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Coherent Absorption Spectroscopy with Supercontinuum for Semiconductor Quantum Well Structure

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We suggest that supercontinuum can be used for absorption spectroscopy to observe the exciton levels of a semiconductor nano-structure. Exciton absorption spectrum of a GaAs/AlGaAs quantum well was observed using supercontinuum generated by a microstructrured fiber pumped by a femtosecond (fs) pulsed laser. Significantly narrower peaks were observed in the absorption spectrum from 11 K up to room temperature than photoluminescence (PL) spectrum peaks. Because supercontinuum is coherent light and can readily provide high enough intensity, this method can provide a coherent ultra-broad band light source to identify exciton levels in semiconductors, and be applicable to coherent nonlinear spectroscopy such as electromagnetically induced transparency (EIT), lasing without inversion (LWI) and coherent photon control in semiconductor quantum structures.

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Coherent control of a single photon for quantum information has attracted much attention in coherently prepared atomic systems. Slow photon propagation in electromagnetically induced transparency (EIT) [1], photon storage [2], correlated two photon generation [3] and coherent photon manipulation techniques [4-6] have been demonstrated experimentally. Recently, quantum optics in semiconductor nanostructures has also gained special attention because it is expected to realize quantum information techniques in semiconductors. A single photon was generated in quantum dots [7], and EIT [8] and lasing without inversion (LWI) [9] were observed in GaAs quantum well (QW) structures.

The EIT effect can be easily demonstrated in alkali atom transition lines, for example, Rubidium D_1, with coupling and probe laser beams using an extended cavity diode laser. The natural linewidth of Rubidium D_1 transition is about 6 MHz in the frequency domain. This transition can be easily resolved with an extended cavity CW diode laser system of which the typical linewidth is narrower than 1 MHz. Meanwhile, for nano-sized semiconductor QW structures, however, much higher intensity is needed to provide enough Rabi frequency for observing the dressed states splitting. The linewidth of exciton transition in a well fabricated GaAs QW system is typically in the order of several hundreds GHz ($\sim$1 nm in wavelength domain). It would be very difficult to get the dressed state splitting in such broad spectral linewidth with a CW coupling laser. Using a femtosecond (fs) pulsed laser as a light source is an alternate way of providing the high enough intensity required for nano-sized QW structures with the minimal thermal effects. The linewidth of fs pulsed lasers is usually broad enough to cover the whole linewidth of exciton transitions providing the high peak intensity. In order to provide a desired laser light source for coherent nonlinear spectroscopy, pulse shaping with a grating (s) and a spatial filter is commonly used to narrow down the linewidth of the light source to resolve these exciton transition lines [8]. In this case, the resolution depends on grating resolution and the effective slit size of spatial filtering. Selection of different wavelengths can be accomplished by scanning the slit in a transverse direction using a mechanical stepping motor.

In this paper, we suggest efficient coherent absorption spectroscopy with supercontinuum (SC) which can be applied to quantum optics experiments in semiconductor nanostructures. Using the SC light source, wavelength scanning of the laser source is not needed at all to observe different levels of exciton transitions in quantum well structure because SC light is extremely broad in spectrum. The SC used in this work was generated by a microstructured fiber pumped by a fs pulsed laser. By adjusting the coupling of laser light to the fiber, the spectral range could reach hundreds of nanometers covering UV to IR (300~1500 nm). The absorption spectrum of a GaAs QW structure was observed with a spectrometer. We compared this SC absorption spectrum with the photoluminescence (PL) spectrum measured using 350 nm fs pulsed excitation. It should be noted that normal arc lamps or flash lamps used in conventional absorption spectroscopic measurement are incoherent light, not suitable for coherent spectroscopy. The major advantage of SC light sources over conventional lamp sources is that the SC is coherent light, which can induce interference effects with the pumping pulsed laser lights [10]. Thus, SC light can be an ideal light source for observing coherent processes in quantum nanostructure by coupling with the original pumping laser.

Since SC was first introduced by Alfano *et al.* in 1970 with picosecond pulses in glass [11], many kinds of nonlinear materials have been used to generate SC, even including gases [12] and liquids [13]. We used a pure silica microstructured optical fiber to generate SC, which is easy to handle and can be integrated with other optical fiber based components for optical communication. Pure silica fibers having periodic arrays of microscopic air holes in the cladding region allow the strong light confinement in the core due to the high index contrast between the core and the cladding [14]. It has been demonstrated that the high nonlinearity induced by the strong light confinement and the controllable dispersion properties can give a rise to ultra-wide SC spectra [15]. Pure silica fibers having irregular arrays of air holes in the cladding region, so called irregularly microstructured fiber, can also guide the light due to the high index contrast between the core and the cladding and tightly confine the light in the core region. Fig. 1 shows the scanning electron microscopic image of the cross section of the fiber used in our experiment. Air holes are irregularly arrayed and their sizes are also irregular. The largest (smallest) size of the air holes is about 1.7 (0.8) μm. The elliptic core has a long (short) axis of about 2.0 (1.0) μm. Because of elliptical shape of the core, this microstructured fiber maintains the polarization of the pumping laser light in generating the polarization maintained SC light. As shown below, exciton transitions of QW is polarization dependent on exciton spin states. The spin states can be selected during exciton-SC field interaction. Thus, the SC generated using this type of microstructured fiber opens the new possibility for coherent manipulation of spin states.

