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Quantum optical properties of a single $In_xGa_{1-x}As$-GaAs quantum dot two-level system

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We report on a two-level system, defined by the ground-state exciton of a single InGaAs/GaAs quantum dot. Saturation spectroscopy combined with ultrahigh spectral resolution gives us a complete description of the system in the steady-state limit. Rabi oscillations and quantum interference experiments, on the other hand, provide a detailed insight into the coherent high excitation regime. All fundamental properties of the two-level system show an excellent quantitative agreement in both domains, even though obtained under entirely different experimental conditions. We thus are able to demonstrate control over an almost ideal two-level system, suitable for possible applications in quantum information processing.

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Coherent optical manipulations of single quantum systems are currently receiving a lot of attention, not only for their fundamental interest but also in view of possible applications like quantum cryptography and quantum computing.[1] When looking for solid-state implementations, self-assembled semiconductor quantum dots (QDs) are promising candidates, due to their well-separated energy levels and high optical quality.[2–6] By placing an InGaAs QD in a diode structure we essentially obtain a single quantum system with electrical contacts. Voltage-based control and electrical detection here not only open up alternate possibilities for fundamental quantum optical experiments but also might provide a link between quantum systems and classical information technology. We present in this paper a comparison of coherent versus steady-state measurements, providing a very complete picture of a QD two-level system.

Our sample consists of molecular beam epitaxy- (MBE) grown InGaAs/GaAs QDs incorporated in the intrinsic region of an *n*-*i*-Schottky diode, as described in Ref. 7. The optical selection of a single QD is achieved via near-field shadow masks. All measurements are performed in a low-temperature microscope at 4.2 K. For basic QD characterizations, we use photoluminescence (PL) spectroscopy. Here the excitation and detection light both have to pass through the shadow mask, which makes this approach very sensitive to the relative position of the QD with respect to the aperture of the shadow mask. We have measured the PL on several sites in order to locate an aperture with optimal QD alignment. The ground-state transition energy of the selected QD is about 1337 meV as compared to an ensemble average of 1310 meV. We assume therefore that the selected QD is comparatively small. This conclusion is confirmed by the fact that no *p*-shell states could be observed on this particular QD.

We are able to control the internal electric field of the sample by adjusting a bias voltage. At sufficiently high electric fields, photogenerated carriers will mostly tunnel out of the QD rather than to recombine radiatively. The occupancy of the QD then can be monitored via the photocurrent (PC),[8,9] enabling us to perform measurements with resonant excitation of the ground state. Under these conditions the QD can be well described by a two-level system, provided that the excitation is spectrally narrow enough not to address any other states. We are able to obtain PC spectra with ultrahigh resolution by tuning the QD energy levels via the Stark effect.[10,11] Figure 1 displays the QD ground-state transition, revealing a linewidth of $\Gamma=5$ μeV as well as a splitting into two peaks, separated by 30 μeV. This effect is due to exchange interaction, and arises in any QD that is not fully symmetric in the *x*-*y* plane.[12,13] By choosing an appropriate polarization of the excitation beam, we are able to completely suppress each of both peaks with respect to the other. With increasing excitation intensities we observe a saturation of the PC peak height as well as power broadening, in excellent agreement with the theory of two-level systems. A detailed analysis of these effects has been published earlier.[11]

In this paper we want to focus on the coherent behavior of the quantum dot. Due to finite dephasing times we use ul-

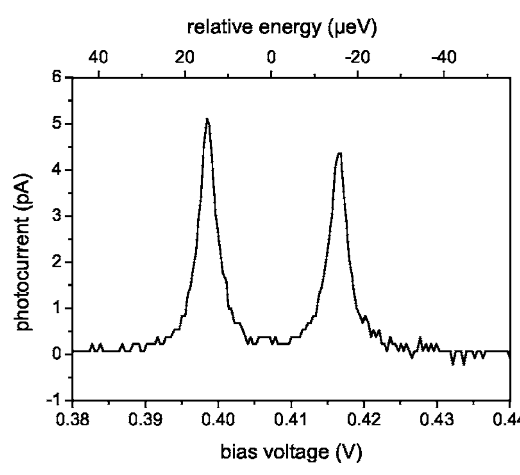

FIG. 1. Photocurrent resonance of the ground-state single exciton at a fixed laser wavelength. Bias voltage levels can be transferred into energy scales via the Stark effect, as indicated by the upper axis. A linewidth of 5 μeV and an asymmetry splitting of 30 μeV are observed.

