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Optical absorption cross section of quantum dots

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Abstract

We have measured the modal optical absorption spectrum of a three-layer system of InAs quantum dots in a slab waveguide geometry, observing distinct absorption peaks for the ground and excited states. The spectrally integrated absorption cross section for the ground and first excited states are determined to be $\sigma_0 = (0.43 \pm 0.1) \times 10^{-15}$ and $(0.92 \pm 0.2) \times 10^{-15}$ cm^2 eV, respectively. Assuming that the spectral shapes are determined primarily by the inhomogeneous size distribution of dots the Gaussian linewidths are 16 and 19 meV for the ground and first excited state transitions, respectively. The peak ground state absorption cross section is 1.1×10^{-14} cm^2. The ground state spectrally integrated cross section estimated by a theory with the envelope function overlap integral taken to be unity is 0.40×10^{-15} cm^2 eV, in agreement with the measured value. We conclude that on the basis of the spectrally integrated cross section there is no evidence for a substantial reduction in the strength of the fundamental light–matter interaction in dots compared with systems of higher dimensionality.

1. Introduction

Semiconductor quantum dots are of considerable interest for use in optoelectronic devices, and the properties of quantum dot laser diodes and optical amplifiers are the subject of extensive investigation [1]. The dots are usually formed by self-assembly by the Stranski–Krastinow process during epitaxial growth and are present in the structure as one or more layers of dots within a waveguide structure [2]. The operation of dots in these devices is controlled at a fundamental level by the strength of the interaction of electromagnetic radiation with the electronic energy level system of the dots and is expressed as the optical cross section, $\sigma(h\nu)$.

For example, for light incident normally to a layer of dots the fraction of light absorbed at low intensity by a layer of dots is given by

$$\left[\frac{\Delta\Phi}{\Phi_0}\right]_{h\nu} = -N_{\text{dots}}\sigma(h\nu), \tag{1}$$

where Φ is the photon flux and N_{dots} is the total number of dots per unit area. A similar cross section determines the maximum optical gain, which can be obtained from a system of dots if the occupation of the states participating in the transition are fully inverted (i.e. all the upper states are filled, all the lower states are empty). The size, shape and composition of the dot and its surrounding matrix determine the energy states of the dot and the associated wavefunctions, and the wavefunctions together with the momentum matrix element determine the optical cross section. The optical cross section is therefore of fundamental importance in making comparisons between experiment and theory, in making comparisons between different dot systems, and in modelling optoelectronic devices. In making these comparisons it is important to remember that most self-assembled quantum dots are not spherical so the orientation of the light beam and its polarization relative to the plane of the dot layer influences the precise value of the cross section.

There have been reports of determinations of the *gain* cross section, σ_{gain}, for example from analysis of threshold current and use of a gain model [3], though in principle the gain cross section can also be obtained from direct measurements of gain by use of an equation similar to equation (1). In laser experiments the light generally propagates in a slab waveguide along the layer of dots and it is necessary to take account of the optical coupling of the guided mode to the dots. Values for the gain cross section have been obtained from the value of the maximum gain which can be obtained from the transition [3] and it is assumed that in such circumstances the carrier population of the system is fully inverted. The *absorption* cross section is usually measured under low injection conditions where the lower level is full and the upper level empty, so the two experiments may be expected to give a similar result for the same geometry, the chief difference being the presence of carrier–carrier interactions in the high injection gain experiment. Recent experimental work has shown that although the optical gain from a quantum dot system saturates at sufficiently high injection, the system may not be fully inverted [4]. This situation may arise from rate-limiting capture processes at high injection and a high density of extended states which controls the quasi-Fermi level positions at high carrier density. In these circumstances the value obtained for σ_{gain} by assuming full inversion will be less than the true cross section, characteristic of the fundamental light–matter interaction. Additionally the gain may be reduced by the presence of 'dark dots' where emission is quenched by non-radiative recombination.