For our experimental study, a Ti:sapphire laser (MIRA900, Coherent) with 150 fs pulse width was employed. The central wavelength of the laser pulse was 800 nm and the linewidth was about 5 nm. The output power used was about 300 mW and the repetition rate was 76 MHz. A 100 cm-long irregularly microstructured fiber with the elliptic core was used. The spectrum of the SC light source and the spectral variance using the SC light were measured by a spectrometer system (Acton SP2150i). The polarization direction of the input laser pulse was controlled by a half-wave plate. The spectrum of the SC is shown in Fig. 2. The spectral distribution can be changed by the polarization, the output power and the coupling alignment of the fs pulsed laser to the fiber. We adjusted the spectrum of the SC to cover the expected exciton

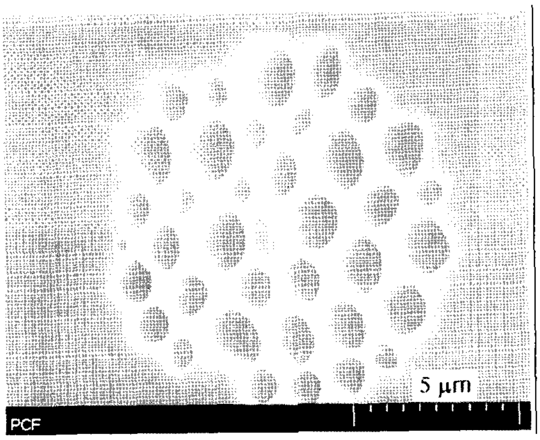

FIG. 1. Scanning electron microscopic image of the cross section of an irregularly microstructured fiber having an elliptic core.

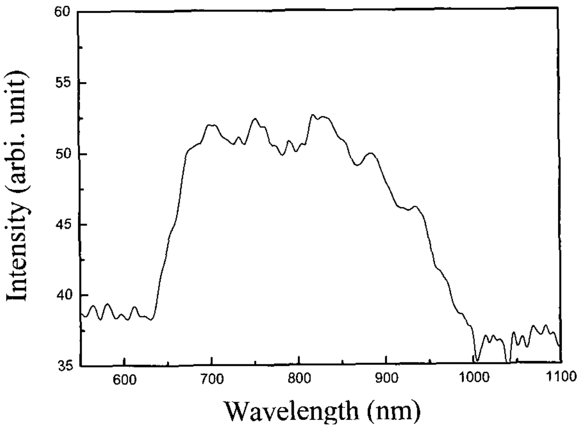

FIG. 2. The spectrum of supercontinuum pumped by a 800 nm wavelength, 300 mW, 150 fs pulsed laser.

levels of our GaAs/AlGaAs QW sample. The GaAs QW sample, grown by molecular beam epitaxy, consists of 20 periods of 10 nm thick GaAs wells and 15 nm thick $Al_{0.35}Ga_{0.65}As$ barriers. The transmission spectroscopic measurements were performed for this QW structure at 11 K up to room temperature by evaluating the transmitted SC intensity normalized by the incident SC intensity. Because we are only interested in the position and the narrowness of the absorption peaks, some transmission loss by the substrate is irrelevant in this case. This transmission loss can be corrected by using a blank substrate without QW if one seeks for the absolute transmittance.

The exciton spin structure with heavy and light holes is shown in Fig. 3. The experimental layout is also shown in Fig. 4. The absorption spectrum of the GaAs QW structure using the SC as the light source was shown in Fig. 5. Two distinct absorption peaks corresponding to the heavy hole and the light hole exciton transitions were clearly observed from 11K up to even room temperature. The peaks of exciton transition lines were both red-shifted together as temperature increased due to the band gap shrinking effect [16]. We compared the absorption spectrum with the PL spectrum measured using 350 nm fs pulsed laser excitation (Fig. 6). The resolving power of distinguishing two closely located, distinct exciton transition lines in the SC absorption spectrum was far superior to that of much broader PL spectrum. The difference is even more apparent at higher temperature. This strongly confirms the possibility of coherent photon control for semiconductor QW structure even in room temperature. We increased the output power of SC to check if there was any power broadening effect in the absorption spectrum, and the power broadening was not observed up to about 30 mW, which is the SC power range we used.

