Generation of one-photon states by a quantum dot

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A technique for preparation of a one-photon wave packet through action of a classical electromagnetic field on a semiconducting quantum dot is proposed. We demonstrate that the Coulomb repulsion between charge carriers allows one to select the frequency, amplitude, and duration of an electromagnetic pulse so that one electron will transfer from an upper size-quantized level of the valence band to a lower size-quantized level of the conduction band with a probability close to unity. As a result of radiative recombination of the produced electron–hole pair, exactly one photon is emitted (a one-photon wave packet). This source of one-photon states can be used in quantum systems of data transmission and in quantum computers. © *1997 American Institute of Physics.* [S1063-7761(97)00810-X]

1. INTRODUCTION

''Classical'' states of the electromagnetic field containing a large (formally infinite) number of photons have by now been studied in detail. This includes coherent and compressed states,[1] in particular. In recent years, interest in ''constructing'' nonclassical photon states has risen. For example, nonlocal two-photon interferometry[2,3] and quantum cryptosystems[4–7] require so-called entangled photon states.[8] A line of research in modern quantum physics such as quantum cryptography is largely based on application of one-photon states, since the secrecy of quantum cryptographic systems depends on the impossibility of cloning (copying) *a priori* unknown quantum states.[9]

Presently the focus of studies in quantum cryptography has shifted from purely theoretical analysis of the problem to designing experimental facilities for quantum data transmission, for the time being over relatively short distances (30 km; see Ref. 5). This effort requires reliable sources of one-photon states. Here, reliability means that the sources generate only one-photon states (but not states with two or more photons).

What can be a source of such states? The simplest method of obtaining an ''almost one-photon'' state is to alternate a coherent laser output originally containing many Fock states[5] $|n\rangle$ (this technique was used in experiments in quantum cryptography based on phase coding[6,7]). The field at the output of an ideal laser above threshold can be described as a coherent state $|\alpha\rangle$ of the form

$$|\alpha\rangle=\exp\left(-\frac{|\alpha|^2}{2}\right)\sum_{n=0}^{\infty}\frac{\alpha^n}{\sqrt{n!}}|n\rangle. \tag{1}$$

The coherent state attenuated downstream of an absorbing medium corresponds to the limit of small $|\alpha|$ (recall that the intensity, i.e., the average number of photons, of a monochromatic coherent field is proportional to $|\alpha|^2$). Expanding Eq. (1) in terms of $|\alpha|\ll 1$, we obtain

$$|\alpha\rangle\approx\left(1-\frac{1}{2}|\alpha|^2\right)|0\rangle+\alpha|1\rangle+\frac{|\alpha|^2}{\sqrt{2}}|2\rangle. \tag{2}$$

This equation shows that the largest contribution to the coherent state of the photon field is due to the vacuum component $(1-|\alpha|^2/2\approx 1)$, whereas the contribution of the one-photon Fock component $|1\rangle$ is small insofar as $|\alpha|\ll 1$. The latter condition means that the probability of detecting a single photon is also small.

It would seem that the situation could be improved by increasing $|\alpha|$, i.e., the intensity. But then, although the contribution of the vacuum component to $|\alpha\rangle$ would decrease (see Eq. (2)), the probability of emitting two photons (Fock state $|2\rangle$) would markedly increase. In the quantum cryptography this would destroy the security of the cryptosystem, since a fraction of photons could be ''extracted'' from the line and used for eavesdropping.

Application of the attenuated state described by Eq. (2) with $|\alpha|\ll 1$ to quantum cryptography leads to another difficulty. The problem is that a cryptosystem based on the principle of phase coding[6] ensures secrecy only if the states with phase shifts 0°/180° and 90°/270° are orthogonal to one another (see Ref. 6 for details). The overlap of these states, however, is always nonzero, being proportional to $\exp(-|\alpha|^2/2)$, and the smaller the value of $|\alpha|$, the closer to unity this overlap.

Note that in quantum cryptography, the requirement that the one-photon state be monochromatic is not absolute. It suffices to prepare a one-photon packet that can be described by the formula[10–12]

$$|1\rangle_f=\sum_{\omega=0}^{\infty}f(\omega)\hat{a}^+(\omega)|0\rangle=\sum_{\omega=0}^{\infty}f(\omega)|1_\omega\rangle, \tag{3}$$

where $\hat{a}^+(\omega)$ is the boson creation operator of monochromatic state $|1_\omega\rangle$. The one-photon nature of the wave packet $|1\rangle_f$ in Eq. (3) means that only states with population $n=1$

are included in the expansion of $|1\rangle_f$ in Fock states $|1_\omega\rangle$, although they may have different frequencies. This state of the photon field is a ''one-particle'' quantum state, and it generates only a single activation of a light detector, or only one transition in a detecting system (it cannot be split so as to generate two detector outputs or two transitions). In this sense, the state $|1\rangle_f$ can be termed as a ''single nonmonochromatic photon.'' The function $f(\omega)$ defines the spectrum of the emitted wave packet, which formally contains an infinite number of modes $|1_\omega\rangle$, but with amplitudes $f(\omega)$ whose absolute values are smaller than unity. This function is normalized,

$$\sum_{\omega=0}^{\infty} |f(\omega)|^2 = 1,$$

and is assumed to be centered about a certain carrier frequency ω_0.

