Coherent properties of a two-level system based on a quantum-dot photodiode

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Present-day information technology is based mainly on incoherent processes in conventional semiconductor devices[1]. To realize concepts for future quantum information technologies, which are based on coherent phenomena, a new type of 'hardware' is required[2]. Semiconductor quantum dots are promising candidates for the basic device units for quantum information processing. One approach is to exploit optical excitations (excitons) in quantum dots. It has already been demonstrated that coherent manipulation between two excitonic energy levels—via so-called Rabi oscillations—can be achieved in single quantum dots by applying electromagnetic fields[3–7]. Here we make use of this effect by placing an InGaAs quantum dot in a photodiode, which essentially connects it to an electric circuit. We demonstrate that coherent optical excitations in the quantum-dot two-level system can be converted into deterministic photocurrents. For optical excitation with so-called π-pulses, which completely invert the two-level system, the current is given by $I = fe$, where f is the repetition frequency of the experiment and e is the elementary charge. We find that this device can function as an optically triggered single-electron turnstile.

Semiconductor quantum dots (QDs), often referred to as artificial atoms, are suitable entities with which to implement arrays of quantum bits (qubits) for quantum information processing. One possible approach to this is the use of excitonic excitations in the ground state of a QD as the basis of a two-level system (Fig. 1a). Rabi oscillations have been successfully demonstrated in the exciton population of single QDs[3–5], showing that the abstract model of a two-level system is in fact applicable to the excitonic ground state of QDs. Low-temperature dephasing times for excitons in self-assembled QDs have been shown to exceed several hundred picoseconds, allowing sufficiently high numbers of coherent manipulations with picosecond pulses[8,9].

Here we report the coherent optoelectronic properties of a single QD in a photodiode, which is essentially a single two-level system with electric contacts. Coherent optical excitations of this system are expected to result in Rabi oscillations, which can be directly and quantitatively transferred into photocurrents[10]. For our experiments we fabricated single-QD photodiodes starting from a layer of self-assembled $In_{0.5}Ga_{0.5}As$ QDs grown by molecular beam epitaxy. Although based on a conventional diode structure, here a GaAs n–i-Schottky structure, the only optically active part is a single self-assembled $In_{0.5}Ga_{0.5}As$ QD contained in the intrinsic layer of the diode (Fig. 1b). A semitransparent Schottky contact is provided by a 5-nm-thick titanium layer. The optical selection of a single QD is done by shadow masks with apertures from 100 to 500 nm, which are prepared by electron beam lithography from a 80-nm-thick aluminium layer (see ref. 11 for further details). A schematic diagram of such a diode is shown in Fig. 1c. The QD represents a single two-level system, which is designed to interact with a coherent optical pump pulse (impinging through the shadow mask) by excitonic transitions, and with an electric circuit by tunnelling. It is this connection to the electric circuit which offers those optoelectronic features described above.

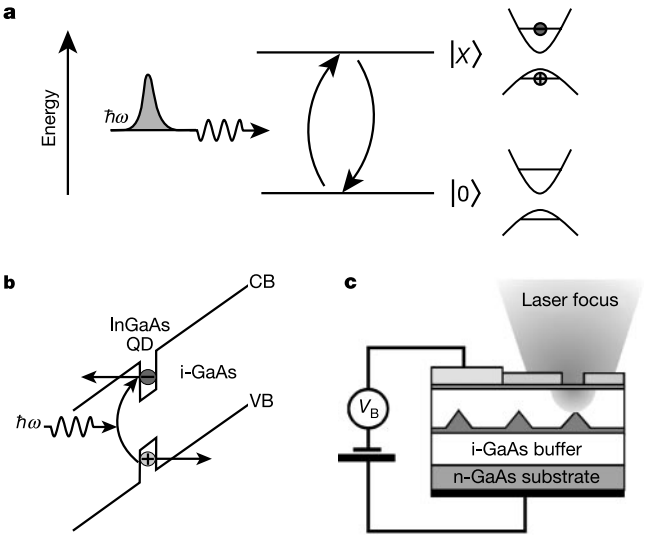

Figure 1 The two-level system in a quantum dot, and the single-quantum-dot photodiode. **a**, Schematic view of an excitonic two-level system in the ground state of a semiconductor QD. The state $|0\rangle$ corresponds to an empty QD, $|X\rangle$ to a one-exciton occupancy. Transitions between both states can be induced by resonant optical driving fields, where the energy $\hbar\omega$ of the optical excitation corresponds to the energetic level spacing between the states $|0\rangle$ and $|X\rangle$. Coherent optical π-pulse excitation in particular then results in an inversion of the two-level system with respect to its initial state: $|0\rangle \Leftrightarrow |X\rangle$. **b**, Intrinsic region of a biased single-QD photodiode: Coherent optical π-pulse excitation applied to the initial state $|0\rangle$ results in the final state $|X\rangle$, which corresponds to the coherent generation of a single electron–hole pair. The photoionization of this state by tunnelling results in the separation of a single electron–hole pair and hence in a deterministic photocurrent. **c**, Schematic view of a single-QD photodiode on the basis of a GaAs n–i-Schottky diode. Optical access to a single QD is provided by a shadow mask. CB, conduction band; VB, valence band.

