Separation of inhomogeneous and homogeneous broadening manifestations in InGaAs/GaAs quantum wells by time-resolved four-wave mixing

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Abstract. Coherent dynamics of excitons in single InGaAs/GaAs quantum well was investigated experimentally. In order to carry out this study a three-pulse setup capable of measuring time-resolved four-wave mixing was build. Measured signal consists of two distinguishable components of similar amplitude corresponding to the fast decaying free polarization decay and the long-lived primary photon echo, which proves comparability of homogenous and inhomogeneous broadenings of the exciton resonance.

1. Introduction

Coherent optical control of localized states in semiconductor nanostructures has promising prospective in optical information processing [1, 2]. Moreover, experiments involving coherent control conveniently provide information on fundamental properties of optical excitations in media under study. The time-resolved four-wave mixing (FWM) could be used in order to explore coherent optical dynamics. This powerful method is able to separate inhomogeneous and homogeneous broadening manifestations [3], to distinguish polarization interference from the quantum beats [4], and to determine important coherent properties such as irreversible ($T_1$) and reversible ($T_2^*$) dephasing times.

In recent years many new systems were studied using FWM-based techniques including halide perovskites [5], transition metal dichalcogenide monolayers [6] InGaAs/GaAs quantum dots embedded in Tamm-plasmon microcavity [7]. The one thing in common in all these experiments is a prevalence of the inhomogeneous broadening of studied resonances over the homogeneous linewidth measured at low temperatures.

On the other hand, there is a special interest in semiconductor nano-heterostructures where inhomogeneous broadening becomes comparable with homogeneous linewidth or even less. Fabrication of such structures requires the usage of the molecular beam epitaxy growth method. Recently we have shown successful growth of InGaAs/GaAs single quantum wells with inhomogeneous linewidth smaller than the homogeneous one [8]. Comparable inhomogeneous and
homogeneous linewidths could give rise to the phenomena such as coexistence of the free polarization decay (FPD) and the photon echo (PE) in the same experiment [4].

In this work we present a time-resolved study of four-wave mixing from InGaAs/GaAs single quantum wells.

2. Sample and experimental setup

The sample P551 under study was grown by the molecular beam epitaxy [8]. It contains single 3 nm thick In$_{0.03}$Ga$_{0.97}$As/GaAs QW. The sample was rotated during the growth to achieve better uniformity across its surface. Basic characterization of the sample was done by the reflection spectroscopy in the Brewster geometry [8].

The experimental setup (see figure 1) is designed to detect the time-resolved degenerate FWM signal in reflection geometry. Optical excitation was performed by few-picosecond laser pulses. This regime is optimal for spectral separation of the coherent dynamics of individual quasiparticle resonance. Laser spectrum is monitored by spectrometer with CCD-detector. In our experiments laser was tuned to the heavy-hole exciton resonance. Laser beam is split into three paths denoted in the figure 1 as 1st, 2nd and reference (Ref) beams. 2nd and Ref pulses pass through optical delay lines. 1st and 2nd pulses are focused by a concave mirror to the same spot on the surface of the sample cooled down to $T=1.4$ K in a closed-cycle helium cryostat. Coherent response from the sample (Signal in figure 1) is comprised of several components – reflected 1st and 2nd pulses with wavevectors $k_1$ and $k_2$ respectively, and FWM response with wavevector $(2k_2-k_1)$. After being collimated by the same spherical concave mirror these components can be separated by shifting transversally the retroreflector installed after the mirror. Selected component is mixed with Ref pulse on a non-polarizing beamsplitter. The pair of mixed beams exiting beamsplitter is focused on the balanced photodetector.

![Figure 1. The scheme of the experimental setup.](image)

The optical frequencies of 1st and Ref beams are shifted by two optical shifters based on travelling wave acousto-optic modulators with independently selected radio frequencies. Two beam frequencies are shifted in opposite directions so that interference signal detected by the balanced photodetector is modulated on the differential frequency. Additional signal-to-noise improvement is achieved by a slow modulation of the 1st pulse by mechanical chopper. Such a dual modulation provides high-sensitive background-free detection of the FWM amplitude.
Figure 2 illustrates spectral- and time-domain profiles of the laser pulse. The spectral width is about 900 µeV and temporal duration extracted from auto-correlation function is approximately 3.1 ps. Time-bandwidth product corresponds to the nearly transformed-limited pulse.

3. Results and discussion

Figure 3. Temporal profile of FWM signal as a function of delay between pumping pulses.

The set of temporal profiles of FWM signal from excitons detected at a number of delays between the 1st and 2nd pulses $\tau_{12}$ is illustrated on the figure 3 with delays indicated in picoseconds (note the logarithmic Y axis). At delay $\tau_{12} \geq 20$ ps the FWM signal is comprised of two distinguishable features.
The first one is prompt with respect to the second pulse and corresponds to the free polarization decay (FPD). The second one is delayed and has maximum at $\tau_{\text{Ref}} = 2\tau_{12}$ corresponding to the two-pulse primary PE. Similar temporal behavior was detected in GaAs/AlGaAs multiple QW [3]. Temporal width of PE is about 10 ps revealing relatively small value of inhomogeneity which generally agrees with previous study of the same sample by another technique [8].

4. Conclusion

Experimental study of the coherent optical dynamics of excitons in InGaAs/GaAs quantum well was performed. Time-resolved four-wave mixing demonstrates coexistence of the free-polarization decay and the photon echo signals. We explain this behavior as clear manifestation of comparability of inhomogeneous and homogeneous broadenings of the exciton resonance. This fact indicates high optical quality of the quantum well sample.

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