The one-photon wave packet (3) can be produced in the following manner. One electron, which is initially in a quasistationary excited state $|e\rangle$ with energy ε_e, is transferred to the ground state $|g\rangle$ with energy $\varepsilon_g < \varepsilon_e$ as a result of interaction with the photon field, which is initially in the vacuum state.

Thus, the problem of generating a one-photon state can be reduced to the problem of preparing a quasistationary excited state of a single electron. This problem has been discussed in the literature. For example, Imamoglu and Yamamoto[13] suggest a method for fabricating such a state in an Al–GaAs–GaAs $p-i_p-i-i_n-n$ heterostructure by applying an ac voltage with specially selected parameters. In this process, one electron and one hole resonantly tunnel from the contacts (across the potential barriers) into the structure and annihilate with emission of a photon. In their opinion, the effects of size quantization and Coulomb blockade should suppress quantum fluctuations associated with the injection of electrons and holes. This scheme, however, like other similar designs, imposes stringent requirements on the parameters of the nanostructure and applied electric field, so that experimental implementation is questionable.

One can suggest another method for exciting a single electron to a quasistationary level, namely, to apply an ac electromagnetic field to an isolated quantum dot with size-quantized levels in both the valence and conduction bands. At first sight, this suggestion seems trivial. Indeed, suppose that all levels of the valence band are occupied, and all conduction band levels are vacant (which is usually the case in a semiconductor). Let us denote by ε_v and ε_c the energies of the upper (occupied) size-quantized level $|v\rangle$ in the valence band and the lower (vacant) level $|c\rangle$ in the conduction band. It is well known that a periodic perturbation $\hat{V}\cos(\Omega t)$ (for example, a classical ac electric field) with frequency $\Omega = \varepsilon_c - \varepsilon_v$ (hereinafter we take $\hbar = 1$) leads to periodic oscillation of the probabilities p_v and p_c of detecting an electron in levels $|v\rangle$ and $|c\rangle$.[14–17] If $p_v(0) = 1$ and $p_c(0) = 0$ at the initial moment, then

$$p_c(t) = \sin^2(\omega_R t), \tag{4}$$

where $\omega_R = |\langle c|\hat{V}|v\rangle|/2$. Here $\langle c|\hat{V}|v\rangle$ is the matrix element of the interband transition. It follows from Eq. (4) that one can select the time T during which the perturbation is on (for example, $T = \pi/2\omega_R$) so that the condition $p_c(T) = 1$ is satisfied (so-called π-pulse[14–17]). Then at time T, the electron is in the quasistationary state $|c\rangle$ with probability one.

This scheme, however, applies only when the levels $|v\rangle$ and $|c\rangle$ are nondegenerate. For example, suppose that they are doubly degenerate in the spin projection, and at $t = 0$ level $|v\rangle$ contains not one but two electrons. One can easily calculate the probability that the periodic perturbation $\hat{V}\cos(\Omega t)$ has transferred only one electron (its spin projection is unimportant) to level $|c\rangle$, whereas the other has remained in level $|v\rangle$:

$$p_{vc}(t) = \frac{1}{2}\sin^2(2\omega_R t). \tag{5}$$

It follows from Eq. (5) that $p_{vc} \leqslant 1/2$, and one cannot be sure that exactly one electron has been transferred to the quasistationary state $|c\rangle$ if levels $|c\rangle$ and $|v\rangle$ are degenerate (the limitation $p_{vc} \leqslant 1/2$ is due to the ''transfer of probability'' to the state with two electrons on level $|c\rangle$, i.e., $p_{cc} \leqslant 1$).

Whereas the degeneracy in both angular momentum and its projection is usually lifted because a quantum dot is usually not spherically symmetric,[18] spin degeneracy persists in the absence of a magnetic field. To lift this degeneracy, one must apply a very strong magnetic field. Let us estimate its strength H. Our estimate is based on the assumption that the longest experimentally attainable electron lifetime in the excited level $|c\rangle$ of the quantum dot is $\tau \approx 10^{-10}$–10^{-9} s. The perturbation duration time T needed to transfer an electron from the valence to conduction band should be much shorter than τ, otherwise the probability of photon emission at $t < T$ is high, and the emitted photon will be ''lost'' in the perturbation field. Thus, we should have $T \approx 10^{-12}$–10^{-11} s. But in this case the perturbating field contains harmonics in the frequency range $\delta\omega \approx 1/T \approx 10^{11}$–$10^{12}$ s^{-1}. The bandwidth $\delta\omega$ should be much smaller than the Zeeman splitting energy ($\approx \mu_B H$) of the size-quantized levels $|v\rangle$ and $|c\rangle$, since otherwise the external field will still mix all the states $|v\uparrow\rangle$, $|v\downarrow\rangle$, $|c\uparrow\rangle$, and as before $|c\downarrow\rangle$, i.e., although the spin degeneracy is formally lifted, its consequences persist. As a result, we have an estimate $H \gg 10$ T, which is very difficult to achieve experimentally.