The more reliable approach to determination of the cross section is to measure the passive optical absorption with a low intensity light beam, but this is difficult in normal incidence geometry because the fraction of light absorbed is small: for $\sigma = 10^{-14}$ cm^2 and $N_{\text{dot}} = 10^{10}$ cm^{-2} the fraction absorbed is 10^{-4}. While this can be overcome to some extent by increasing the number of dot layers, an alternative approach is to measure the passive modal absorption in a waveguide geometry. Although the coupling of the light to the dots is reduced by the optical confinement factor, it is possible to use long path lengths to obtain a measurable effect. The experiment has the merit that the geometry is the same as that for optical gain so direct comparisons can be made with the gain cross section.

In this paper we report the results of measurements of modal absorption of a three-layer system of InAs dots in a waveguide geometry. In these samples the modal gain saturates at a value which is only about one third of the magnitude of the modal absorption. The measured absorption spectrum can be well described by a simple theory for an inhomogeneous size

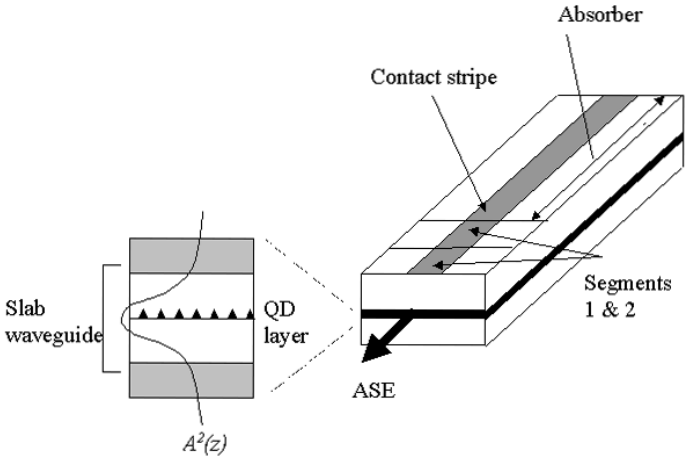

Figure 1. Diagram of the slab waveguide geometry used for the segmented contact measurements of modal absorption.

distribution of dots, using best estimates of the dot density. In the next section we describe the segmented contact technique used to measure the modal absorption, together with the experimental results. Derivation of the cross section, comparisons with theory and implications for the modal gain are covered in the remaining sections.

2. Absorption measurements

The modal absorption was measured in a waveguide geometry using a laser structure with an electrical stripe contact divided into a series of segments which could be driven independently, illustrated in figure 1. The sample was grown by MBE and comprised three layers of InAs dots, each embedded in a $In_{0.15}Ga_{0.85}As$ well of thickness 9.6 nm and surrounded by undoped GaAs to provide a waveguide core of total thickness 230 nm [5]. The cladding layers were $Al_{0.7}Ga_{0.3}As$. The dots have a base diameter of about 15 nm and height of about 7 nm and the density for one layer of dots was $(2.5 \pm 0.5) \times 10^{10}$ cm^{-2} obtained by AFM measurements on a calibration wafer (and quoted in [5]).

The material was processed into 100 μm wide oxide-isolated structures with the contact isolated into segments each $L = 293$ μm long in a device of overall length 3 mm. These devices were mounted onto copper heatsinks. The contact segments were driven separately with pulses of 1 μs duration at a duty cycle of 0.1%. The amplified spontaneous emission was collected from the end of the structure and detected using a grating spectrometer and a Hamamatsu cooled photomultiplier sensitive to a wavelength of 1.4 μm. The spectral response of the overall measurement system was determined using a standard lamp.

