Second-harmonic generation resonant with *s-p* transition in InAs/GaAs self-assembled quantum dots

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(Received 22 September 2000; published 1 March 2001)

We have investigated the second-harmonic generation with the conduction states of InAs/GaAs self-assembled quantum dots. The harmonic generation is resonant with intersublevel transitions, and in particular with the *s-p* transition. This transition, polarized along the [110] and the [−110] directions, exhibits a large dipole matrix element (3 nm). The frequency doubling is achieved around 20 μm wavelength using the *s-p* and the *p-d* transitions. The double resonance leads to a resonant enhancement of the susceptibility with a linewidth lower than 1 meV. We show that the polarization dependence of the susceptibility follows the polarization selection rule of the intersublevel transitions. The susceptibility amplitude is deduced by comparison with phase-matched second-harmonic generation in bulk GaAs. A susceptibility as large as 2.5×10^{-6} m/V for one quantum dot plane is measured. This susceptibility is four orders of magnitude larger than the susceptibility of bulk GaAs.

DOI: 10.1103/PhysRevB.63.113312 PACS number(s): 78.66.Fd, 42.65.Ky, 78.30.−j

I. INTRODUCTION

Optical nonlinearities associated with intersubband transitions in semiconductor quantum wells have been successfully reported during the last decade.[1,2] More recently, second-and third-order nonlinear processes associated with intersublevel transitions in the valence band of semiconductor quantum dots were investigated.[3,4] In all cases, an enhancement of the nonlinear susceptibility for second- and third-harmonic generation as compared to the susceptibility of bulk materials was reported. This enhancement is a consequence of the resonance conditions between the pump exciting field and the intersublevel (or intersubband) transitions. In a resonant system, the nonlinear susceptibility of nth order is proportional to the product of $n+1$ dipole matrix elements. The enhancement of the resonant nonlinear susceptibility first originates from the maximization of the product of the dipole matrix elements. Intersublevel transitions with the largest dipole matrix elements should thus lead to large nonlinear coefficients. The achievement of the resonance condition is a second prerequisite for a large nonlinear susceptibility. In the case of second-harmonic generation in quantum dots, this resonance condition corresponds to the resonance of the pump and the harmonic field with intersublevel transitions exhibiting nonvanishing dipole matrix elements.

The amplitude and the direction of the dipole matrix elements of intersublevel transitions in quantum dots depend on the geometry of the dots. In the conduction band of InAs/GaAs self-assembled quantum dots with a flat lens-shaped geometry, few electron states are confined in the dots. The largest dipole matrix elements for intersublevel transitions involving the ground state is associated with the transition with the fourfold degenerate first excited state. This transition is usually referred to as the ''*s-p*'' transition in the literature. For quantum dots with a typical height of 2.5 nm and a diameter of 25 nm, the dipole of the *s-p* transition is around 3 nm. This transition is in-plane polarized. In a previous article, we have shown theoretically that double resonant second-harmonic generation involving this transition could be achieved.[4] The double resonant process is based on the e_{000}-$e_{100}(s\text{-}p)$ intersublevel transition, the e_{100}-e_{110} and the e_{000}-e_{110} intersublevel transitions. In this notation, the states are classified by reference to the parallelepipedal quantum dot with infinite barriers, i.e., according to the number of nodes of the eigenfunction along the *x*, *y*, and *z* directions that correspond to the [110], [−110], and [001] directions, respectively. There is no dipole matrix element between the ground-state e_{000} and $e_{200}(e_{020})$. Since the $e_{100}-e_{110}$ and the $e_{000}-e_{110}$ intersublevel transitions are respectively polarized in the layer plane and along the growth axis of the dots, the susceptibility for second-harmonic generation is expected to be of the type $\chi^{(2)}_{zxx}$. We emphasize that the specific polarization selection rules of the intersublevel transitions generate susceptibility components different from those associated with intersubband transitions in quantum wells (only $\chi^{(2)}_{zzz}$ is allowed in the conduction band of the quantum wells). The existence of these components is a direct consequence of the lack of inversion symmetry of the lens-shaped quantum dots.

