Wavelength modulation spectroscopy of single quantum dots

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We demonstrate that external cavity diode lasers with large mode-hop-free tuning ranges (up to 80 GHz) together with wavelength modulation spectroscopy can be used to study excitonic transitions in semiconductor nanostructures. Such transitions are characterized by homogeneous linewidths typically on the order of a few GHz. Wavelength modulation spectroscopy offers a high signal-to-noise method for the determination of resonance line shapes. We have used this technique to accurately measure dipole moments and dephasing rates of single semiconductor quantum dot eigenstates. These measurements are important for the use of quantum dots in semiconductor cavities and quantum logic gates, and for an improved understanding of the physics of exciton confinement. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1461071]

Absorption spectroscopy based on the modulation of the wavelength of the exciting field is a common technique in atomic and molecular optics.[1–4] Early experiments using diode lasers focused on fast modulation of the injection current, a technique typically referred to as frequency modulation spectroscopy because the modulation frequency was larger than the linewidth of the system under investigation. Such studies benefit from extremely low $1/f$ noise, but are difficult to implement on resonances broader than the modulation frequency.

Wavelength modulation spectroscopy (WMS) utilizes modulation frequencies smaller than the resonance linewidth, and has recently become simpler to implement due to the availability of external cavity diode lasers (ECDLs).[5–7] The large mode-hop-free tuning range available with current ECDLs allows WMS to be used to study resonances whose linewidths are as large as tens of GHz, such as those of interband transitions in high-quality semiconductor heterostructures. Although the modulation frequencies for this technique are limited by the mechanical tuning speeds of the mirror angle in the external cavity, they are sufficient to allow for the use of standard lock-in detection techniques.

An understanding of the optical transition strength of confined excitons in semiconductors is important both to clarify the physics of quantum mechanical confinement in all three dimensions, and to apply this knowledge to the development of nanoscale optoelectronic devices. Ongoing experiments in quantum dot (QD) microcavities in which the excitonic transition is strongly coupled to a resonant optical field of the cavity depend critically on the magnitude of the dipole moment of the confined exciton.[8,9] Also, proposals to implement quantum computing in semiconductors using QDs as qubits in a logic gate rely on π pulses to carry out the gate operations.[10,11] The strength of the optical fields for these π pulses is inversely proportional to the size of the QD dipole moment.

Traditional semiconductor modulation spectroscopy techniques,[12] such as piezomodulation, thermomodulation, optomodulation, or electromodulation, all rely upon a shift in the resonant frequency arising from that property of the semiconductor being modulated. However, a precise physical model of this shift often does not exist, so it must be treated as a fit parameter in the spectral line shape analysis. For energy shifts considerably smaller than the linewidth, fitting procedures cannot accurately distinguish the line shift from the line strength. Thus, systematic errors associated with this technique can be large. WMS, on the other hand, relies upon modulation of the laser frequency itself, allowing for an accurate calibration of the depth of modulation and therefore the transition strength.

WMS data for this letter was obtained using a Littman–Metcalf ECDL (Newport Model 2010) whose output is actively power stabilized using a photodiode to provide feedback to an AOM. The PZT that controls the fine position of the mirror angle (and therefore the wavelength) was driven with a 50 Hz sinusoidal voltage at an amplitude that corresponded to 2.5 GHz. This oscillating voltage was added to a dc voltage that continuously stepped the wavelength over a range of more than 80 GHz. The beam was focused onto the sample and part of it was transmitted through an aperture at the sample surface (discussed below) and excited the QD states. The transmitted beam was then refocused onto an avalanche photodiode (APD) biased below breakdown. The change in transmission detected by the APD at the modulation frequency was monitored using a phase-sensitive lock-in amplifier. The experiments were performed at 6 K to minimize phonon interactions.

The QDs are formed naturally in a narrow (6.2 nm) GaAs layer deposited between 25 nm $Al_{0.3}Ga_{0.7}As$ barriers. 2 min growth interruptions at the interfaces lead to the formation of large monolayer-high islands which give rise to a bimodal distribution of excitonic states, similar to that reported in earlier studies of these structures.[13,14] These two classes of excitonic transitions are indicated in Fig. 1(a) by photoluminescence (PL) (upon optical excitation at ~1.7