Details of the derivation of the modal absorption and gain spectra from analysis of edge-emitted spontaneous emission spectra are given in [6]. Round-trip amplification in the structure was inhibited by the 3 mm long passive absorbing section beyond the pumped regions (see figure 1) so the observed emission makes only a single pass through the structure. The passive absorption of the dot system was measured as the ratio of emission when the first ($I_{\mathrm{meas}}(1)$) and second ($I_{\mathrm{meas}}(2)$) section segments were driven separately, the light from the second segment being passively absorbed by the first segment before being detected:

$$(A_{\mathrm{mode}} + \alpha_i) = \frac{1}{L} \ln \left\{ \frac{I_{\mathrm{meas}}\,(1)}{I_{\mathrm{meas}}\,(2)} \right\}, \tag{2}$$

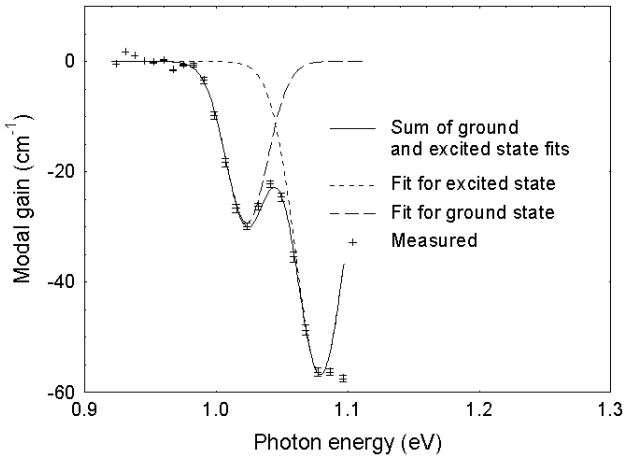

Figure 2. Modal absorption spectra, with the experimental results shown as data points and the curves show the fit to individual ground and excited state transitions using equation (6) and the sum of these contributions.

where A_{mode} is the *modal* absorption coefficient of the dot layers and α_i represents the optical loss from the waveguide due to scattering processes. The gain was measured by driving section one, then section one and two together and so on, to replicate a stripe-length determination of modal gain [7]. All measurements were done at room temperature and for light polarized in the plane of the layers of the structure (TE).

Figure 2 shows the measured net modal absorption spectrum for light propagating along the layer of dots, measured with a drive current through the exciting segment of 100 mA. At photon energies below the absorption edge of the dots absorption is due to losses in the waveguide of about 5 ± 1 cm^{-1}. The absorption peaks at 1.02 and 1.08 eV are due to the ground and excited states of the dot system and the modal absorption at each peak (after removing the waveguide loss) is 30 ± 1 and 56 ± 1 cm^{-1}, respectively.

Figure 3 is the gain spectrum measured at a drive current per segment of 200 mA. The gain from the ground state is observed to saturate with current above about 130 mA so this spectrum shows the saturated ground state gain. When the waveguide loss is taken into account, the maximum peak modal gain due to the dots is 10 cm^{-1} which is only one third of the peak modal absorption.

3. Derivation of the absorption cross section

The spectral dependence of the optical absorption cross section of a *single* dot is a Lorentzian line given by

$$\sigma(\hbar\omega) = \left\{ \frac{2 \times 4\pi\hbar}{n\varepsilon_0 c\hbar\omega} \left(\frac{e}{2m}\right)^2 \gamma(M)^2 \int F_2(\mathbf{r}) F_1(\mathbf{r})\, \mathrm{d}^3\mathbf{r} \right\} L(E_i, \hbar\omega). \tag{3}$$

This equation is for a simple two-level system with two electrons of opposite spin in each dot state with the lower state of the transition being full and the upper state empty. M is the momentum matrix element, γ is a factor which takes account of the relative orientation of the optical field and the dot geometry, and F_1 and F_2 are the three-dimensional envelope functions for the upper and lower dot states. $L(E_i, \hbar\omega)$ is a normalized homogeneous Lorentzian lineshape function centred on the transition energy of the ith dot E_i. If the homogeneous linewidth is small compared with the photon energy and terms within { } are independent of