In this paper we suggest an alternative approach to the problem. It is based on consideration of the Coulomb repulsion between electrons with different spin projections that occupy (in the absence of interaction between them) the same one-electron level (in a real quantum dot, Coulomb repulsion is always present, and the smaller the dot the stronger it is). It is clear from qualitative considerations that if the Coulomb repulsion (for example, in level $|c\rangle$) were infinite, the transition of a second electron to this level would be forbidden, and one could generate the one-electron state with probability $p_{vc} = 1$ at some time $t = T$. But if the Coulomb repulsion energy is finite, this is not obvious.

Our aim is to check whether the transition of one electron from the valence to conduction band is possible under

the action of a periodic perturbation on a quantum dot with finite Coulomb interaction. It will be shown that for realistic parameters of the system (Coulomb repulsion energy, frequency and amplitude of applied periodic electric field, and lifetime of the electron in the quasistationary state), one can select a pulse duration T such that the probability of driving one electron to the conduction band is close to unity. Thus, it is possible to design a source of one-photon wave packets on the basis of an isolated quantum dot without resorting to sophisticated techniques.

2. QUALITATIVE ESTIMATES. DESCRIPTION OF THE MODEL

Let us first approximately estimate the splitting $\Delta\varepsilon$ of the doubly degenerate (in the spin projection) size-quantized level $|v\rangle$ due to the Coulomb interaction between electrons in states $|v\uparrow\rangle$ and $|v\downarrow\rangle$:

$$\Delta\varepsilon \approx U = \left\langle v\uparrow \left| \frac{e^2}{\varepsilon|\mathbf{r}_1-\mathbf{r}_2|} \right| v\downarrow \right\rangle \approx \frac{e^2}{\varepsilon a} \tag{6}$$

is the matrix element of the Coulomb interaction, ε is the static dielectric constant, and a is the characteristic linear dimension of the quantum dot. It follows from Eq. (6) that at $\varepsilon \approx 10$ and $a \approx 10$ nm, we have $U \approx 10$ meV. Our estimate of U is in agreement with the experimental data to order of magnitude.[19] A similar estimate is, naturally, obtained for the splitting of level $|c\rangle$ in the conduction band due to the Coulomb repulsion between electrons in states $|c\uparrow\rangle$ and $|c\downarrow\rangle$.

As was demonstrated in Introduction, it is possible to excite exactly one electron to the conduction band with probability $p_{vc}=1$ only when $T \ll \tau$ and $1/T \ll \Delta\varepsilon$. Thus, we obtain from Eq. (6)

$$1/\tau \ll 1/T \ll U. \tag{7}$$

Since $1/\tau \approx 10^{-3}-10^{-2}$ meV, we have $1/T \approx 0.1-1$ meV, so condition (7) can be satisfied for $U \geqslant 1-10$ meV, i.e., as follows from Eq. (6), for $a \leqslant 10-100$ nm. The range of admissible T can, of course be broadened. To this end, one should increase τ and/or reduce a. For our purposes, however, this range of T is sufficient, since condition (7) is then satisfied at realistic values of τ and a in semiconducting quantum dots fabricated using contemporary technologies.

Note also that we would be satisfied, in principle, by the situation in which the maximum value of $p_{vc}(t)$ is not exactly unity, but very close to it. Therefore, we determine the time T of the external perturbation duration by the condition

$$p_{vc}(T) = \max[p_{vc}(t)]. \tag{8}$$

In addition, let us agree to select the minimal value of T among those satisfying condition (8) (otherwise the first of the inequalities in Eq. (7) can be violated).