In conclusion, we used SC generated using a microstructured fiber pumped by a fs pulsed laser to observe exciton transition lines in GaAs Quantum well struc-

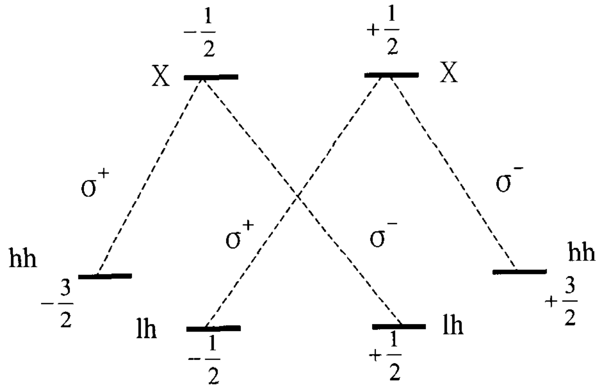

FIG. 3. Exciton spin states, X : exciton, hh : heavy hole, lh : light hole.

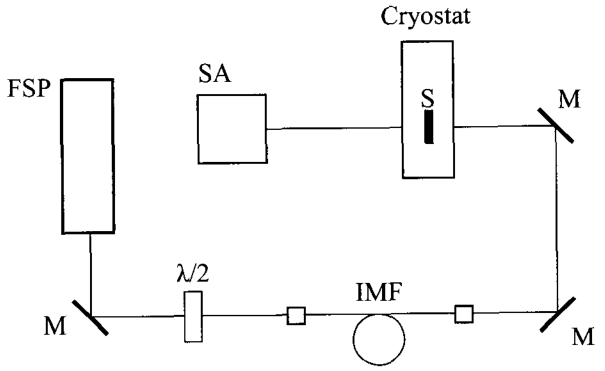

FIG. 4. Experimental configuration for coherent absorption spectroscopy with SC, FSP: 150 fs pulsed Ti:sapphire laser (MIRA900), M: mirror, IMF: irregular microstructured fiber, SA: spectrometer, S: QW sample.

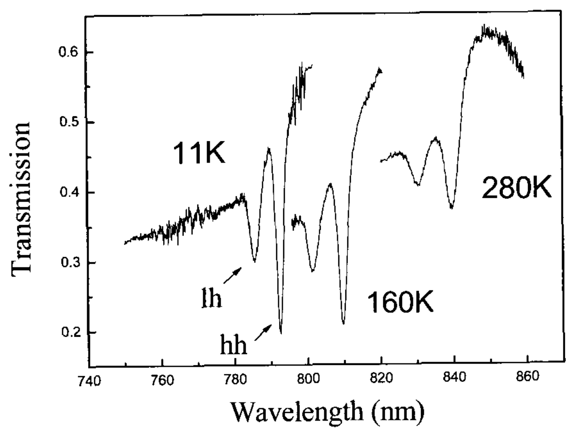

FIG. 5. The SC absorption spectrum of GaAs QW at three different temperature.

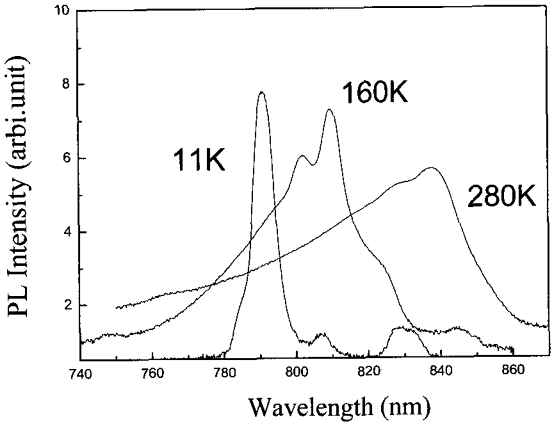

FIG. 6. Photoluminescence spectrum of GaAs QW, excited by a 350 nm wavelength, 150 fs pulsed laser.

ture. The resolving power of closely located, distinct exciton transition lines in the absorption spectrum was much superior to the regular PL spectrum. The coherent absorption spectroscopy using SC makes it possible to observe exciton transitions even at room temperature, which is not so apparent in the PL spectrum. The pumping power of fs pulse laser to generate SC can be minimized to less than 100 mW, and the rest of the output of the fs pulse laser can be used as coupling light for coherent nonlinear optics. Using the SC generated in this work, further experimental investigation in coherent nonlinear optics with coupling and probe lasers is under way.

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