Here, we report the first observation of second-harmonic generation with intersublevel transitions with the conduction states of self-assembled quantum dots. The nonlinear susceptibility is enhanced because of the resonance with the *s-p* intersublevel transition. We show that the spectral dependence of the susceptibility exhibits a very narrow resonance.

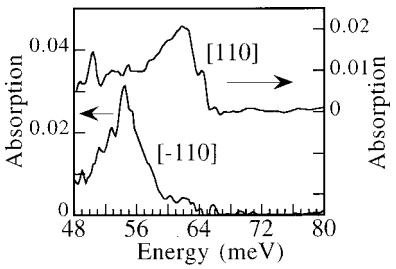

FIG. 1. Room-temperature absorption of the *n*-doped quantum dot sample. The absorption is measured at normal incidence. The transmission is normalized by the transmission of an undoped GaAs substrate with an identical thickness. The absorption is polarized along the [100] and [−110] directions.

This narrow resonance is a signature of the double resonance between the intersublevel transitions. This feature indicates that the confined states in quantum dots with a lens-shaped geometry can be equally spaced in energy.

II. EXPERIMENTAL SETUP

The InAs/GaAs self-assembled quantum dots were grown by molecular-beam epitaxy. The sample consists of 30 quantum dot layers separated by 50-nm thick GaAs barriers.[5] The dot density is $4\times10^{10}\,\mathrm{cm}^{-2}$. The quantum dots are *n* doped with Si with a nominal delta doping of $8\times10^{10}\,\mathrm{cm}^{-2}$ lying 2 nm below the quantum dot planes. This doping corresponds to an average density of two carriers per dot. For second-harmonic generation experiments, the sample was polished with 45° wedge. The length of the sample (1.8 mm) allows two passes of the infrared excitation within the quantum dot layers. A reference sample with undoped quantum dots was also used in order to measure the contribution of bulk GaAs to the second-harmonic generation. The experiments were performed at low temperature (10 K). The cryostat was equipped with a KRS-5 optical window for light injection around 20 μm wavelength. The excitation source was provided by the picosecond pulses delivered by a free-electron laser.[3] A long-wave pass filter was inserted in the laser beam in order to reject the harmonics of the free-electron laser. The light collected after the sample was analyzed with a spectrometer and detected with a broadband mercury-cadmium-telluride photodetector. The spectrometer did allow a good rejection of the pump. In the same time, the second-harmonic nature of the signal was unambiguously determined.

III. RESULTS

Figure 1 shows the infrared absorption of the quantum dot sample at room temperature. The absorption was measured at normal incidence. The transmission for a given polarization is normalized by the transmission of a GaAs reference sample with the same thickness. The intersublevel absorption between the ground state and the first excited state exhibits two resonances at 54 and 62 meV. The absorption at 54 meV is polarized along the [−110] direction, while the absorption at 62 meV is polarized along the [110] direction. The lines

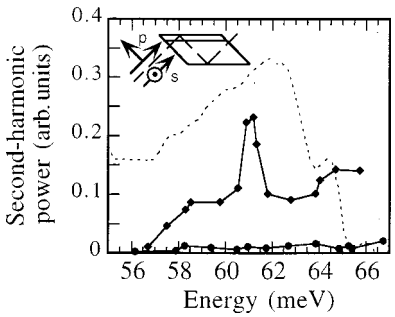

FIG. 2. Second-harmonic power as a function of the pump energy. Diamonds: *n*-doped quantum dot sample; dots: undoped GaAs reference sample. The dashed line shows the absorption along the [110] direction. The inset shows the waveguide geometry. The pump beam is *p* polarized. The lower amplitude for the GaAs reference sample is attributed to a difference in the injection and collection efficiencies.

