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Quantized Photocurrent in a Single-Exciton Pump.

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Abstract. – We propose that a vertical quantum dot under illumination of a pulsed laser can operate as a single-exciton pump. The photocurrent resulting from the creation of a single exciton per laser pulse is equal to the electron charge times the frequency of the laser pulses. The experimental conditions for realizing such a single-exciton pump and its applications are discussed.

The ability to control the transport of single electrons is a fascinating new field of research, not only from a fundamental point of view, but also for obtaining an accurate current standard and for various device applications. So far, most of the effort has been put in the control of single-electron transport through small metallic tunnel junctions. Due to the small junction capacitance C (of order 10^{-16} F), the tunnelling of an electron alters the energy of the system by a charging energy $E_c = e^2/2C$ (of order meV) which exceeds the thermal energy at low temperature ($T < 1$ K). This can result in a Coulomb blockade for other electrons to tunnel [1]. Geerligs *et al.* [2] were able to modulate the Coulomb blockade with a radio-frequent (RF) signal, resulting in the passage of one electron per cycle through their so-called *single-electron-turnstile* device. The current I was found to be given within the error of the experimental set-up ($\sim 0.3\%$) by $I = ef$, where f is the frequency of the RF signal. In a similar system, Pothier *et al.* [3] have realized a *single-electron pump*. Two phase-shifted RF signals were used to generate a current $I = ef$, even for zero bias voltage across the junctions. In a pioneering experiment of Delsing *et al.* [4], single-electron-tunnelling oscillations were phase-locked with a RF signal, also resulting in a current $I = ef$. A semiconductor variant of the single-electron turnstile, which is based on oscillating tunnel barriers, has recently been discussed by Odintsov [5] and realized by Kouwenhoven *et al.* [6].

Besides the above experimental realizations of passing electrons one-by-one, Niu [7] has proposed a system in which the velocity of a Bloch wave can be controlled. Guinea and Garcia [8] proposed that with an oscillating tip of an STM near a metallic grain, one could realize the passage of one electron per cycle from the tip to the grain. The above experiments and proposals are all based on a frequency modulation of the conduction band. In this letter,

we consider an alternative way to obtain a frequency-determined current. We propose that due to the creation of a single exciton in a semiconductor quantum dot per cycle of a pulsed laser, the resulting photocurrent is determined by the frequency of the laser pulses. We will first point out the general operating principles of this *single-exciton pump* and specify the details later.

The nonlinear transport properties of a quantum dot have been studied by Reed *et al.* [9]. Their quantum dot was defined by etching techniques in a vertical layered heterostructure, consisting of n^+ GaAs (contact), AlGaAs (tunnel barrier), InGaAs (quantum dot), AlGaAs (tunnel barrier), and n^+ GaAs (contact). The structure observed in the *I-V* characteristics was identified to result from zero-dimensional (0D) electron states with energy separations of about 25 meV. Figure 1*a*) shows schematically the bottom of the conduction band and the top of the valence band vertically through the quantum dot structure of fig. 1*b*), whose optical properties we discuss in this letter. In the regions I, II, and III of fig. 1*a*), the Fermi energy E_F is taken to lie in the gap. The energy states in the quantum dot (*i.e.* between the barriers) are quantized in all three spatial directions and are therefore 0D electron states. The 0D states can be doubly occupied due to spin-degeneracy. We denote the energy of the n-th 0D state by E_n. The 0D states can be populated by applying a large electric field over the structure, which lifts the Fermi energy of one of the contacts above the conduction band in the buffer region, allowing electrons to tunnel through the barrier into the dot. This is the way Reed *et al.* [9] observed the 0D states, demonstrated by resonant transmission occurring when E_F was lined up with the discrete energy of a 0D state.

An alternative way to populate the 0D states is by creating excitons with a laser [10]. Information about the confined exciton energy states can be obtained by measuring the luminescence spectrum ([1]). We take the energy of the laser $h\nu_1 = E_1 + E_1' + E_{gap} - E_B$ such that at most two excitons can be created in the lowest energy state, while the higher states corresponding to $E_{n>1}$ stay empty. The binding energy E_B includes the Coulomb interaction between the electrons and holes, which gives a correction to the single-particle discrete states E_n and E_n'. Due to stimulated emission, which is of equal probability as the absorption of a photon, the mean occupation is only one exciton. The statistical deviation from this is