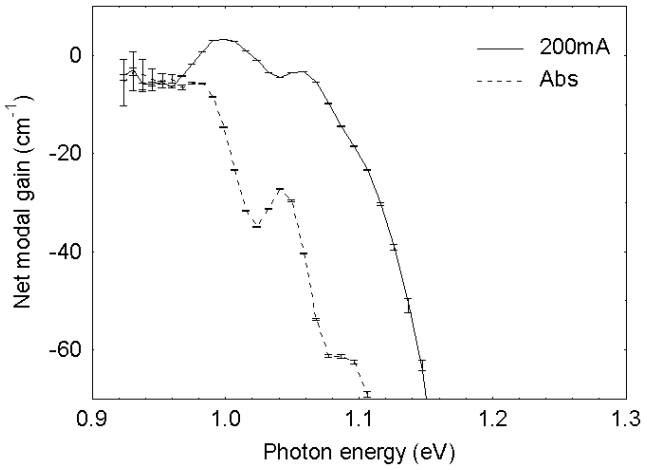

Figure 3. Modal gain spectrum measured for a drive current per segment of 200 mA.

energy over the line, then when the cross section is integrated over the linewidth we obtain the total absorption strength per dot σ_0, in units of [cross section area] $\times$ [energy], of

$$\sigma_0(E_i) = \left\{ \frac{2 \times 4\pi\hbar}{n\varepsilon_0 c\hbar\omega} \left(\frac{e}{2m}\right)^2 \gamma(M)^2 \int F_2(\mathbf{r}) F_1(\mathbf{r})\, \mathrm{d}^3\mathbf{r} \right\}. \tag{4}$$

The modal absorption of light in a slab waveguide containing a layer of N_{dots} per unit area in the plane of the slab is given by:

$$A_m(\hbar\omega) = \frac{N_{\mathrm{dots}}}{w_{\mathrm{mode}}} \left\{ \frac{2 \times 4\pi\hbar}{n\varepsilon_0 c\hbar\omega} \left(\frac{e}{2m}\right)^2 \gamma(M)^2 \int F_2(\mathbf{r}) F_1(\mathbf{r})\, \mathrm{d}^3\mathbf{r} \right\} \times \int L(E_i, \hbar\omega) P(E_i)\, \mathrm{d}E_i, \tag{5}$$

where the guided mode has an effective width [8] w_{mode} and we have assumed that the electric field of the mode is uniform over the layer (or layers) of dots, as discussed in [8]. The total absorption at a specific photon energy ($\hbar\omega$) is obtained by integrating over dots of different transition energy E_i, in the Gaussian inhomogeneous distribution $P(E_i)$, which have homogeneous Lorentzian distributions which embrace the photon energy of interest. We have assumed that the factors in $\{\,\}$ have only a small variation with photon energy over the inhomogeneous distribution. The chief variable is the photon energy itself so we are assuming that the inhomogeneous linewidth is small compared with the photon energy. If the inhomogeneous linewidth is greater than the homogeneous linewidth we can write equation (5) as

$$A_m(\hbar\omega = E_i) = \frac{N_{\mathrm{dots}}}{w_{\mathrm{mode}}} \sigma_0(E_i) P(E_i). \tag{6}$$

The value of $\sigma_0(E_i)$ can be obtained from experimental data most directly by simply integrating the measured modal absorption spectrum, an approach which has no implied assumptions regarding the inhomogeneous lineshape:

$$\sigma_0 = \int_{-\infty}^{\infty} \sigma_0 P(E_i)\, \mathrm{d}\,(E_i = \hbar\omega) = \frac{w_{\mathrm{mode}}}{N_{\mathrm{dots}}} \int_{-\infty}^{\infty} A(\hbar\omega)\, \mathrm{d}\hbar\,\omega. \tag{7}$$

Alternatively, equation (6) shows that the form of the absorption spectrum should follow the inhomogeneous distribution and the value of $\sigma_0(E_i)$ can be obtained by fitting the spectrum

with an appropriate normalized lineshape function. This approach can be used where the spectra for ground and excited state transitions partially overlap and is preferable to the use of spontaneous emission spectra as a means of determining the linewidth. The expression for $\sigma_0(E_i)$ given by equation (4) is consistent with both of these methods of data analysis.