We use linear photocurrent spectroscopy (Ti:sapphire laser) with continuous wave (c.w.) excitation to characterize our single-QD photodiode[11,12]. With a fixed laser energy E_L close to the QD exciton resonance E_X, we tune the bias voltage V_B and, as a consequence of the quantum confined Stark effect (QCSE), $E_X(V_B)$ is shifted through E_L (see Fig. 2 for $E_L = 1.31$ eV). By repeating this procedure for different E_L, we obtain a magnitude of the QCSE of about $2.4\,\mathrm{meV\,V^{-1}}$ (Fig. 2 inset). The excitonic photocurrent resonance exhibits a typical width of about 30 mV, which corresponds to a spectral linewidth of about 72 μeV. Owing to the nearly linear Stark shift, the V_B axis can be directly transferred to an energy axis. On the basis of this known (approximately linear) QCSE, a single-QD photodiode can be operated as detector with resonant, electric field tunable sensitivity. Within a limited energy range it can be applied as an 'all in one' mesoscopic optical spectrum analyser.

We now consider the coherent properties of a single-QD photodiode, and address the hierarchy of relevant and characteristic timescales in this system. The lifetime of a coherent polarization is given by the decoherence or dephasing time $\tau_{dephase}$. Only within this characteristic time is it possible to observe coherent interactions or to perform coherent manipulations in the system. In the field of semiconductor QDs, experiments on self-assembled InGaAs QDs have shown low-temperature dephasing times in excess of 500 ps (refs 8, 9). The spontaneous radiative recombination lifetime of the exciton τ_{rad} is of the order of 1 ns in typical III/V semiconductor QDs with direct bandgap. It is the longest timescale relevant for excitonic two-level systems. In biased single-QD photodiodes, the tunnelling time τ_{tunnel} is also important. Depending on the electric field, τ_{tunnel} is tunable from infinity to less than 3 ps (ref. 11). For

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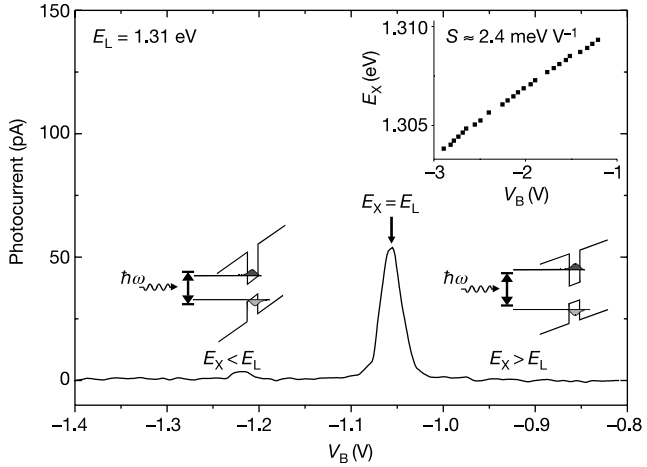

Figure 2 Photocurrent spectrum of a single QD in the region of the excitonic ground state energy E_X. In this c.w. experiment, the laser energy E_L is fixed, whereas E_X is tuned by V_B via the quantum confined Stark effect, QCSE. Within the QCSE tuning range (inset), the single-QD photodiode can be operated as a V_B tunable spectrometer. On the left side of the resonance ($E_X < E_L$), the QD appears red-shifted with respect to the fixed laser energy $E_L = \hbar\omega$, on the right side ($E_X > E_L$), it appears blue-shifted.

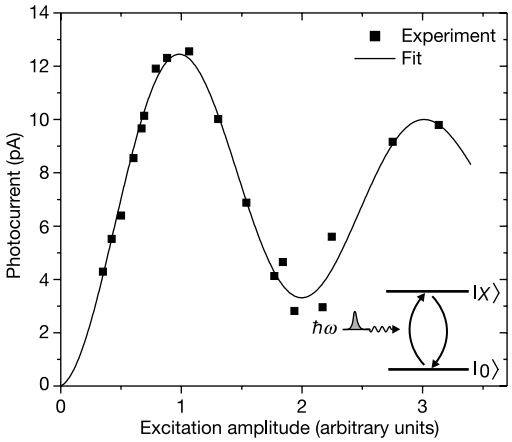

Figure 3 Rabi oscillations of the photocurrent at resonance for increasing excitation amplitude A_{exc}. Coherent π-pulse excitation corresponds to $A_{exc} = 1$. The experimentally observed peak photocurrent is 12.6 pA, only slightly below the predicted limit of $I = fe = 13.1$ pA ($f = 82$ MHz). For π-pulse excitation, the single-QD photodiode therefore performs as optically triggered single-electron turnstile device. The fitted curve assumes an exponential damping of the Rabi oscillations with increasing A_{exc}, and is included as a guide to the eye.

