlattice samples. We show that they decay radiatively, much faster than thermalized excitons, and that their electron momentum is initially concentrated around the superlattice Brillouin zone center. Later, their electron momentum gradually spreads towards the zone edge, in which case the excitons cannot recombine radiatively.

In geometrically restricted semiconductor quantum structures, such as quantum wells (QWs) and superlattices (SLs), the translation symmetry is removed in one direction. Therefore, excitons with in-plane momentum, which is smaller than the momentum of light in the host crystal, can decay radiatively within their intrinsic radiative lifetime [1]. Accordingly, there is a marked difference between excitonic resonant excitation, in which excitons recombine radiatively immediately after pulse excitation, and non-resonant excitation, in which dephasing must occur before recombination kicks-in. Visible-infrared (IR) [2] and visible-TeraHertz [3] dual beam time-resolved spectroscopy were used recently, in order to study these processes directly. Our method is based on a 500 femtoseconds (fs) pulse excitation by tunable interband laser pulse, followed by 500 fs IR probe pulse, tuned to the intersubband (ISB) resonances of the system. The ISB probe pulse induces an optical transition of photoexcited electrons. Hence, it is very sensitive to the momentum of the carrier whose optical transition is being induced.[2] ISBA spectroscopy in SLs provides a sensitive tool for probing the electronic momentum component along the SL symmetry axis.

The $GaAs/Al_{0.33}Ga_{0.67}As$ heterostructures were grown by molecular beam epitaxy on a (100)-oriented GaAs substrate. They consisted of 33 periods of undoped GaAs quantum wells of thickness 6nm and $Al_{0.33}Ga_{0.67}As$ barriers of thickness 14.7 nm and 6.2 nm for the multi-QW (MQW) sample and for the SL sample, respectively. The thicknesses of the layers were measured by high resolution X-ray diffraction. Two edges of the sample were polished at 45^o to the growth axis, in order to form a waveguide for the mid-IR radiation with an electric field component parallel to the growth direction. The measurements were done with the sample mounted on a cold finger cryostat at $\simeq$15 K. We estimated an exciton density per period of $\simeq 2\times 10^{10} cm^{-2}$ from the measured beam average power, repetition rate, and spot diameter on the sample, together with the calculated interband absorption coefficient [4]. Fig. 1 summarizes the measured experimental data from both samples. In Fig. 1a and Fig. 1c the cw measured photoinduced ISBA spectra of the MQW and of the SL sample, respectively, are shown. In Fig. 1b and Fig. 1d we present the temporally integrated PL and PL excitation (PLE) spectra together with the time resolved, photoinduced, intersubband absorption (ISBA) excitation (PIAE) spectra for both samples. The spectra are normalized at their highest energy edge ($\simeq$1.64 eV).

We note that the PLE spectrum and the 3 ps PIAE spectrum of the MQW sample are almost identical, while the 16 ps PIAE spectrum is different. We clearly observe that the PIAE spectrum evolves with time in such a way that the relative intensity of the heavy hole exciton (HH_X) resonance is reduced. We attribute this reduction to radiative losses of low in-plane momentum excitons. The inset summarizes the temporal dependence of the HH_X resonance intensity on a logarithmic time scale.

The ISBA is a direct measure of the density of electrons in the first electronic subband of the QWs. Thus, the initial fast decay and the later slower decay of the PIAE excitonic resonance, demonstrate that resonantly excited excitons, "remember" their history, and they decay faster than non resonantly excited electron-hole (e-h) pairs. The SL sample offers further insight into this "excitonic memory". The spectral shape of the optical transition between the first electronic miniband, and the second miniband is mainly due to the dispersion of the