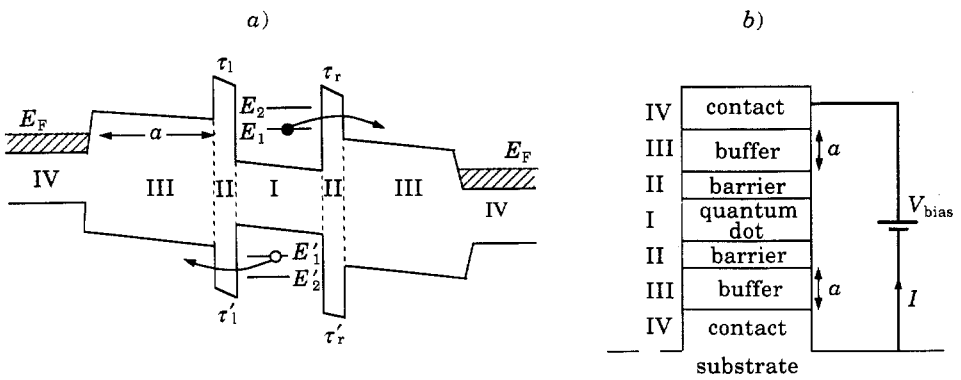

Fig. 1. – *a*) Bottom of the conduction and top of the valence band, which defines a quantum dot (region I) between two barriers (regions II) and separated by two buffers (regions III) from the two electron reservoirs (regions IV). The bands are slanted by a voltage between the reservoirs. *b*) Pilar of different layers of semiconductor alloys, which has the band gap modulation of *a*), see also Reed *et al.* [9].

([1]) The 1D quantum-confined energy states of a narrow wire have been observed in photoluminescence measurements by Kohl *et al.* [11]. To our knowledge no successful luminescence experiments have been performed, which probe the 0D states in quantum dots.

small if many events are considered. For the moment we take the occupation to be exactly one and return to this point later.

We denote the time for an electron to tunnel to the left as τ_l and to the right as τ_r. Similar, τ_l' and τ_r' denote the tunnel times for the hole (we neglect the dependence of the times $\tau_{l,r}$ and $\tau_{l,r}'$ on the discrete energies E_n and E_n'). If the bands are slanted by a voltage across the structure, as indicated in fig. 1*a*), the excited electron has a preference to tunnel to the right ($\tau_r \ll \tau_l$). Similarly, the hole will preferably tunnel to the left ($\tau_l' \ll \tau_r'$). The energy E_B to break up the exciton is delivered by the voltage source. With a continuous operation of the laser, the resulting photocurrent is of order $e/(\tau_{exc} + \tau_r + \tau_l')$, where τ_{exc} is the time to create an exciton. It is important to note that this current from the left- to the right-reservoir results from the pumping action by the laser in the quantum dot only. The excitations in the reservoirs relax in the same reservoir, if the relaxation length is much shorter than the distance a of the buffer regions (the regions III in fig. 1), which separate the quantum dot from the reservoirs. In the buffer regions no excitations can occur, because the energy gap there is larger than the laser energy. *So the pumpimg takes place in the quantum dot, and only the first state contributes to the photocurrent.*

We now consider the pumping if the laser is pulsed with a pulse time T_{pulse} and a waiting time T_{wait} between pulses, so that the pulse frequency $f = 1/(T_{pulse} + T_{wait})$. If $T_{pulse} \gg \tau_{exc}$, the first state will be populated with one exciton. If $\tau_r \ll \tau_{rec}$ (τ_{rec} is the recombination time of the exciton, or equivalently, the radiative life time), and taking $T_{wait} \gg \tau_r$, the electron will tunnel out to the right-reservoir before recombining and before the next pulse. Taking also $T_{wait} \gg \tau_l'$, *i.e.* the hole tunnels to the left, then one electron has been transported from the left- to the right-reservoir in the cycle time $T_{pulse} + T_{wait}$. Note that «multi-pumping» can occur when T_{pulse} is of order $\max(\tau_r, \tau_l')$. In this case, an exciton can tunnel out of the dot within the pulse time, which leaves the possibility that an extra exciton is created during the pulse. Multi-pumping is suppressed if $T_{pulse} \ll \max(\tau_r, \tau_l')$. If we combine all the above conditions for the different times we get

$$\tau_{exc} \ll T_{pulse} \ll \max(\tau_r, \tau_l') \ll \tau_{rec}, T_{wait} . \tag{1}$$

If condition (1) *is satisfied, one electron is transported from the left to the right reservoir within the cycle time* $T_{pulse} + T_{wait}$. *Repeating this process with a frequency* f *yields a quantized photocurrent* $I = ef$. Returning to the statistical deviation in the occupation of the first state by one exciton due to stimulated emission, we note that currents are measured on a time scale of a second, which makes the relative deviation $\sqrt{f}/f \approx 10^{-4}$ for a frequency of 100 MHz. The practical implications of condition (1) will be discussed below. Furthermore, we note that in contrast to the electron turnstiles and pumps [2, 3, 6], the charging energy does not affect the operation of this exciton pump.