typical bias conditions in our present device ($V_B = -1.04$ V), we infer $\tau_{tunnel} \approx 10$ ps from the photocurrent linewidth. According to simple rate equations, the excitonic excitation decays mostly by tunnelling for $\tau_{tunnel} < \tau_{rad}$, and hence contributes to the photocurrent. Tunnelling on a timescale $\tau_{tunnel} < \tau_{dephase}$, on the other hand, leads to enhanced dephasing, and limits the available time range for coherent interactions to τ_{tunnel}. For future applications, $\tau_{dephase}$ and τ_{tunnel} could be chosen independently by introducing a time-dependent bias voltage V_B (ref. 10). In such an arrangement, coherent manipulations of the two-level system could be performed at zero electric field under the condition of weak dephasing, whereas subsequent read-out would be performed at high electric field.

To reach conditions for coherent excitation for the present case of time-independent V_B, we apply 1-ps pulses from a mode-locked Ti:sapphire laser. Within the laser line width of about 0.7 meV, no other QD state contributes to the photocurrent. With a biexciton binding energy of 3 meV, the two-photon biexciton resonance[13] is sufficiently out of range, 1.5 meV below E_X, which corresponds to a 0.6-V separation on the V_B axis. We varied the pulse intensity over a range of up to two orders of magnitude. For the following analysis, we follow the photocurrent signal on the main resonance at about -1.04 V. In Fig. 3 we show the on-resonance photocurrent as a function of the excitation amplitude (A_{exc}), which is the square root of the peak pulse intensity, the relevant parameter for coherent phenomena. Here the relative unit of 1 corresponds to a time-averaged c.w. excitation power of about 1×10^3 W cm^{-2} (within a 1-μm laser focus) incident on the shadow mask of the diode, for a pulse width of about 1 ps and a pulse repetition frequency of 82 MHz. In the present work we have scaled the excitation amplitude such that $A_{exc} = 1$ corresponds to the condition of an optical π-pulse.

With increasing A_{exc}, we first observe an increasing photocurrent signal from the QD. At $A_{exc} \approx 1$ the current reaches a maximum; a further increase of A_{exc} then leads to a reduction of the photocurrent. This behaviour is in contrast to the well-known characteristics of a conventional, incoherent photodiode, but is expected for the coherent population of a two-level system with increasing pumping power[6,7]. Over the whole range of A_{exc} we observe more than one period of a damped Rabi oscillation in the photocurrent, which reflects directly and quantitatively the resulting occupancy in the two-level system. The observed Rabi oscillations appear damped at high A_{exc}, as previously observed in related work[3–5]. The exact nature of the underlying dephasing mechanism(s) remains an open question. The range around the first maximum of the Rabi oscillations, which corresponds to an excitation with a π-pulse ($A_{exc} = 1$), appears effectively undamped. Applied to a two-level system with an initial occupancy of 0, a single π-pulse projects the system to occupancy 1 (complete inversion). The photoionization of this excitation by tunnelling leads then to the separation of (in the ideal case) exactly one electron–hole pair, and hence to the net transport of one elementary charge between the contacts of the photodiode. In this sense a single-QD photodiode, excited with a π-pulse in the coherent regime, is therefore a deterministic current source, which delivers one elementary charge e to an outer circuit per laser pulse. With the pulse repetition frequency f of our laser (82 MHz), we expect therefore a time-integrated net current $I = fe$ of 13.1 pA. In our present experiment, we obtain a peak photocurrent in the first maximum of about 12.6 pA. This quantitative measure of the occupancy results in about 96% of the maximum possible photocurrent. The demonstration of such a high efficiency in probing the occupancy of a specific quantum-mechanical state became possible by applying the above-described concept of electric readout.