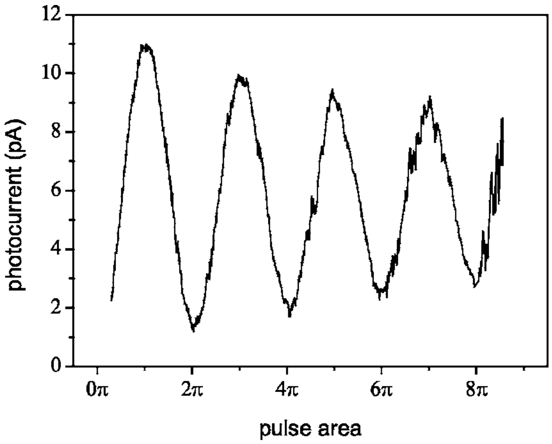

FIG. 2. Exciton Rabi oscillation at excitation with picosecond laser pulses (bias voltage 0.6 V). The oscillation is only slightly damped towards high pulse areas.

trashort laser pulses for excitation. The fundamental experiment in the coherent regime is the observation of Rabi oscillations.[14] The occupancy of the upper level of a two-level system under coherent resonant excitation is given by $\sin^2(\Omega t/2)$,[15,16] where the Rabi frequency Ω is proportional to the square root of the laser intensity and t corresponds to the pulse length. A π pulse thereby results in a complete inversion of the two-level system. In the context of quantum computing, this represents a qubit rotation analogous to the classical NOT operation. We define the pulse area, i.e., the rotation angle $\theta = \Omega t$, by adjusting the excitation amplitude rather than the pulse length. A π pulse typically corresponds to an average laser power on the sample of about 2 μW at a pulse length of 2.3 ps and a repetition frequency of $f_{Laser} = 80$ MHz. If the tunnel efficiency of our device was 100%, any π pulse would contribute to the PC with an elementary charge, resulting in a maximum value of $I = f_{Laser} \times e = 12.8$ pA.[4] At low bias voltage however, the tunneling time increases to values similar to the radiative lifetime, which causes a quenching of the PC. At 0.4 V for example, the maximum observed PC is only about 6 pA, as compared to 12 pA at 0.8 V.

Figure 2 shows the upper level occupancy measured in the photocurrent as a function of the excitation pulse area. At the highest excitation intensities the system undergoes almost nine full inversions with each laser pulse. We use here circular polarized light so that biexciton generation is inhibited not only by spectral separation but also by Pauli blocking.[17] The original measurement is corrected for a background that increases linearly in power (i.e., quadratic in pulse area) and accounts for only 6% of the total signal at π-pulse excitation but exceeds the coherent part at very high intensities. The subtraction of a purely linear background yields a very reasonable result and we hence conclude that the dominant part is caused by linear absorption of stray light in the surrounding of the QD. This assertion is justified by the fact that the photocurrent in our sample is collected over the whole area of a 300×400 μm^2 mesa structure. The data displayed in Fig. 2 were measured at a bias voltage of 0.6 V, but similar results are obtained in the whole range between 0.4 and 0.8 V. This is remarkable because the dephasing time of the system varies within this range by about a factor of 6, as discussed later in this paper. The damping finally increases at voltage levels where the dephasing time is less than 30 ps, which is about 10 to 15 times the pulse length.

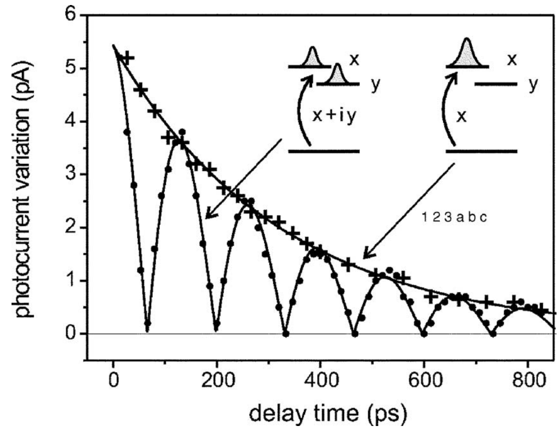

FIG. 3. Quantum interference experiment with varying delay between two $\pi/2$ pulses (bias voltage 0.4 V). The crosses correspond to a measurement at which only one of the asymmetry split levels is addressed. Circles on the other hand, correspond to an excitation of both lines, resulting in quantum beats. Solid lines represent fit curves, from which we deduce a dephasing time of 322 ps and an asymmetry splitting of 31 μeV.