are inhomogeneously broadened (full width at half maximum ~6–8 meV) by the dot size distribution. The splitting between the two lines is most likely attributed to an elongation of the quantum dots along the [−110] direction.[6] The 8 meV splitting would correspond to a 10% elongation of the dots along one direction, which means that the quantum dots do not exhibit a cylindrical symmetry. The number of carriers transferred in the dots at room temperature can be deduced from the amplitude of the absorption. Starting from the theoretical value (3 nm) for the dipole matrix element of the *s*-*p* transition, one finds a carrier density transferred in the dots of $2\times10^{10}\,\mathrm{cm}^{-2}$. We emphasize the importance of the polarization dependence of the absorption. For second-harmonic generation experiments, the sample was polished with 45° facets in order to couple in *p* polarization the infrared beam with the in-plane polarized transition at high energy (62 meV).

Figure 2 shows the spectral dependence of the second-harmonic power of the *n*-doped quantum dot sample. The second-harmonic power is compared to the power collected with the undoped reference sample. In the latter case, only bulk GaAs contributes to the second-harmonic generation. The dashed line shows the spectral dependence of the infrared absorption along the [110] direction. The inset shows the waveguide geometry. The incident light was *p* polarized (50% of the electric field in the layer plane along the [110] direction and 50% of the electric field along the *z*-growth axis). The excitation beam was focused with a Ge lens with a spot size of ~300 μm. The incident intensity was around 100 MW cm^{-2}. A narrow resonance for the second-harmonic power is observed at 61 meV for the *n*-doped quantum dot sample. This resonance is superimposed on a background signal corresponding to the contribution of bulk GaAs. The 61 meV resonance is a signature of the resonant enhancement of the second-harmonic generation associated with the intersublevel transitions. The measured linewidth of the second-harmonic power (0.6 meV) is limited by the linewidth of the midinfrared picosecond pulses. This broadening is, to our knowledge, the narrowest linewidth reported for second-harmonic generation with intersubband or intersublevel transitions. This narrow linewidth indicates the

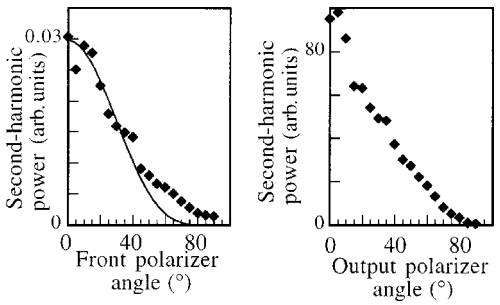

FIG. 3. Second-harmonic power as a function of the polarization angle of the incident excitation beam. 0° (90°) corresponds to a $p(s)$ polarized excitation. In p polarization, part of the electric field is along the [110] direction. In s polarization, the electric field is along the $[-110]$ direction. The pump energy is 61 meV. The figure on the right hand side shows the second-harmonic power as a function of the *output* polarizer angle. The pump at 61 meV was set in p polarization.

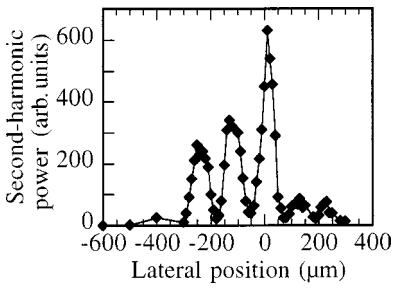

FIG. 4. Second-harmonic power of bulk GaAs as a function of the lateral position of the sample. The maxima correspond to the achievement of phase matching. The distance between two maxima corresponds to twice the coherence length.