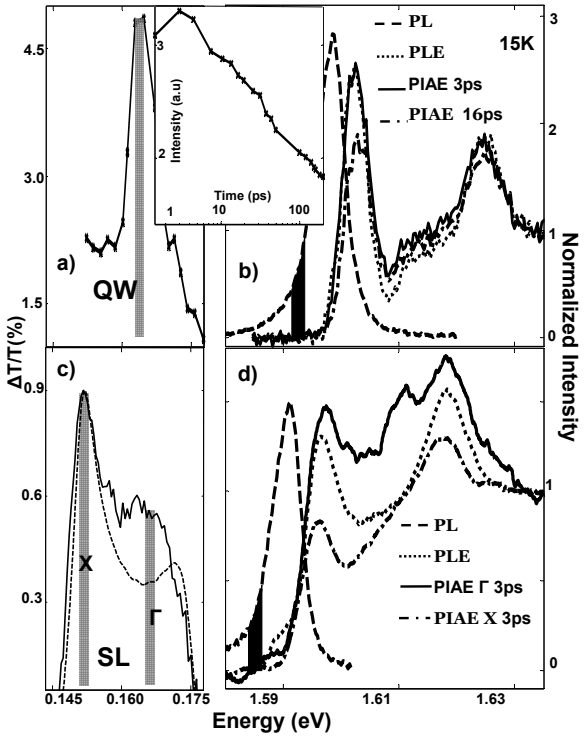

FIGURE 1. a),(c)) Temporally integrated photoinduced ISBA spectrum of the MQW (SL) sample. The dashed line in (c) represents thermal equilibrium calculations after Ref.[4]. b),(d)) Temporally integrated PL and PLE spectra together with time resolved ISBA excitation spectra for the MQW (SL) sample. The shadowed areas under the PL and under the ISBA spectra indicate the spectral ranges from which the PL and ISBA excitation spectra, respectively, were obtained. Inset: The exciton resonance intensity vs. time after the excitation pulse for the MQW sample.

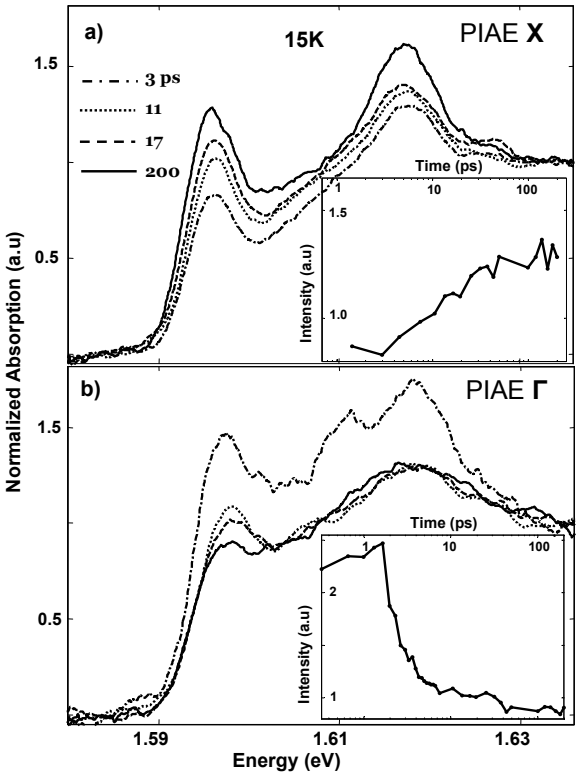

FIGURE 2. a),(b)) Time resolved PIAE spectra for various times after the excitation pulse for the SL sample probed at the X (Γ) point of the ISBA transition. Insets: The exciton resonance intensity vs. time after the excitation pulse.

second miniband along the growth axis. In particular, the SLs zone center (Γ) and zone edge (X) transitions are clearly identified spectroscopically (see Fig.1c), thus correlating the ISB transition energy and the momentum states of the probed photoexcited electron in the first miniband. By probing at different ISB energies, the population of resonant excitons with different momenta along the SL symmetry axis is measured. This is demonstrated in Fig. 1d, in which the PL and PLE spectra of the SL sample are quite similar to those of the MQW sample, but the 3 ps PIAE spectra are vastly different. In the PIAE spectrum probed at the X point, the excitonic resonance is much weaker than that observed in the PLE spectrum, while in the PIAE from the Γ point the excitonic resonances are stronger.

In Fig. 2a (2b) we present PIAE spectra probed at the X (Γ) ISBA energy for various times after the excitation pulse. The temporal dependence of the excitonic resonance is displayed in the insets. It is clearly seen that the excitonic strength increases with time when probed at the X point and drastically decreases with time when probed at the Γ point. Our observations can be qualitatively understood in terms of the electronic component of the excitonic momentum. For resonantly excited excitons, this component is initially concentrated around the superlattice Brillouin zone center as clearly shown by our measurements. This momentum redistributes in time, such that it gradually increases around the zone edge. At the zone edge the radiative decay of the excitons is prohibited, and therefore, at long times the strength of the resonance grows back to its original strength at the moment of excitation.

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REFERENCES

1. B. Deveaud et al., Phys. Rev. Letters **67**, 2355 (1991).
2. R. Duer et al., Phys. Rev. Letters **78**, 3919 (1997)
3. R.A. Kaindl et al., Nature **423**, 734 (2003).
4. D. Gershoni et al., IEEE J. QE, **29**, 2433 (1993).