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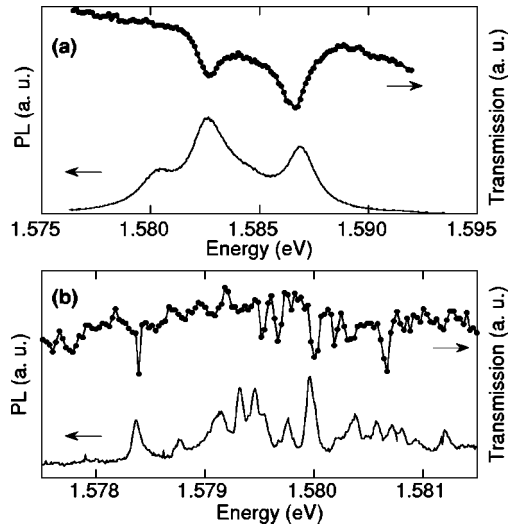

FIG. 1. Photoluminescence (PL) and direct optical absorption from heavy hole QD states (a) in a sample without an apertured aluminum mask on the surface, and (b) probed through an ~0.5 μm aperture on the sample surface. Note the change in energy scale from (a) to (b).

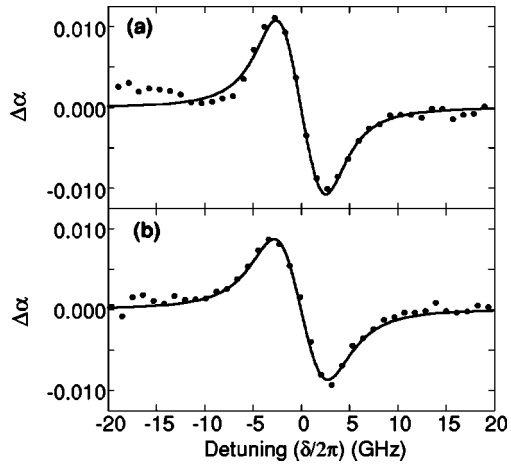

FIG. 2. Wavelength modulation data from two different quantum dot eigenstates. The curve fits have two fit parameters, α_0 and γ. The change in absorption ($\Delta\alpha$) is the rms signal obtained at the modulation frequency divided by the (off-resonance) transmission.

eV) and direct optical transmission, both obtained from an ensemble of excitonic states. The higher (lower) energy inhomogeneously broadened resonance corresponds to heavy hole excitons in the 22 (23) ML thick GaAs region. These width fluctuations give rise to exciton localization within the plane normal to the growth direction in the 23 ML thick regions. The islands are often elongated in the $[1\bar{1}0]$ direction, leading to linear polarization selection rules for the excitonic transitions. The experiments described in this letter were restricted to one of these linear polarization directions.

Excitons in isolated QDs are probed through ~0.5 μm apertures in an aluminum mask laid directly onto the sample's surface. Transmission and PL collected through a typical aperture are shown in Fig. 1(b). The dips in transmission represent absorption from single homogeneously broadened QD states. The bottom curve is the PL collected through the aperture. States that appear in absorption but not in PL are those that decay primarily through nonradiative processes. The strength of the direct absorption signal depends only on parameters specific to the state in resonance (i.e., the dipole moment and the dephasing rate), unlike the strength of PL signal which depends on spectral (and often spatial) coupling rates between states. Spectra similar to that of Fig. 1(b) are used to coarsely position the wavelength of the ECDL at a QD resonance for the data shown below.

Optical absorption from a localized exciton is related to the measured transmission, T, by $T=T_0-T_{\text{abs}}=T_0[1-\alpha(\delta)]$, where T_0 is the off-resonance transmission, T_{abs} is the absorbed power, $\alpha(\delta)$ is the absorption line shape, and $\delta=\omega-\omega_0$ is the laser detuning from the exciton resonance. For a homogeneously broadened transition in a single two-level system;

$$\alpha(\delta)=\frac{\alpha_0}{1+(\delta/\gamma)^2}, \tag{1}$$

where $\alpha_0=\omega\mu^2/(An\hbar c\epsilon_0\gamma)$, μ is the optical dipole moment, A is the area of the optical field that excites the QD, γ/π is the (FWHM) linewidth (in Hz), and n is the index of refraction of the semiconductor (3.66 for GaAs). A is given by the size of the aperture, about 400 nm in diameter for the data discussed below.

