Determination of the Intrinsic Radiative Lifetime of Quantum Well Excitons by Time-Resolved Intersubband Absorption Excitation Spectroscopy

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Abstract. We demonstrate a new method for measuring the intrinsic radiative life time of 2D excitons in semiconductor quantum structures. The method utilizes time resolved intersubband absorption photoinduced by interband picosecond pulse excitation. Measuring concurrently, the time evolution of the excitation spectra of the photoinduced intersubband absorption and that of the photoluminescence, we find that the heavy hole exciton resonance as measured by the intersubband absorption loses intensity much faster than the decay of the photoluminescence. Using a simple rate equations model, we demonstrate that our observation provides a novel way of directly measuring the excitonic intrinsic radiative lifetime, which is $\approx$20 ps for the 6 nm wide GaAs/GaAlAs quantum well sample that we studied. Our measurements reveal also that the exciton resonance is formed within 8 picoseconds after the resonant pulse excitation.

1. Introduction

In intrinsic direct bandgap semiconductors the interaction of excitons with the electromagnetic radiation field leads to a stationary state called exciton-polariton. This elementary excitation is a coherent state of the entire macroscopic three dimensional (3D) crystal, and as such it does not decay radiatively [1]. In geometrically restricted semiconductor quantum structures, the translational symmetry of the crystalline potential is removed in at least one direction. As a result, the 3D photon field interacts with excitons of lower dimensionality. Thus, in quantum wells, a 2D exciton with an in-plane momentum smaller than the momentum of the 3D photon of same energy, can decay radiatively within its intrinsic radiative life time of few tens picoseconds [2, 3, 4]. The excitonic intrinsic radiative lifetime was so far inferred rather indirectly, from time resolved photoluminescence (PL) measurements [5, 6, 7]. Here, instead, we use visible–infrared (IR) dual beam time-resolved absorption excitation spectroscopy together with PL excitation spectroscopy to directly measure this intrinsic excitonic radiative lifetime.

2. Experimental

The sample was grown by molecular beam epitaxy on a (100)-oriented GaAs substrate. It consists 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. 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 ≃15 K. The details of the time resolved PIA setup are described elsewhere[9]. The setup allows us to spectrally tune the IR probe pulse and to variably delay it relative to the visible pump pulse. The temporal resolution of the system is ≃4 ps, and the spectral resolution is better than 0.5 meV, both for the pump and for the probe energies. We estimate the exciton density per period from the measured beam average power, repetition rate, and spot diameter on the sample. These estimates are corroborated by comparisons between the measured and calculated intersubband absorption coefficient [8]. Thus, by measuring the strength of the differential photoinduced intersubband absorption (ISBA), following a pulsed resonant or non resonant optical excitation, we directly measure the population of photoexcited electrons in the structure's first conduction subband [9]. In Fig. 1 we present the temporally integrated