The nature of the damping of pulse area dependent Rabi oscillations has recently been subject of intensive theoretical work (see Refs. 18 and 19 and references therein). The accuracy of the different theoretical models up to now could not be proven with much certainty as experimental data mostly were restricted to pulse areas $<3\pi$ and also showed quite large error bars.[3–6,20,21] By performing accurate measurements on the *ground state* of a well-defined and precisely characterized InGaAs QD we are able to establish here a reliable basis for future theoretical work in the field of dephasing mechanisms in semiconductor QDs.

While a measurement of Rabi oscillations represents the occupancy of a two-level system, we have to perform quantum interference experiments to also gain access to the phase of coherent excitations. First experiments of this kind have been performed in the weak excitation regime, i.e., at pulse areas much less than 1π.[22,23] In the context of quantum information processing, these experiments have to be extended to the strong excitation regime.[3,5,24] We have performed here experiments with $\pi/2$ pulses, which represent a 1-qubit Hadamard transformation.[25] The first pulse thereby creates a coherent superposition of the "0" and "1" state of the QD two-level system. The second pulse then follows with a variable delay in the range of 0 to 1000 ps. The relative phase of the second pulse can be controlled via an additional fine delay with subfemtosecond resolution. If coherence is maintained, the superposition state is expected to be transferred into the pure "1" or "0" state, depending on whether the two pulses are of the same or opposite phase, respectively. When varying the phase continuously, we observe an oscillation of the PC at the same period as for the optical interference at overlapping pulses. The amplitude of these oscillations versus delay time is displayed in Fig. 3. The data represented by crosses correspond to a polarization at which only one of the asymmetry split levels is excited. A fit to these data points reveals a purely exponential decay at delay times >10 ps, corresponding to a dephasing time of T_2=322±5 ps. The analysis of the first few picoseconds is complicated by the fact that an overlap of both pulses to some degree influences the measurement results. We still are able to determine some initial dephasing, though, in the best measurements, this amounts to less than 4%. Going back to Fig. 3, the circles correspond to a polarization at which both states of the asymmetry split doublet are excited. We observe here an additional beat with a period of T=133 ps. As our experiment is insensitive to a phase shift of 180, the data have to be fitted by an exponential decay modulated not by a plain cosine function but by its absolute $|\cos(\pi t/T)|$. The period T can be converted into an energy difference of $\Delta E = h/T = 31\ \mu$eV, in good agreement with the value observed in a direct measurement as displayed in Fig. 1. Moreover, in contrast to any previously reported measurements,[13,26–28] the modulation here amounts to the full amplitude of the reference signal and we are able to fully turn it on and off. This confirms that the exciton superpositions in the asymmetry split levels can be well prepared by selecting the polarization of optical excitation or, more generally, again gives evidence for an excellent control of our quantum system.

We further are able to compare dephasing times measured by quantum interference experiments with those derived by a linewidth analysis. At low bias voltage and accordingly long tunneling times, however, saturation effects result in a broadening of the linewidth even at low excitation intensities.[11] We consequently performed a full power broadening analysis with an extrapolation to zero excitation for all measurements up to 0.6 V. At higher bias voltages, the PC saturation value is high enough so that the linewidth of single low-power spectra will already yield the correct results. The linewidth Γ can be converted into a dephasing time T_2 via $T_2 = 2\hbar/\Gamma$.[15,16] The corresponding data are plotted versus bias voltage in Fig. 4 (full circles). The strong voltage dependence indicates that the dephasing time is primarily determined by the tunneling time. Optical recombination should be only of significance at very low voltage levels. In the investigated regime we therefore have the opportunity to control the main dephasing mechanism of our quantum system by changing the tunneling time via the bias voltage.