The measured spectrum in figure 2 has been fitted with equation (6) using a Gaussian inhomogeneous distribution, and with the cross section $\sigma_0(E_i)$ and the Gaussian halfwidth (σ_E) as the fitting parameters. We used $N_{dots} = 3 \times (2.5 \times 10^{10})$ cm^{-2} and $w_{mode} = 0.28$ μm, the latter obtained from a mode solver for the nominal waveguide structure. $\sigma_0(E_i)$ is constant over the inhomogeneous line of each transition, but $\sigma_0(E_i)$ and σ_E are allowed to be different for the ground state and each excited state transition. The fit is shown in figure 2 and the fitting parameters were $\sigma_0(E_i) = (0.43 \pm 0.1) \times 10^{-15}$ and $(0.92 \pm 0.2) \times 10^{-15}$ cm^2 eV for the ground and first excited state transitions, respectively, and $\sigma_E = 16$ and 19 meV for the two transitions, respectively. The peak ground state cross section is 1.1×10^{-14} cm^2.

4. Comparison with theory and implications for modal gain

It is enlightening to compare the experimental value of the spectrally integrated cross section with the value given by equation (4). The simplest situation is where the overlap integral of the envelope functions $F(\mathbf{r})$ is unity, however a further question arises regarding the value to be used for the polarization factor γ. In a quantum well this has a value of (1/3) at $k = 0$ for TE polarized light, that is light with the electric field vector in the plane of the well. Our experiments have a similar geometry with the light polarized in the plane of the dots which also corresponds to the plane of the largest dimension of the dots. However the 3D localization in the dots means the electron states are described by a combination of states with a range of k vectors which reduce γ below its value at $k = 0$ [9]. For simplicity and definiteness we take $\gamma = (1/3)$, recognizing that the value derived is likely to be an overestimate. With a value of the matrix element M for InAs given by $(2M^2/m_0) = 21.1$ eV and $\hbar\omega = 1.12$ eV, corresponding to the ground state peak in figure 2, we obtain $\sigma_0(E_i) = 0.40 \times 10^{-15}$ cm^2 eV from equation (4).

This simple application of equation (4) agrees with the experimental measurements within the estimated uncertainty. We would expect our calculated value to overestimate the true value due to our neglect of the effect of k values greater than zero and the assumption of complete envelope function overlap. The greatest uncertainly in the experimental data is the dot density ($\pm$20%) and uncertainties also arise in the effective mode width due to uncertainty in layer thicknesses and composition of the waveguide.

We conclude that the measured overall absorption strength is in reasonable agreement with theoretical predictions and we find no evidence for *large* differences which could arise from built-in piezoelectric fields in the dots [10] (though modest effects due to incomplete overlap may occur). Nevertheless it is certainly the case that the modal gain of a system of dots (5–10 cm^{-1}) is an order of magnitude smaller than that of a quantum well (50–100 cm^{-1}), but this is not due to fundamental differences in the light–matter interaction as represented by equation (5), but rather to the effect of inhomogeneous broadening reducing the peak gain. We can illustrate this by calculations of the peak modal absorption: this gives the maximum peak modal gain which can be obtained if the system can be fully inverted in the absence of any dark dots.

In the absence of inhomogeneous broadening (i.e. for a system of identical dots), the *peak* modal absorption can be obtained from equation (5) as:

$$A_{pk} = \frac{N_{dot}}{w_{mode}} \sigma_0 L\,(E_i, \hbar\omega = E_i) = \frac{N_{dot}}{w_{mode}} \sigma_{pk} = \frac{N_{dot}}{w_{mode}} \left(\frac{\sigma_0}{\pi \Lambda}\right), \tag{8}$$

Table 1. Peak cross section and modal absorption for homogeneous and inhomogeneously broadened dot distributions. The number in [ ] is the effective coverage for a single layer of dots, i.e. ($N_{\rm dot} \times \sigma_{\rm pk}$) for $N_{\rm dots} = 2.5 \times 10^{10}$ cm^{-2}. The peak modal gain is calculated for three layers of dots as in the sample used for the experiments.