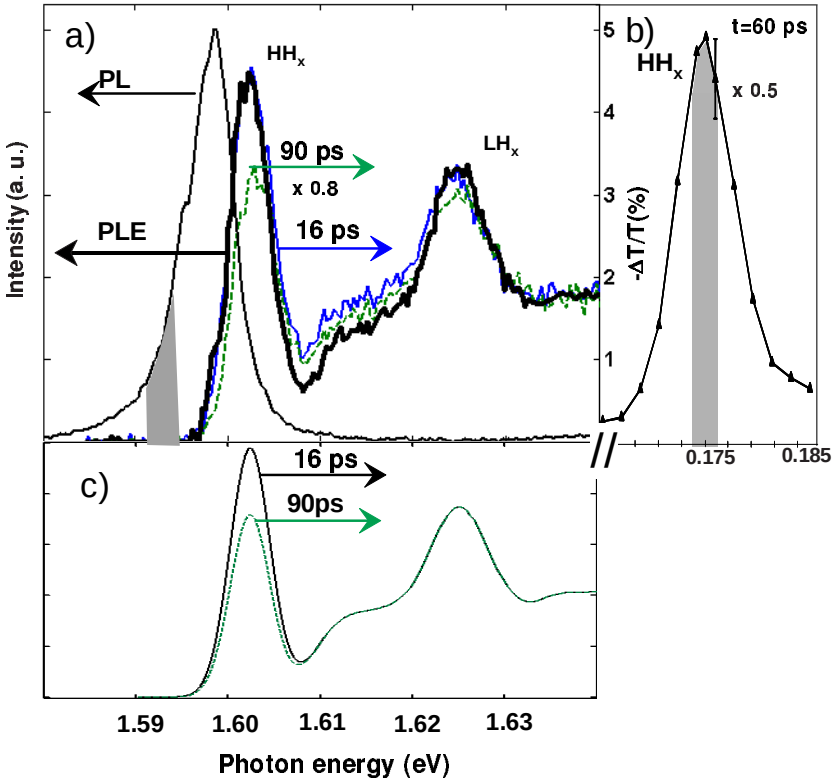

Figure 1. a): Temporally integrated PL and PLE spectra (left axis) together with time resolved ISBA excitation spectra for two times after the excitation pulse (right axis). b): Photoinduced ISBA spectrum at resonant HH excitation 60 ps after the excitation pulse. The shadowed areas under the PL and under the ISBA spectra indicate the spectral ranges from which the PL and ISBA excitation spectra, respectively, were obtained. c): Best theoretical fit to the time resolved photoinduced ISBA excitation spectra (see text)

PL and PLE spectra (left axis) together with time resolved ISBA excitation spectra for 15 and for 90 ps after the excitation pulse (right axis). In the right panel the photoinduced ISBA spectrum at resonant heavy hole (HH) excitation, 60 psec after the excitation pulse is shown. The shadowed areas under the PL and under the ISBA spectrum indicate the spectral ranges from which the PLE and intersubband photoinduced absorption excitation (PIAE) spectra, respectively, were obtained. The three spectra are normalized at energy of 1.64 eV. We note that the PLE spectrum and the 15 ps PIAE spectrum are almost identical, while the 90 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 (HHx) resonance is reduced.

Thus, while at 15 ps the relative intensity of the resonance equals that observed in the PLE spectrum, at later times it significantly loses intensity.

In order to accurately quantify the rate by which the HHx exciton loses intensity with time, we carefully measured ISBA transients for each excitation energy around the HHx resonance. A typical such transient is depicted in the inset to Fig. 2. Four different time domains are observed in the transients.

(i) During the first time domain, at negative delay times, the probe beam comes before the pump and thus E_1 is not populated and the PIA signal is negligible.

(ii) During the second time domain a fast rise of the PIA signal due to the photogeneration of excitons by the 3 ps long laser pulse is observed.

(iii) At the end of the pulse a third region of slower rise, of 30–100 ps long duration, is observed. We believe that this is the time it takes for the photogenerated carrier population to reach thermal equilibrium at the lattice temperature [10, 11]. During this process the PIA signal rises, probably due to the intersubband oscillator transition strength increase of the thermally distributed excitons [12].

(iv) In the fourth domain, the photogenerated population decays by radiative recombination of excitons, on a sub nanosecond time scale as expected by the exciton-polariton model [4, 13]. The fitted effective decay time at long delays is 410 ps as shown by the overlaid solid line.

The time resolved photoinduced ISBA excitation spectra in Fig. 2 were generated from a set of similar transient measurements at various excitation energies. This procedure provides more accurate pulse onset determination and better control over the excitation intensity. As in Fig. 1, the various time resolved spectra in Fig. 2 are normalized at the highest measured energy. In this way the evolution of the spectrum with time is separated from the overall intensity variations of the PIA spectra.