T_2 times derived from the quantum interference experiments are shown in the same figure, indicated by triangles. Both sets of data show excellent agreement up to a bias voltage of 0.7 V. This indicates that dephasing times at excitation with $\pi/2$ pulses are exactly the same as for vanishing excitation power. This equality is found to hold for dephasing times between 80 and 320 ps and has to be regarded as an important but previously inaccessible system property. At higher voltages we observe quantum beats independent of the choice of polarization. In this regime it is therefore difficult to infer a dephasing time. The interference at the first maximum of the quantum beats (delay time 133 ps), however, is higher than expected from the linewidth analysis, as indicated by the open triangles. The reason therefore is not fully understood up to now.

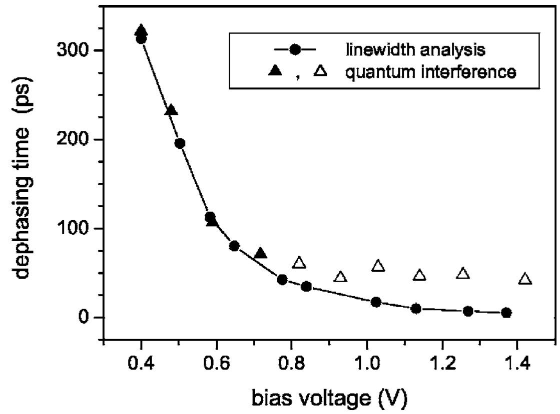

FIG. 4. Comparison of dephasing times derived by linewidth analysis (circles) and quantum interference experiments (triangles). Both sets of data show very good agreement at voltage levels up to 0.7 V. At higher bias voltage, quantum beats could not be suppressed, independent of the polarization of the excitation beam. Dephasing times here are derived from the ratio of the initial signal vs the first maximum of the beats (open triangles).

Resuming the comparison of coherent versus steady-state measurements, we get an agreement in several aspects. The linewidth of the ground-state resonance complies with the decay time of quantum interference. The asymmetry-induced splitting of energy levels is reflected in the period of quantum beats. The polarization at which these effects are suppressed is the same in both measurements. Furthermore, the power dependence observed in cw and pulsed experiments should show some kind of correlation, since the underlying physics in both regimes contain the transition matrix element of the QD. From saturation and power broadening experiments we derive a characteristic dimensionless power level $\widetilde{P} = \Omega^2 T_1 T_2$.[11,15] As T_1 and T_2 times are also obtained from these experiments, we can compare the resulting Rabi frequency Ω to a direct measurement of Rabi oscillations. In cw measurements we typically derive a value of $\Omega \approx 0.2$ GHz at a laser power of P=100 nW. From a comparison of different measurements we get the more general ratio $\Omega/\sqrt{P} = 0.19 \pm 0.4$ THz/$\sqrt{\text{mW}}$. In a measurement of Rabi oscillations as shown in Fig. 2, π-pulse excitation is achieved at an *average* laser power of 2 μW. A conversion to continuous excitation[29] results in a value of $\Omega/\sqrt{P} = 0.25 \pm 0.3$ THz/$\sqrt{\text{mW}}$. This is in remarkably good agreement with saturation measurements, even though the optical peak power typically differs by five orders of magnitude.

In summary, we have performed a whole range of fundamental experiments with respect to two-level systems, both in the steady state as well as the coherent regime. All experimental results can be brought down to few basic properties of the investigated single QD, giving evidence for an almost ideal quantum system. By drawing a comparison of complementary experimental methods we are able to prove that fundamental system properties, including the dephasing time, remain valid over an extraordinarily wide range of experimental conditions. Furthermore, many results mark a major advance in experimental quality and thus are suitable to test and improve the accuracy of theoretical models. In the context of quantum computing the present paper demonstrates excellent control over any 1-qubit manipulation.