achievement of the double resonance condition for a fraction of the dots. With a single resonance, the linewidth would be reduced by a factor of $\sqrt{2}$ as compared to the linewidth of the absorption. This resonance has to be compared with the theoretical predictions reported in Ref. 4. In this report, the confined energy states in the quantum dots were calculated by solving the three-dimensional Schrödinger equation for the lens-shaped quantum dots. An aspect ratio of 1:10 was taken between the height and the diameter of the dots. An effective mass of $0.04\,m_0$ was taken for the electrons. An enhancement of the susceptibility was predicted because of the double resonance between the confined states. The data reported in Fig. 2 confirm this theoretical prediction. The difference in the resonance energy between the theoretical and the experimental data is mostly attributed to an under estimation of the lateral size or aspect ratio of the quantum dots and an under estimation of the electron effective mass. In Ref. 4, the energy of the s-p transition for a 2.5 nm height quantum dot was calculated at 80 meV, while it is experimentally found resonant around 60 meV. We emphasize that this double resonance demonstrates that the energy levels are equally spaced for a fraction of the lens-shaped quantum dots, i.e., the energy of the transition $e_{000}-e_{100}$ is strictly equal within the homogeneous broadening to the energy of the transition $e_{100}-e_{110}$. This feature could be of interest to test the prediction of more sophisticated energy level calculation in the dots.[7]

Figure 3 shows the dependence of the second-harmonic power as a function of the polarization of the pump set at 61 meV. The polarization was set with a combination of two polarizers placed in front of the sample. 0° corresponds to a p polarization, while 90° corresponds to an in-plane (s) polarization. The incident power was normalized by the transmission of the two polarizers. The second-harmonic generation is strongly polarized. As expected, the signal vanishes when the polarization is set along the $[-110]$ direction. The full line in Fig. 3 corresponds to a fit expected from theory $[P_{2\omega}\alpha\cos^4(\theta)$ where θ is the polarizer angle]. The inset of Fig. 3 shows the dependence of the second-harmonic power as a function of the output polarization angle. The incident pump beam was p polarized. 0° corresponds to a second-harmonic signal polarized along the p direction, while 90° corresponds to the in-plane direction. The second harmonic is clearly p polarized. This feature is coherent with the predicted orientation of the susceptibility $\chi^{(2)}_{zxx}$.

The measurement of the nonlinear susceptibility through the measurement of the harmonic power requires a careful calibration. We have chosen to measure the quantum dot nonlinear susceptibility by comparison with the susceptibility of bulk GaAs. It is well-known that the conversion efficiency with bulk GaAs depends on the phase-matching condition, i.e., on the propagation length normalized by the number of coherence lengths (L_c). In GaAs, quasiphase matching can be achieved by several techniques. Thompson *et al.* have shown that the stacking of GaAs plates with a thickness of one coherence length and the appropriate orientation can lead to a high-conversion efficiency.[8] Quasiphase matching can also be obtained in a multipass waveguide geometry like the one that we have investigated. In this case, phase matching is achieved by taking advantage of the phase shift upon total internal reflection. This idea was first proposed by Armstrong *et al.*[9] Boyd *et al.* have experimentally shown that by choosing an appropriate tilt of the sample, i.e., varying the number of total internal reflections and the propagation length, quasiphase matching over several coherence lengths can be achieved.[10] The conversion efficiency in bulk GaAs can be increased if the phase shift upon total internal reflection occurs after a propagation of an odd number of coherence lengths. According to the present experimental setup, the propagation length before the first total reflection can be changed by moving the sample laterally. Note that the total propagation length remains constant. As shown in the inset of Fig. 2, the infrared beam is incident on the 45° wedge of the sample. The second-harmonic generation is therefore expected to exhibit maxima as a function of the lateral position of the sample when the propagation length before reflection corresponds to an odd number of coherence length. This effect is shown in Fig. 4. The spacing between two successive maxima correspond to $2L_c$ (i.e., a coherence length of 60 μm for a 20 μm pump wavelength). We emphasize that the coherence length measured with this technique is very close to the theoretical coherence length estimated from the difference in refractive index at this wavelength (53 μm). The amplitude of the peaks are not