Before proceeding to the description of our model, let us somewhat simplify the problem. Assume that both the valence and conduction bands contain only one size-quantized level that is doubly degenerate in the spin projection. Generally speaking, this is not so. Physically, this simplification means that we take into account only electronic transitions from the upper valence band level to the lower conduction band level, and ignore low-lying (fully occupied) levels of the valence band and upper (vacant) levels of the conduction band (the effect of these levels will be discussed below). In describing Coulomb repulsion, we use the Hubbard model,[20] which is widely used in the theory of systems with strong Coulomb correlations.[21]

Thus, taking into account the discussion above, we express the model Hamiltonian of a semiconducting quantum dot in a periodic electric field as

$$\hat{H}(t) = \sum_{\sigma} (\varepsilon_v \hat{a}^+_{v\sigma}\hat{a}_{v\sigma} + \varepsilon_c \hat{a}^+_{c\sigma}\hat{a}_{c\sigma}) + U_v \hat{n}_{v\uparrow}\hat{n}_{v\downarrow} + U_c \hat{n}_{c\uparrow}\hat{n}_{c\downarrow} + E(t)\sum_{\sigma} (d_{cv}\hat{a}^+_{c\sigma}\hat{a}_{v\sigma} + d_{vc}\hat{a}^+_{v\sigma}\hat{a}_{c\sigma}), \tag{9}$$

where $\hat{a}^+_{q\sigma}(\hat{a}_{q\sigma})$ is the creation (annihilation) operator for an electron with spin $\sigma=\uparrow$ or $\downarrow$ in size-quantized levels $|q\rangle=|v\rangle$ and $|c\rangle$ (the subscript v refers everywhere to the valence band, and subscript c to the conduction band); $\hat{n}_{q\sigma}=\hat{a}^+_{q\sigma}\hat{a}_{q\sigma}$ is the particle number operator (population number); ε_q are energies of one-electron levels; U_q is the Hubbard repulsion energy between two electrons in the same level; d_{cv} is the matrix element of optical dipole transitions ($d_{vc}=d^*_{cv}$); and $E(t)$ is the external classical ac field.

Let us suppose that the external field is turned on at $t=0$ and turned off at $t=T$, and has a carrier frequency Ω, i.e.,

$$E(t) = E_0 \cos(\Omega t)\,\theta(t)\,\theta(T-t),$$

where E_0 is the field amplitude, $\theta(t)$ is the Heaviside step function. The duration T of the applied field action and its frequency Ω are to be drived by maximizing the probability that only one electron is transferred from the valence band to the quasistationary level of the conduction band (Eq. (8)).

Since we consider two electrons with different spin projections in two different one-electron energy levels, the total number of two-electron states of the system is four. We denote these states by

$$|1\rangle=|v\uparrow,v\downarrow\rangle, \quad |2\rangle=|v\uparrow,c\downarrow\rangle, \quad |3\rangle=|v\downarrow,c\uparrow\rangle, \quad |4\rangle=|c\uparrow,c\downarrow\rangle, \tag{10}$$

where, for example, $|v\uparrow,c\downarrow\rangle$ means that the electron with $\sigma=\uparrow$ is in the valence band, and the electron with $\sigma=\downarrow$ is in the conduction band, etc. For the system under consideration, the set of states defined by Eq. (10) is complete, so the wave function $\Psi(t)$ can be expressed at any moment as

$$\Psi(t) = \sum_{i=1}^{4} A_i(t)\exp(-iE_i t)|i\rangle, \tag{11}$$

where E_i are eigenvalues of the time-independent Schrödinger equation $\hat{H}|i\rangle = E_i|i\rangle$ in the absence of an applied field ($t \leqslant 0$). With due account of Eq. (10), we derive from Eq. (9)

$$E_1 = 2\varepsilon_v + U_v, \quad E_2 = \varepsilon_v + \varepsilon_c, \quad E_3 = \varepsilon_v + \varepsilon_c, \quad E_4 = 2\varepsilon_c + U_c. \tag{12}$$

Note once again that we use the complete set of two-electron states defined by Eq. (10) and schematically shown in Fig. 1.

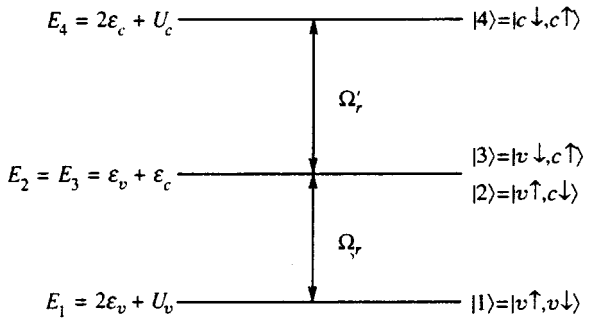

FIG. 1. Diagram of energy levels of two-particle states of a semiconducting quantum dot.

The values $A_i(0)$ define the system wave function at the initial moment (moreover, $A_i(0)=A_i(t<0)$ since the $|i\rangle$ are eigenstates of Hamiltonian (9) for $t\leqslant 0$). We assume that at $t\leqslant 0$ the semiconductor is in its usual state, namely, the valence band is fully occupied and the conduction band is empty, i.e., $A_1(0)=1$, $A_2(0)=A_3(0)=A_4(0)=0$. The probability $p_i(t)$ of detecting the system in state $|i\rangle$ at an arbitrary time t is $|A_i(t)|^2$. We are interested in the value of $p_{vc}(t)=p_2(t)+p_3(t)$, which is the probability (see Eq. (10)) that both the valence and conduction bands contain one electron each (it follows from the normalization condition (11) that, naturally, $p_1(t)+p_2(t)+p_3(t)+p_4(t)=1$ at any t).