A coherently driven single-QD photodiode is an optically triggered single-electron source, which offers, just like the related single-electron turnstile device[14], single electrons (or holes) on demand. Both devices have the potential to deliver frequency-controlled currents according to $I = fe$. With circularly polarized excitation, a single-QD photodiode has the potential to provide deterministic streams of spin-polarized charges. More subtle points concerning the absolute accuracy of this device, like the influence of incomplete tunnelling or possible avalanche multiplication, will certainly be the subject of future work. By means of single-QD photodiodes, it becomes possible to transfer optical excitations of single quantum systems into deterministic electric currents. This optoelectronic functionality provides a link between coherent optical excitations and electric currents on the single-electron level. We believe that this will allow the electric readout of excitonic quantum gates, and could have a substantial impact on quantum information technology. □

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Competing interests statement

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Evidence for spin–charge separation in quasi-one-dimensional organic conductors

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Interacting conduction electrons are usually described within Fermi-liquid theory[1], which states that, in spite of strong interactions, the low-energy excitations are electron-like quasiparticles with charge and spin. In recent years there has been tremendous interest in conducting systems that are not Fermi liquids, motivated by the observation of highly anomalous metallic states in various materials, most notably the copper oxide superconductors[2,3]. Non-Fermi-liquid behaviour is generic to one-dimensional interacting electron systems, which are predicted to be Luttinger liquids[4,5]. One of their key properties is spin–charge separation: instead of quasiparticles, collective excitations of charge (with no spin) and spin (with no charge) are formed, which move independently and at different velocities. However, experimental confirmation of spin–charge separation remains a challenge. Here we report experiments probing the charge and heat current in quasi-one-dimensional conductors—the organic Bechgaard salts[6–10]. It was found that the charge and spin excitations have distinctly different thermal conductivities, which gives strong evidence for spin–charge separation. The spin excitations have a much larger thermal conductivity than the charge excitations, which indicates that the coupling of the charge excitations to the lattice is important.

Quasi-one-dimensional conductors have been realized in carbon nanotubes[11], in nanoscopic quantum wires based on GaAs–AlGaAs heterostructures[12], in conducting polymers[13] and in crystalline solids[6–10]. The one-dimensionality of these systems results either from a geometrical restriction as in a quantum wire, or from the particular crystal and electronic structure of a three-dimensional material. This is the case in the Bechgaard salts $(TM)_2X$, which consist of stacks of the organic molecules tetramethylselenafulvalene (where TM is TMTSF) or tetramethyltetrathiofulvalene (where TM is TMTTF) with different anions X^- such as PF_6^- and ClO_4^-. The electronic properties are strongly anisotropic, because the overlap of the relevant orbitals is much larger in the stacking direction than perpendicular to this direction. The materials therefore provide excellent model systems for (weakly coupled) chains of interacting electrons. Owing to the full charge transfer of one electron from the chains to each of the anions the band is quarter-filled (or half-filled if a small structural dimerization along the chain direction is taken into account). Because of the large Coulomb interaction between the chain-electrons, the materials must be considered to be strongly correlated Mott–Hubbard systems. The electronic phase diagram (Fig. 1) is extraordinarily rich. Various ordered states occur at low temperatures, where the weak inter-chain coupling causes a higher-dimensional behaviour. Here, we focus on the one-dimensional Luttinger-liquid behaviour expected at elevated temperatures.

To detect signatures of spin–charge separation we studied the electrical (σ) and thermal (k) conductivities of three different materials, the spin-Peierls system $(TMTTF)_2PF_6$, the spin-density wave system $(TMTSF)_2PF_6$, and the superconductor $(TMTSF)_2ClO_4$, which we abbreviate in the following as TM-SP, TM-SDW, and TM-SC, respectively. The experiments are based on a simple

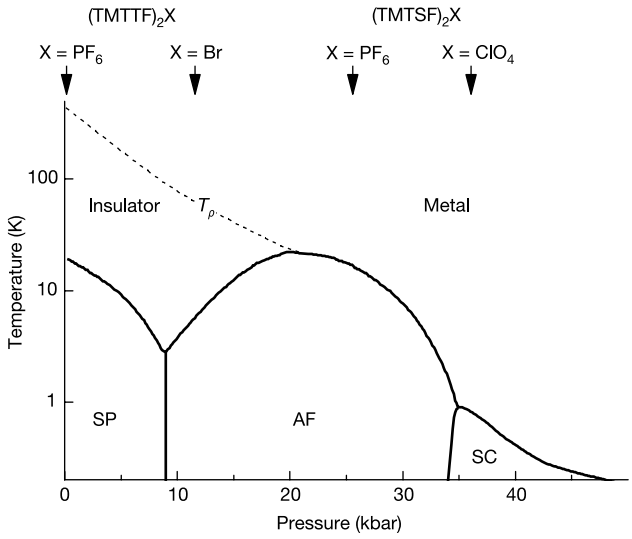

Figure 1 Phase diagram for the $(TM)_2X$ compounds. Pressure or anion (X) substitution yields various phases. The arrows mark the ambient-pressure offset for each compound. The sulphur series (TMTTF) is conducting at high temperatures and insulating below the localization temperature T_ρ. Further cooling results in either spin-Peierls (SP) or antiferromagnetic spin density wave (AF) order. SP order is non-magnetic and results from the dimerization of the lattice and of the spins. The selenide-based salts (TMTSF) are metallic at high temperatures. At low temperatures various ordered phases are found (AF and superconducting (SC)). After ref. 10.

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