Wavelength modulation may be represented by $\delta(t)=\delta+D\cos(\Omega t)$, where D is the depth of modulation ($D/2\pi=2.5$ GHz) and Ω is the modulation frequency ($\Omega/2\pi=50$ Hz). $\Delta\alpha(\delta)$ is the rms transmission signal observed at the modulation frequency divided by the off-resonance transmission (T_0) and is in-phase with the frequency modulation. For small modulation ($D\ll\gamma$), $\Delta\alpha(\delta)$ is simply a derivative line shape, but for large modulation ($D\sim\gamma$), it is expressed for a Lorentzian as:[15]

$$\Delta\alpha(\delta)=\frac{\alpha_0[\text{sign}(\delta)\sqrt{x-y}-(\delta/y)\sqrt{x+y}]}{(D/\gamma)x}$$

$$x=\sqrt{y^2+4(\delta/\gamma)^2}$$

$$y=(D/\gamma)^2-(\delta/\gamma)^2+1, \tag{2}$$

where $\text{sign}(\delta)=\{-1$ if $\delta<0$; 0 if $\delta=0$; 1 if $\delta>0\}$.

Equation (2) is fit to the WMS data shown in Figs. 2(a) and 2(b), representing two different QD states obtained from an aperture similar to the one discussed in Fig. 1(b). The two free parameters, α_0 and γ, are used to find μ using Eq. (1). The state in Fig. 2(a) is found to have $\alpha_0=0.038^{+0.007}_{-0.014}$, $\mu=57^{+6}_{-10}$ Debye, and $\gamma/\pi=6.1\pm0.4$ GHz. The state in Fig. 2(b) is found to have $\alpha_0=0.034^{+0.007}_{-0.012}$, $\mu=58^{+6}_{-10}$ Debye, and $\gamma/\pi=7.2\pm0.3$ GHz. Using $f=2m_0\omega\mu^2/(e^2\hbar)$ (where m_0 is the free electron mass), the oscillator strengths are found to be (a) $f=58^{+12}_{-21}$ and (b) $f=60^{+12}_{-22}$. The notation X^{+du}_{-dl} denotes lower (dl) and upper (du) error bars on X. Data from other states (not shown) find dipole moments typically to be in the range 50–100 D and the FWHM linewidths to be in the range 6–12 GHz. The signal-to-noise ratio for these data is about 20, though this can be improved by averaging over multiple data sets. A signal-to-noise ratio of one therefore corresponds to a minimum measurable value of μ of about 12 D using this method.

For the sample employed in these studies, the refractive index mismatch between the semiconductor and vacuum contributes a standing-wave component (in addition to the

traveling wave component) to the electric field experienced by the quantum dot. Matching the boundary conditions throughout the entire sample, the actual absorption by the QD is found to be no more than ~30% smaller than that inferred by the measured change in transmission. Diffraction by the subwavelength aperture, which has not been considered, would make this effect (and the percent error) even smaller. A knowledge of the area illuminated by the optical field at the QD (A) is necessary for a determination of the dipole moment from Eq. (1). The aperture diameters are determined postprocessing to about 20% accuracy using optical transmission calibrated to known ($\sim\mu$m sized) apertures. These two factors are the dominant source of uncertainty in the reported dipole moments, and determine the error bars given above.

The linewidths obtained by WMS are similar to those previously reported using nonlinear pump–probe spectroscopy[16] and PL excitation spectroscopy[17] in similar apertured QD systems. In addition, both linewidths and dipole moments agree with those obtained from direct optical absorption measurements performed without modulation on the same QD states.[18] Calculations of transition dipole moments in GaAs/AlGaAs QDs with a height of 3 nm and a diameter of about 30 nm find the dipole moment to be 81 D.[19] Andreani *et al.* calculate a dipole moment of 67 D for a 4 nm quantum well width and 40 nm quantum dot diameter.[9] These calculated values are similar to those reported in this letter, but larger than those previously reported using optomodulation spectroscopy.[20] However, the optomodulation technique is now believed to systematically underestimate the transition strength. This issue, as well as an in-depth comparison between the theoretical and experimental line strengths and linewidths of QDs in thin growth-interrupted GaAs layers is discussed elsewhere.[18]

The large dipole moment associated with these QD excitonic transitions shows that optical absorption from GaAs interface fluctuation QDs is very strong, making them attractive candidates for nanoscale optoelectronic devices. This enhanced interaction with light also suggests applications in QD-cavity strong coupling experiments, which require a large transition moment to achieve the required coupling strength.[8,9] Furthermore quantum logic gates that require π pulses for the gate operation[10,11] benefit from the weak field strengths that result from a large dipole moment. The recent demonstration of Rabi oscillations of excitons in single interface fluctuation QDs is an important step towards the realization of a solid state quantum logic gate, and confirms the large transition moments associated with these systems.[21]

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