The coefficients $A_i(t)$ in expansion (12) can be calculated by solving the time-dependent Schrödinger equation

$$i\,\frac{\partial\Psi(t)}{\partial t}=\hat{H}(t)\Psi(t), \tag{13}$$

where $\hat{H}(t)$ is given by Eq. (9), i.e., it explicitly depends on time when $0\leqslant t\leqslant T$.

3. RESONANT APPROXIMATION

In order to solve the problem as formulated, we use the resonant approximation,[14–17] which enables us to progress reasonably far analytically and obtain an accurate solution (in some limiting cases in analytic form). One can, certainly, go beyond the resonant approximation, but in this case the solution can be obtained only numerically and, as will be shown below, this will not lead to any significant changes in the results. In the resonant approximation, the Hamiltonian (9) has the form

$$\begin{aligned}\hat{H}(t)=&\sum_\sigma\,(\varepsilon_v\hat{a}^+_{v\sigma}\hat{a}_{v\sigma}+\varepsilon_c\hat{a}^+_{c\sigma}\hat{a}_{c\sigma})+U_v\hat{n}_{v\uparrow}\hat{n}_{v\downarrow}\\&+U_c\hat{n}_{c\uparrow}\hat{n}_{c\downarrow}+\sum_\sigma\left[\frac{\lambda}{2}\exp(-i\Omega t)\hat{a}^+_{c\sigma}\hat{a}_{v\sigma}\right.\\&\left.+\frac{\lambda^*}{2}\exp(i\Omega t)\hat{a}^+_{v\sigma}\hat{a}_{c\sigma}\right],\end{aligned} \tag{14}$$

where we have introduced the notation $\lambda=d_{cv}E_0$.

In what follows, we will obtain exact solutions. It is noteworthy that, unlike the elementary case of a resonant transition between two one-electron levels $|v\rangle$ and $|c\rangle$, which occurs at the frequency $\Omega=\varepsilon_c-\varepsilon_v$,[14] we consider all allowed transitions between four two-electron states of the system. Therefore the resonant frequency is different from $\varepsilon_c-\varepsilon_v$ in the presence of finite Coulomb repulsion.

To solve the Schrödinger equation (13) with Hamiltonian (14), it is convenient to go to a representation with a time-independent Hamiltonian. To this end, we use the unitary transformation

$$\hat{U}(t)=\exp\left[-\frac{i\Omega t}{2}\sum_\sigma\,(\hat{a}^+_{c\sigma}\hat{a}_{c\sigma}-\hat{a}^+_{v\sigma}\hat{a}_{v\sigma})\right], \tag{15}$$

which was used in Ref. 16 to describe the interaction between an intense electromagnetic field and a semiconductor.

We replace the wave function $\Psi(t)$ with $\tilde{\Psi}(t)=\hat{U}^+(t)\Psi(t)$. Substituting

$$\Psi(t)=\hat{U}(t)\tilde{\Psi}(t) \tag{16}$$

into the Schrödinger equation (13) for $\Psi(t)$, and taking into account Eq. (15), we obtain the Schrödinger equation for $\tilde{\Psi}(t)$:

$$i\,\frac{\partial\tilde{\Psi}(t)}{\partial t}=\hat{\tilde{H}}\tilde{\Psi}(t), \tag{17}$$

with the Hamiltonian

$$\begin{aligned}\hat{\tilde{H}}=&\hat{U}^+(t)\hat{H}(t)\hat{U}(t)-i\hat{U}^+(t)\,\frac{\partial\hat{U}(t)}{\partial t}\\=&\sum_\sigma\left[\left(\varepsilon_v+\frac{\Omega}{2}\right)\hat{a}^+_{v\sigma}\hat{a}_{v\sigma}+\left(\varepsilon_c-\frac{\Omega}{2}\right)\hat{a}^+_{c\sigma}\hat{a}_{c\sigma}\right]\\&+U_v\hat{n}_{v\uparrow}\hat{n}_{v\downarrow}+U_c\hat{n}_{c\uparrow}\hat{n}_{c\downarrow}+\sum_\sigma\left(\frac{\lambda}{2}\,\hat{a}^+_{c\sigma}\hat{a}_{v\sigma}\right.\\&\left.+\frac{\lambda^*}{2}\,\hat{a}^+_{v\sigma}\hat{a}_{c\sigma}\right).\end{aligned} \tag{18}$$