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[1] D. Bouwmeester, A. Ekert, and A. Zeilinger, *The Physics of Quantum Information* (Springer-Verlag, Berlin, 2000).
[2] X. Li, Y. Wu, D. Steel, D. Gammon, T. H. Stievater, D. S. Katzer, D. Park, C. Piermarocchi, and L. J. Sham, Science **301**, 809 (2003).
[3] H. Kamada, H. Gotoh, J. Temmyo, T. Takagahara, and H. Ando, Phys. Rev. Lett. **87**, 246401 (2001).
[4] A. Zrenner, E. Beham, S. Stufler, F. Findeis, M. Bichler, and G. Abstreiter, Nature (London) **418**, 612 (2002).
[5] H. Htoon, T. Takagahara, D. Kulik, O. Baklenov, A. L. Holmes, Jr., and C. K. Shih, Phys. Rev. Lett. **88**, 087401 (2002).
[6] P. Borri, W. Langbein, S. Schneider, U. Woggon, R. L. Sellin, D. Ouyang, and D. Bimberg, Phys. Rev. B **66**, 081306 (2002).
[7] F. Findeis, M. Baier, A. Zrenner, M. Bichler, G. Abstreiter, U. Hohenester, and E. Molinari, Phys. Rev. B **63**, 121309 (2001).
[8] P. W. Fry *et al.*, Phys. Rev. Lett. **84**, 733 (2000).
[9] F. Findeis, M. Baier, E. Beham, A. Zrenner, and G. Abstreiter, Appl. Phys. Lett. **78**, 2958 (2001).
[10] A. Hogele, S. Seidl, M. Kroner, K. Karrai, R. J. Warburton, B. D. Gerardot, and P. M. Petroff, Phys. Rev. Lett. **93**, 217401 (2004).
[11] S. Stufler, P. Ester, A. Zrenner, and M. Bichler, Appl. Phys. Lett. **85**, 4202 (2004).
[12] D. Gammon, E. S. Snow, B. V. Shanabrook, D. S. Katzer, and D. Park, Phys. Rev. Lett. **76**, 3005 (1996).
[13] W. Langbein, P. Borri, U. Woggon, V. Stavarache, D. Reuter, and A. D. Wieck, Phys. Rev. B **69**, 161301 (2004).
[14] I. I. Rabi, Phys. Rev. **51**, 652 (1937).
[15] L. Allen and J. H. Eberly, *Optical Resonance and Two Level Atoms* (Wiley, New York, 1975).
[16] P. Meystre and M. Sargent III, *Elements of Quantum Optics*, 2nd ed. (Springer-Verlag, Berlin, 1991).
[17] K. Brunner, G. Abstreiter, G. Bohm, G. Trankle, and G. Weimann, Phys. Rev. Lett. **73**, 1138 (1994).
[18] J. Forstner, C. Weber, J. Danckwerts, and A. Knorr, Phys. Rev. Lett. **91**, 127401 (2003).
[19] J. M. Villas-Boas, S. E. Ulloa, and A. O. Govorov, Phys. Rev. Lett. **94**, 057404 (2005).
[20] T. H. Stievater, X. Li, D. G. Steel, D. Gammon, D. S. Katzer, D. Park, C. Piermarocchi, and L. J. Sham, Phys. Rev. Lett. **87**, 133603 (2001).
[21] L. Besombes, J. J. Baumberg, and J. Motohisa, Phys. Rev. Lett. **90**, 257402 (2003).
[22] N. H. Bonadeo, G. Chen, D. Gammon, D. S. Katzer, D. Park, and D. G. Steel, Phys. Rev. Lett. **81**, 2759 (1998).
[23] P. Borri, W. Langbein, S. Schneider, U. Woggon, R. L. Sellin, D. Ouyang, and D. Bimberg, Phys. Rev. Lett. **87**, 157401 (2001).
[24] P. Machnikowski and L. Jacak, Phys. Rev. B **69**, 193302 (2004).
[25] P. Bianucci, A. Muller, C. K. Shih, Q. Q. Wang, Q. K. Xue, and C. Piermarocchi, Phys. Rev. B **69**, 161303 (2004).
[26] T. Flissikowski, A. Hundt, M. Lowisch, M. Rabe, and F. Henneberger, Phys. Rev. Lett. **86**, 3172 (2001).
[27] K. Nishibayashi, T. Okuno, Y. Masumoto, and H.-W. Ren, Phys. Rev. B **68**, 035333 (2003).
[28] A. I. Tartakovskii, J. Cahill, M. N. Makhonin, D. M. Whittaker, J.-P. R. Wells, A. M. Fox, D. J. Mowbray, M. S. Skolnick, K. M. Groom, M. J. Steer, and M. Hopkins, Phys. Rev. Lett. **93**, 057401 (2004).
[29] Assuming sech^2 pulses with an intensity full width at half maximum of 2.3 ps at a repetition frequency of 80 MHz.