Spin-flip processes do not affect the photocurrent. In fact, a spin-flip of the excited electron suppresses stimulated emission, which enhances the probability of an occupation of one exciton. Spin-flip processes do affect the photocurrent if the laser light is circular polarized, such that only electrons with the proper spin direction are excited. This results in an occupation probability of 1/2, and consequently in a photocurrent $I_{pol} = ef/2$. In this case, spin-flip processes increase the photocurrent to a value between $ef/2$ and ef.

If the (nonpolarized) laser energy $h\nu$ is increased to $h\nu_n = E_n + E_n' + E_{gap} - E_B$, excitons can be excited to the n-th state. All states up to E_n can be populated if the laser has a (quasi-continuous) energy band with minimum energy $h\nu_1$ and maximum $h\nu_n$. The pulsed mode now results in the transportation of n electrons per cycle, yielding a photocurrent $I = n \cdot ef$. The photocurrent increases in steps of ef when the maximum laser energy is increased. The plateau width is a direct measure of the energy difference between consecutive exciton states.

If the pulse frequency f is changed, the photocurrent will change proportionally with a slope ne depending on the laser energy $h\nu_n$. Deviations from this dependence can give information about the tunnel, recombination and excitation times. For instance, if $I > ef$ for $h\nu = h\nu_1$, multi-pumping may occur when $T_{\text{pulse}} \approx \max(\tau_r, \tau_l')$. On decreasing T_{pulse} until $I = ef$, one can obtain information about the tunnel times τ_r and τ_l'. In a similar way, if $I < ef$ for $h\nu = h\nu_1$, one can compare T_{pulse} with τ_{exc} by changing the power of the laser.

The photocurrent can, in principle, be enhanced to $I = N \cdot ef$ by putting N equal quantum dots in parallel, which can considerably increase the quantized photocurrent. The condition is that the spread in energy levels should be much smaller than the energy separation between consecutive states.

A realization of a single-exciton pump depends on the possibility of satisfying condition (1). Starting from the left, we note that the time τ_{exc} to create an exciton depends on the laser power and is typically of order 1 fs. The spread $\delta h\nu$ in the laser energy should be much smaller than the separation ΔE_{0D} between consecutive 0D states. This gives a lower limit for the pulse time, because of Heisenberg's uncertainty principle: $\delta h\nu \approx \hbar / T_{\text{pulse}} \ll \Delta E_{0D}$. Taking $\Delta E_{0D} \approx 25$ meV [9], this yields $T_{\text{pulse}} \gg 0.01$ ps. For the waiting time there is no fundamental restriction. Technically, T_{wait} is tunable in multiples of ~ 10 ns. The typical quantized photocurrent is then $e \cdot 100\,\text{MHz} = 16\,\text{pA}$, which is easily measurable. The times $\tau_{l,r}$ and $\tau_{l,r}'$ to tunnel out of the quantum dot include the single-particle dwell time (following from the confinement), and the effect of the binding energy on the nonradiative exciton life time, which both depend strongly on the voltage across the sample. With appropriate barriers, the tunnel times can, therefore, be tuned by the voltage across the sample. Tunnel times of order 10 ps would still satisfy condition (1). The most uncertain time is the recombination time τ_{rec}, which is unknown for quantum dots. In 3D GaAs a recombination time of 3.3 ns was found [12]. When the volume of the quantum dot becomes smaller than the exciton coherence volume, it is expected that the recombination time increases [13]. For our purposes a recombination time of order 1 ns is required.

Combining again the different times, we get

$$\tau_{\text{exc}} \approx 0.001\,\text{ps} \ll T_{\text{pulse}} \approx 0.1\,\text{ps} \ll \max(\tau_r, \tau_l') \approx 10\,\text{ps} \ll \tau_{\text{rec}} \approx 1000\,\text{ps}.$$

With these numbers the photocurrent is quantized in multiples of ef with an accuracy of about 1%.

The main problem in realizing the single-exciton pump is the fabrication of the quantum dots. The etching usually induces a depletion region containing trapped surface states. This gives the possibility for the hole to hop out of the confined state to a surface state. This destroys luminescence, because the excited electron cannot recombine anymore with a hole in the confined state in the valence band. Consequently, such a process may be advantageous for the exciton pump, because it suppresses recombination. The exciton pump may therefore serve as a tool to study the time and energy properties of 0D exciton states.

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