This Hamiltonian $\hat{\tilde{H}}$ is independent of time; therefore the general solution of the time-dependent Schrödinger equation (17) for $0\leqslant t\leqslant T$ has the form

$$\tilde{\Psi}(t)=\sum_{i=1}^{4}\,B_i\,\exp(-i\tilde{E}_i t)|\,\tilde{i}\,\rangle, \tag{19}$$

where $|\,\tilde{i}\,\rangle$ and $\tilde{E}_i$ are the eigenstates and eigenenergies of the time-independent Schrödinger equation

$$\hat{\tilde{H}}|\,\tilde{i}\,\rangle=\tilde{E}_i|\,\tilde{i}\,\rangle \tag{20}$$

with Hamiltonian $\tilde{H}$ in the form given by Eq. (18). The coefficients B_i in Eq. (19) should be derived from the initial conditions (predetermined values of $A_i(0)$ in Eq. (11)) and satisfy the normalization condition

$$\sum_{i=1}^{4}\,|B_i|^2=1. \tag{21}$$

We seek solutions of Eq. (20) in the form

$$|\,\tilde{i}\,\rangle=\sum_{k=1}^{4}\,C_{ik}|k\rangle, \tag{22}$$

where the $|k\rangle$ are two-electron eigenstates defined by Eq. (10) for $t\leqslant 0$. Substituting Eq. (22) into Eq. (20), we obtain a set of equations that determines $\tilde{E}_i$ and C_{ik}:

$$\sum_{k=1}^{4} C_{ik}(\langle i|\hat{\tilde{H}}|k\rangle - \delta_{ik}\tilde{E}_i)=0, \tag{23}$$

where $i=1,2,3,4$, $\langle i|\hat{\tilde{H}}|k\rangle$ are the matrix elements of Hamiltonian (18) in terms of the basis states (10). The Hamiltonian matrix $\langle i|\hat{\tilde{H}}|k\rangle$ has the form

$$\begin{pmatrix} 2\varepsilon_v+U_v+\Omega & \lambda^*/2 & \lambda^*/2 & 0 \\ \lambda/2 & \varepsilon_v+\varepsilon_c & 0 & \lambda^*/2 \\ \lambda/2 & 0 & \varepsilon_v+\varepsilon_c & \lambda^*/2 \\ 0 & \lambda/2 & \lambda/2 & 2\varepsilon_c+U_c-\Omega \end{pmatrix}.$$

From Eqs. (19) and (22) we obtain

$$\tilde{\Psi}(t)=\sum_{i=1}^{4} D_i(t)|i\rangle, \tag{24}$$

where

$$D_i(t)=\sum_{k=1}^{4} B_k C_{ki}\exp(-i\tilde{E}_k t). \tag{25}$$

Since $\tilde{\Psi}(0)=\Psi(0)$ (see Eqs. (15) and (16)), we have $D_i(0)=A_i(0)$, where the coefficients $A_i(0)$ determine the state (11) for $t\leqslant 0$. Therefore, we obtain from Eq. (25) an equation to determine the coefficients B_i in terms of given $A_i(0)$:

$$A_i(0)=\sum_{k=1}^{4} B_k C_{ki},$$

whence

$$B_i=\sum_{k=1}^{4} A_k(0)C_{ki}^{-1}, \tag{26}$$

where C^{-1} is the matrix inverse of C. From Eqs. (25) and (26) we obtain

$$D_i(t)=\sum_{k=1}^{4}\sum_{l=1}^{4} A_l(0)C_{li}^{-1}C_{ki}\exp(-i\tilde{E}_k t). \tag{27}$$

Given Eq. (16) relating the function $\tilde{\Psi}(t)$ to $\Psi(t)$, and taking into account that the operator $\hat{U}(t)$ defined by Eq. (9) is unitary, we obtain an expression for the probability $p_i(t)$ for the transition to state $|i\rangle$:

$$p_i(t)=|D_i(t)|^2. \tag{28}$$

In the case under investigation, $A_1(0)=1$, $A_2(0)=A_3(0)=A_4(0)=0$ (at time $t=0$, all levels of the valence band are occupied and the levels of the conduction band are vacant; see Eq. (10)), expression (27) for $D_i(t)$ can be simplified somewhat. Nonetheless, the calculation of $D_i(t)$ still requires finding $\tilde{E}_i$ and C_{ik} from Eq. (23). This eigenvalue problem reduces to the solution of a cubic algebraic equation. The resulting expressions for $\tilde{E}_i$ and C_{ik} are cumbersome and difficult to analyze, so it is more convenient at this stage to solve Eq. (23) numerically. However, before discussing numerical calculations, we consider an important limiting case that admits of an analytic solution which is in excellent agreement with the numerical solution (obtained using the scheme described above) in the parameter range of interest.