	Experimental absorption strength $\sigma_0 = 0.44 \times 10^{-15}$ cm^2 eV	
Broadening	Peak cross section (cm^2)	Peak modal absorption, (cm^{-1})
Homogeneous $\Lambda = 200\ \mu$eV	0.7×10^{-12} [1.7×10^{-2}]	1850
Inhomogeneous $\sigma_{\rm E} = 16$ meV	1.11×10^{-14} [2.8×10^{-4}]	30

where Λ is the homogeneous Lorentzian linewidth. If we use the experimental value for the spectrally integrated absorption strength ($\sigma_0 = 0.44 \times 10^{-15}$ cm^2 eV), and take $\Lambda = 200\ \mu$eV which corresponds to a dephasing time of about 3 ps, we obtain a peak cross section of $\sigma_{\rm pk} = 0.7 \times 10^{-12}$ cm^2 and the peak modal absorption for a three-layer dot system in our waveguide ($N_{\rm dot} = 7.5 \times 10^{10}$ cm^{-2}) without size dispersion would be 1850 cm^{-1}. At the absorption peak the ($N_{\rm dot} \times \sigma_{\rm pk}$) product for a single layer of dots (2.5×10^{10} cm^{-2}) is 1.75×10^{-2}, which is the effective area coverage of the dots and represents the effect of the reduced volume due to fragmentation of the gain medium into a system of dots.

For an inhomogeneous distribution the peak modal absorption is given by

$$A_{\rm pk} = \frac{N_{\rm dot}}{w_{\rm mode}}\sigma_{\rm pk} = \frac{N_{\rm dot}}{w_{\rm mode}}\left(\frac{\sigma_0}{\sqrt{2\pi}\sigma_{\rm E}}\right), \tag{9}$$

so for a halfwidth $\sigma_{\rm E} = 16$ meV (as determined for our sample) $\sigma_{\rm pk} = 1.1 \times 10^{-14}$ cm^2 and the peak modal absorption is 30 cm^{-1}.

These values for homogeneous and inhomogeneous distributions are summarized in table 1, and they illustrate the crucial role of inhomogeneous broadening in determining the absorption coefficient measured at the peak of the absorption line. The modal absorption represents the likely value of the maximum peak modal gain which can be obtained if the ground state can be fully inverted.

5. Discussion and conclusions

We can draw a number of conclusions from the experimental results and data presented in table 1. The measured value of *spectrally integrated* absorption cross section of the ground state transition, which represents its total absorption strength, is similar to the value predicted by simple theory assuming complete overlap of the envelope functions and using the matrix element at $k = 0$. There is no evidence for major discrepancies between experiment and theory for the strength of the basic light–matter interaction in a quantum dot.

The low *peak* optical cross section and *peak* modal absorption (indicative of the maximum available peak modal gain) arise from the reduced effective area coverage compared with a well, and the effect of inhomogeneous broadening spectrally dispersing the optical strength. A three layer system of dots of uniform size (2.5×10^{10} cm^{-2} per layer) should have a modal gain in our waveguide geometry of 1850 cm^{-1}, whereas in the presence of the measured inhomogeneous broadening of 16 meV this is reduced by a factor 60 to only 30 cm^{-1}. These simple calculations reveal the sensitivity of values of *peak* absorption and gain to broadening

and highlight the perils of making comparisons between experiment and theory, and data for different dot systems, in terms of cross section and gain, both of which always refer to a value at a specific photon energy and therefore incorporate differences due to the inhomogeneous broadening of each specific sample. We re-iterate that these comparisons have been made in terms of modal absorption and are not affected by incomplete inversion of the dot states nor by the presence of dark dots. These effects will further reduce the peak modal gain.

It follows from this discussion and equation (4) that for dots in a given material system (in this case InAs) and emitting at the same photon energy, differences in the value of spectrally integrated cross section arise primarily from the overlap integral and the degree of localization. The physical size of the dot affects the absorption only indirectly through these factors. We therefore expect InAs dot systems operating at the same wavelength to have similar values of σ_0.

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