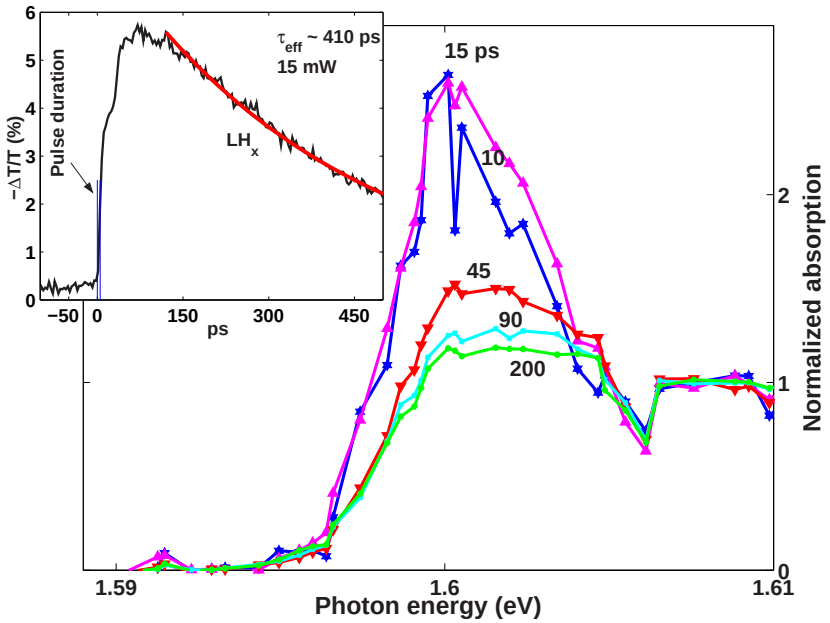

Figure 2. Normalized PIAE spectra for various times after the pulse excitation, as obtained from transient measurements of PIA for various excitation energies.

Fig. 2 clearly demonstrate that the relative intensity of the HHx resonance reaches maximum within the first 10 ps after the excitation. It then quickly decays within the following 90 ps,

after which it remains almost unchanged as long as the photoinduced ISBA could have been measured. In order to quantify the temporal evolution of the HHx resonance we spectrally integrated its normalized PIAE intensity as shown in Fig. 2 for various delay times. We then subtracted the steady-state integrated intensity (as measured at longer than 120 ps times) and plotted the resulting integrated intensity, ΔI_{HHx}, as a function of the time elapsed after the excitation pulse in Fig. 3 (dotted line).

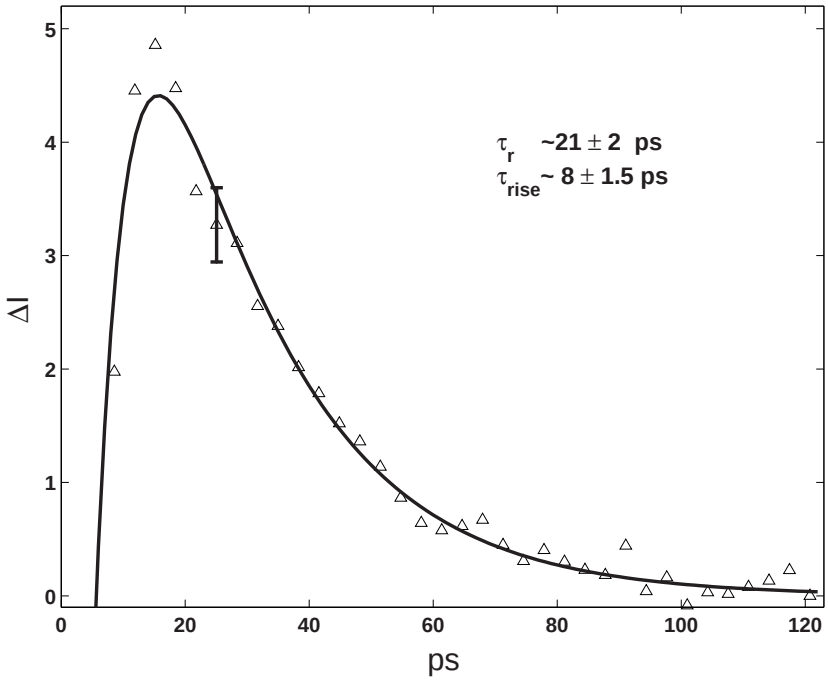

Figure 3. The spectrally integrated intensity of the HH_1 exciton resonance and a phenomenological model fit of Eq.1 (solid line).

The resulting temporal dependence of ΔI_{HHx} can be approximated by a simple rise and decay time model of the form:

$$\Delta I_{HHx} \propto \left(1 - a\, e^{-\frac{t}{\tau_{rise}}}\right) e^{-\frac{t}{\tau_{decay}}} \ . \tag{1}$$

The best fit to the experimental data is overlaid on the data in Fig.3, with the following values for the parameters: $t_{rise} = 8 \pm 1.5$ps, $t_{decay} = 21 \pm 2$ ps.