Suppose that the Coulomb repulsion between electrons in the conduction band is infinite, $U_c=\infty$. The two-electron state $|4\rangle=|c\uparrow,c\downarrow\rangle$ (see Eq. (10)), whose energy is $E_4=2\varepsilon_c+U_c=\infty$, is then obviously always empty, i.e., the coefficient $A_4(t)$ in Eq. (11) is zero for all t. For the same reason, the matrix elements C_{i4} $(i=1-4)$ in Eq. (22) are also zero. Therefore, Eq. (23) can be solved by diagonalizing a 3×3 matrix, which can be done analytically. Thus, having calculated the eigenvalues $\tilde{E}_i$ and the matrix of eigenvectors C_{ik}, we obtain from Eqs. (27) and (28) the following expressions for the transition probabilities $p_i(t)$ from the state $\Psi(0)=|1\rangle=|v\uparrow,v\downarrow\rangle$ to the state $|i\rangle$:

$$\begin{aligned} p_1(t)&=1-\left(1-\frac{\delta^2}{4\omega_R^2}\right)\sin^2(\omega_R t),\\ p_2(t)&=\frac{1}{2}\left(1-\frac{\delta^2}{4\omega_R^2}\right)\sin^2(\omega_R t),\\ p_3(t)&=\frac{1}{2}\left(1-\frac{\delta^2}{4\omega_R^2}\right)\sin^2(\omega_R t),\\ p_4(t)&=0, \end{aligned} \tag{29}$$

where

$$\delta=\Omega-(\varepsilon_c-\varepsilon_v-U_v),\quad \omega_R=\frac{\sqrt{\delta^2+2|\lambda|^2}}{2}. \tag{30}$$

From Eq. (29) we derive the desired probability $p_{vc}(t)=p_2(t)+p_3(t)$ for the transition of one electron to the conduction band:

$$p_{vc}(t)=\left(1-\frac{\delta^2}{4\omega_R^2}\right)\sin^2(\omega_R t). \tag{31}$$

It follows from Eqs. (30) and (31) that $p_{vc}(T)=1$ at $\delta=0$ and

$$T=\frac{\pi}{2\omega_R}. \tag{32}$$

The equality $\delta=0$ has a simple physical meaning: the frequency Ω is equal to $\Omega_r=E_2-E_1=E_3-E_1$ (see Eq. (12) and Fig. 1), i.e., the difference between the energy of the pair of degenerate levels $|2\rangle$ and $|3\rangle$, and that of level $|1\rangle$. Only these two-electron states are resonant at $\delta=0$. Since $\varepsilon_c-\varepsilon_v\approx 1$ eV and $U_v+U_c\ll\varepsilon_c-\varepsilon_v$, we have $\Omega_r\approx 10^{15}\ \mathrm{s}^{-1}$. When the frequency is offset from resonance $(\delta\neq 0)$, the value of $p_{vc}(T)$ derived from Eq. (8) deviates from the maximum value by a quantity of order $|\delta|^2/|\lambda|^2$, i.e., the following inequality should hold:

$$|\delta|\ll|\lambda|. \tag{33}$$

It is clear that at $\delta=0$ our results should also be valid when U_c is finite (the lower boundary will be determined below), since the system can then only be in states $|1\rangle$, $|2\rangle$,

and $|3\rangle$ (if $U_c=\infty$, this statement is true at any δ). Moreover, if $\delta=0$, the sum U_v+U_c should be finite (i.e., U_c may be zero), which follows from the condition that the resonant frequencies Ω_r and $\Omega_r'=E_4-E_2=E_4-E_3$ (Fig. 1) for transitions between the levels $|1\rangle\leftrightarrow|2\rangle,|3\rangle$ and $|4\rangle\leftrightarrow|2\rangle,|3\rangle$, respectively, be different (at $U_v=U_c=0$ we have $p_{vc}\leqslant 1/2$; see Eq. (5)). At $\delta\neq 0$, the necessary condition for the validity of Eq. (31) for $p_{vc}(t)$ is obviously

$$|\delta|\ll U_v+U_c, \tag{34}$$

and the important parameter is the sum U_c+U_v, rather than U_v and U_c individually.

Inequalities (33) and (34) do not specify the ratio between $|\lambda|$ and U_v+U_c. Since for $|\delta|\ll|\lambda|$ we have $\omega_R\approx|\lambda|$, by generalizing condition (7), discussed in Sec. 2, we obtain $|\lambda|\ll U_v+U_c$, i.e., it follows from Eqs. (7) and (33) that

$$1/\tau,|\delta|\ll|\lambda|\ll U_v+U_c \tag{35}$$

(recall that τ is the radiative recombination time). Thus, if condition (35) is satisfied, the ''three-level approximation'' should apply to electronic transitions in the investigated four-level system under an applied an periodic perturbation.