correlated since the collection optics were not optimized as the sample was laterally moved. The amplitude of the pump field is also decreased because of the phonon absorption. We have simulated the propagation of the harmonic field in the wave-guide as a function of the total length of the sample and as a function of the distance between two total internal reflections. In order to obtain a curve as shown in Fig. 4 with a marked peak-to-valley ratio ($\sim$30), the conversion length was found to be equal to $2\times L_c$ ($\pm 50\%$). The amplitude of the second-order nonlinear susceptibility can now be calibrated by reference with bulk GaAs ($\chi^{(2)}_{\alpha\beta\gamma}=1.9\times10^{-10}$ m/V).[11] The propagation length in the quantum dot layers is only 34 nm as compared to the 120 μm length for the harmonic conversion in bulk GaAs. In *p* polarization, the effective susceptibility for GaAs resulting from the projection of the susceptibility tensor along the propagation axis is $[3/(2\sqrt{2})\chi^{(2)}_{\mathrm{GaAs}}]$. The nonlinear polarization for the quantum dots is oriented along the [001] direction. The same projection procedure leads to an effective susceptibility equal to $\sqrt{2}/4$ times $\chi^{(2)}_{zxx}$ for the quantum dots. The susceptibility for one quantum dot plane is found four orders of magnitude larger than the susceptibility of bulk GaAs (2.5×10^{-6} ($\pm1.25\times10^{-6}$) m/V as compared to 1.9×10^{-10} m/V). This susceptibility amplitude is the largest susceptibility reported to date to our knowledge in semiconductor materials. It is much larger than the resonant susceptibility reported for midinfrared intersubband transitions in quantum wells[12] and for intersublevel transitions in the valence band of the quantum dots.[4] This giant value of the susceptibility is a clear signature of the enhancement resulting from the double resonance with intersublevel transitions.

The amplitude of the susceptibility can be compared to the theoretical value. In Ref. 4, the susceptibility in the conduction band of one quantum dot plane was predicted to be six orders of magnitude larger ($\sim10^{-4}$ m/V) than the susceptibility of bulk GaAs. The discrepancy by a factor of $\sim$40 between the theoretical and the experimental values is accounted for by a difference in the input parameters: dipole matrix element of the intersublevel transitions, effective carrier density populating the dots, the homogeneous broadening, and the fraction of dots satisfying the double-resonance condition. We emphasize that the second-order nonlinear susceptibility in m/V is calculated with an equivalent three-dimensional carrier density, as done previously with quantum wells. The thickness of the deposited InAs was used in the calculation, and also to deduce the experimental value of the susceptibility. However, isolated quantum dots require a distance of at least 5 nm between the wetting layers. This thickness could also be used to obtain a three-dimensional carrier density. In this case, the theoretical and the experimental values of the susceptibilities would be reduced accordingly. It is important to observe that a more pertinent element of comparison to evaluate the conversion efficiency in these systems (quantum wells or quantum dots) is the product in m^2/V of the susceptibility by the interaction length. We note that the model does not account for the asymmetry of the quantum dots in the layer plane. We emphasize that we cannot deduce the homogeneous broadening of the intersublevel transition from the present experiment. This feature is in contrast with results reported for nonlinear optical spectroscopy of single dots where energy relaxation and dephasing rates were shown to be comparable.[13] In the present experiment, the broadening is limited by the spectral width of the picosecond pulses. An upper limit (0.8 meV at low temperature) can only be estimated. This limit corresponds to a coherence time of $\sim$1 ps. More information on the homogeneous linewidth of intersublevel transitions should be obtained with four-wave mixing experiments.

IV. CONCLUSION

In conclusion, we have observed second-harmonic generation in the conduction band of InAs/GaAs self-assembled quantum dots. The second-harmonic generation is enhanced by the double resonance between the pump beam around 20 μm wavelength and the intersublevel transitions [$e_{000}-e_{100}$ (or *s-p* transition) and $e_{000}-e_{110}$ (or *s-d* transition)]. This double resonance enhancement shows that the energy levels in the conduction band can be equally spaced for lens-shaped quantum dots. The experimental second-order nonlinear susceptibility of one quantum dot plane is found as large as 2.5×10^{-6} m/V, i.e., four orders of magnitude larger than the bulk GaAs susceptibility.

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