The meaning of the rise time is that the exciton formation, as measured by the ISBA technique above, is not instantaneous with the laser pulse. Similar observation using interband measurements are explained in terms of emission and recombination processes [14], intrinsic for these experiments, but bypassed by the use of the ISBA technique, here. Thus, the origin of this rise time is not fully understood. We believe, that this rise time has to do with the time it takes for the resonantly photogenerated HHx to distribute within their inhomogeneously broadened resonance [16]. Indeed, when the temporal analysis of the HHx exciton is performed without the spectral integration over the HHx excitonic resonance (not shown), much shorter rise times are deduced.

The decay time that we measure, however, can be clearly interpreted in terms of the intrinsic radiative lifetime of the 2D excitons as discussed below.

3. Discussion

Our measurements can be modeled by simple rate equations, with minimal assumptions. We first assume that the photogenerated e - h population can only decay radiatively, and second, that radiative decay takes place only via annihilation of excitons. Since the radiative recombination of excitons is only allowed for exciton states with in plane momentum $k_{\|} < k_0$, where k_0 is the photon momentum in the matter, [3, 4, 13] we separate the total exciton population into two sub-groups, denoted n_0 - with $k_{\|} < k_0$ and n_k with $k_{\|} > k_0$, respectively. The ISBA intensity, which is proportional to the number of electrons within the first conduction subband, is therefore proportional to n_0+n_k. Under these simple assumptions the temporal evolution of the two exciton sub-groups can be described by two coupled rate equations :

$$\frac{dn_0}{dt} = -\frac{n_0}{\tau_r} - \frac{n_0}{\tau_{k^+}} + \frac{n_k}{\tau_{k^-}} \tag{2}$$

$$\frac{dn_k}{dt} = \frac{n_0}{\tau_{k^+}} - \frac{n_k}{\tau_{k^-}} \tag{3}$$

where τ_{k^+} (τ_{k^-}) characterizes the transfer of excitons from n_0 (n_k) to n_k (n_0) population and τ_r is the radiative decay time (see inset to Fig. 4). The initial conditions of our system is determined by the type of excitation. For resonant excitation into the HHx line, the initial conditions are: $n_0(0) = N, n_k(0) = 0$, while at any non resonant excitation $n_0(0) = 0, n_k(0) = N$. It is straightforward to show that at long times (relative to τ_r) after pulse excitation, regardless of the initial conditions, the total population decays with an effective decay time τ_{eff}, given by: $\tau_{\text{eff}} = (A+1)\tau_r$ where A is the ratio between the two sub groups at long times, given by,

$$A = \lim_{t\to\infty}\left(\frac{n_k}{n_0}\right) \approx \frac{\int_0^{\varepsilon_0/k_BT} e^{-x}dx}{\int_{\varepsilon_0/k_BT}^{\infty} e^{-x}dx} \approx \frac{k_BT}{\varepsilon_0}\ . \tag{4}$$

Here $\varepsilon_0 = \hbar k_0/n - Eg \ll k_BT$ is the maximal kinetic energy of n_0 excitons. Thus τ_{eff} is eliminated (in Eq. 2), since it either can be calculated if the exciton mass and sample temperatures are accurately known [4, 13], or simply deduced directly from our measurements. We analytically solved the rate equations. The temporal evolution of the total exciton population for resonant and non resonant excitations using typical characteristic times of $\tau_{\text{eff}} = 420ps$,$\tau_r = 20$ ps and $\tau_{k^-} = 120$ ps are shown in Fig. 4. Our simple model clearly mimics the experimental observations and it shows that at resonant excitation the total population decays faster at short time and that after about 50 ps it continues to decay with τ_{eff}, which is common for both resonant and non resonant excitations.
Under the assumption that the thermalization processes are much faster than the radiative decay, one may safely deduce that the efficiency of the excitation transfer to the PL emission is independent of the excitation energy. Therefore, it follows that the measured continuous wave (cw) PLE spectrum is directly proportional to the absorption spectrum, $\alpha(\varepsilon)$ [15].

In order to handle $\alpha(\varepsilon)$ analytically we have fitted the PLE spectrum to a compound line shape including two Gaussians, one at the HH and the second at the light hole (LH) exciton resonance, and two broadened "step" functions at their Density of the States onsets. ¿From the measured absorption, we can now calculate the time resolved photoinduced ISBA spectra, by solving the above rate equations for each excitation energy ε, where resonant and non resonant excitations are weighted by the relative magnitude of the HHx resonance at this energy. The calculated spectra are displayed for comparison with the measured spectra in Fig. 1c. The quality of the fits in Fig. 1c and our ability to quite accurately, quantitatively describe the PIAE spectrum and its temporal evolution, give an additional overall strength to our model.

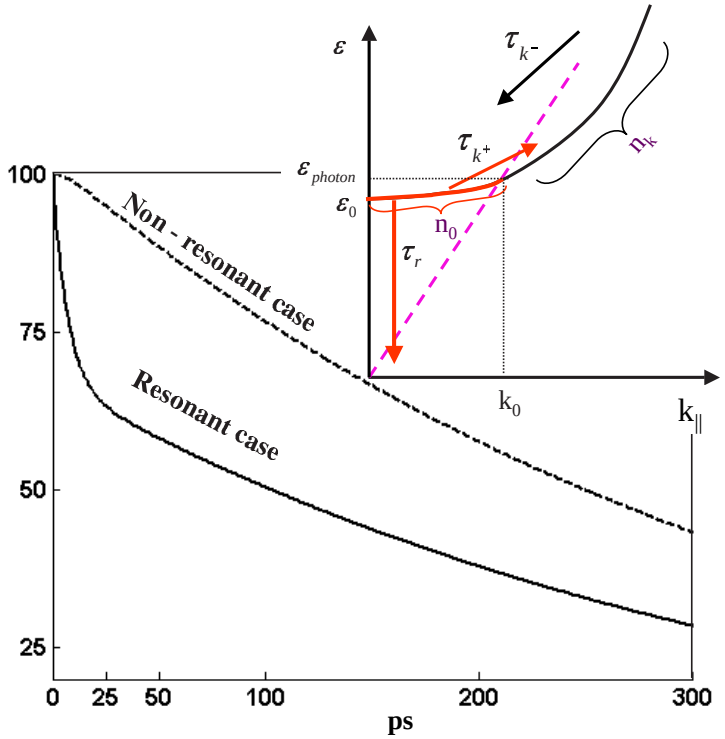

Figure 4. Analytical solution for the rate equation a) at resonant excitation, b) at non - resonant excitation, with characteristic times τ_r = 20 ps and τ_k = 120 ps, and measured τ_{eff} ≈400 ps. Inset shows a schematic representation of the various characteristic times involved in the rate equation model

4. Summary

We demonstrated in this work a new method for measuring the intrinsic radiative lifetime of 2D excitons. The method utilizes time resolved measurements of the intersubband absorption, photoinduced by interband picosecond pulse excitation. These measurements directly follow the dynamics of resonantly and non-resonantly photoexcited carriers in $GaAs/AlGaAs$ quantum structures on a sub nanosecond time scale with temporal resolution of a few picoseconds.

Our main results can be summarized as follows:

The excitation spectrum of the photoinduced intersubband absorption (PIAE) closely resembles the excitation spectrum of the photoluminescence (PLE). In particular, at very short time after the excitation pulse, the two are almost identical. At later times the heavy hole exciton resonance in the excitation spectrum of the intersubband transition lose strength quite rapidly, and it deviates quite significantly from the strength of the resonance as measured in the excitation spectrum of the photoluminescence. The decrease in the strength of the exciton resonance stops after 90 -120 picoseconds, after which the photoexcited carriers are at thermal distribution.

These results can be quantitatively explained by a simple rate equation model, which distinguishes between two populations of excitons. One population, having smaller momentum than that of light in the semiconductor, can decay radiatively, and the second, with higher momentum, and consequently, it cannot decay radiatively. Resonant and off resonant excitations, determine the initial conditions of these populations.

¿From the experimentally deduced temporal evolution of the exciton resonance we find

that the intrinsic radiative lifetime of 2D excitons in 6 nm GaAs quantum well is 21 ± 2 ps.

We also observed that the HH exciton resonance formation, as evidenced by the electronic intersubband absorption is not instantaneous. Its formation within 8 ± 1.5ps, can be probably attributed to exciton-exciton scattering times[16].

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