We emphasize that conditions (35) can be satisfied in a real experiment. In fact, since $U_v+U_c\geqslant 1-10$ meV and $1/\tau\approx 10^{-3}-10^{-2}$ meV (see Sec. 2), we should have $|\lambda|\approx 0.1-1$ meV (the upper boundary of this interval can be increased by increasing U_v+U_c, i.e., decreasing the quantum dot size a, while the lower boundary is determined by the lifetime τ). Recalling that the parameter $|\lambda|$ is a product of the optical dipole matrix element $|d_{cv}|$ and the electric field amplitude E_0, and noting that $|d_{cv}|\approx ea$, where $a\approx 10-100$ nm, we obtain $E_0\approx 10^2$ V/cm, which can be easily realized in experiment. As concerns the condition $|\delta|\ll|\lambda|$, we have $|\delta|\leqslant 0.01$ meV, i.e., the frequency Ω of the external source should be accurate to within 10^{10} s^{-1}. Modern experimental methods make this possible.

We now compare the numerical solutions of Eqs. (23)–(28) to the analytic solution of Eqs. (30)–(32) in the ''three-level approximation.'' Let us take for definiteness $\varepsilon_c-\varepsilon_v\approx 1$ eV. Figure 2 shows p_{vc} as a function of t at $\delta=0$, $|\lambda|=0.01$ eV, and $U_v+U_c=0.1$ eV (the numerical calculations indicate that $p_{vc}(t)$ is only determined by the sum U_v+U_c, at least when $U_v+U_c\ll\varepsilon_c-\varepsilon_v$). Figure 2 demonstrates that the analytic solution is in good agreement with numerical calculations, as expected, since the selected values of the system parameters satisfy (34). The numerical calculations yield $p_{vc}^{\max}$ and T equal to 0.9935 and $1.4606\cdot 10^{-13}$ s, respectively, and the analytic solution yields 1 and $\pi/|\lambda|\sqrt{2}=1.4622\cdot 10^{-13}$ s, respectively.

Figures 3 and 4 illustrate the transition from the ''regime of noninteracting electrons'' ($U_v+U_c=0$, $p_{vc}^{\max}=1/2$, and $T=\pi/2|\lambda|=1.0339\cdot 10^{-13}$ s; see Eq. (5)) to the ''regime of strong Coulomb repulsion,'' for which $U_v+U_c\gg|\lambda|$ (Eq. (35)). This transition occurs when $U_v+U_c\approx\lambda$; when $U_v+U_c\geqslant 3\lambda$, the probability $p_{vc}^{\max}$ is larger than 0.95 and T is within 3.5% of its asymptotic value corresponding to $U_v+U_c=\infty$.

We now discuss the effect of an offset from resonance ($\delta\neq 0$) on $p_{vc}^{\max}$ and T. The corresponding curves are given

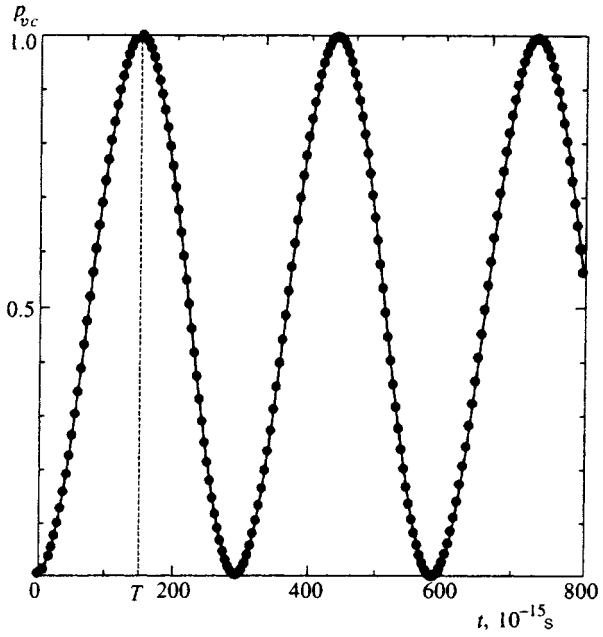

FIG. 2. Probability p_{vc} of one-electron transition from the valence band to the conduction band versus duration t of action of a periodic perturbation with frequency $\Omega=\varepsilon_c-\varepsilon_v-U_v$ ($\delta=0$); $\varepsilon_c-\varepsilon_v=1$ eV, $U_v+U_c=0.1$ eV, $|\lambda|=0.01$ eV. Numerical calculations with Eqs. (23)–(28) are plotted as dots, and the analytic solution defined by Eqs. (30)–(32) in the ''three-level approximation'' is shown by the solid line.

in Figs. 5 and 6 for $U_v+U_c=0.1$ eV and $|\lambda|=0.01$ eV, which satisfy the second inequality in (35). One can see that the analytic solution is in excellent agreement with the numerical calculations even when the first inequality in (35) is not satisfied. Here, of course, $p_{vc}^{\max}\approx 1$ only when $|\delta|\ll|\lambda|$.

Figures 7 and 8 show $p_{vc}^{\max}$ and T calculated in the immediate neighborhood of resonance for $U_v+U_c=0.01$ eV

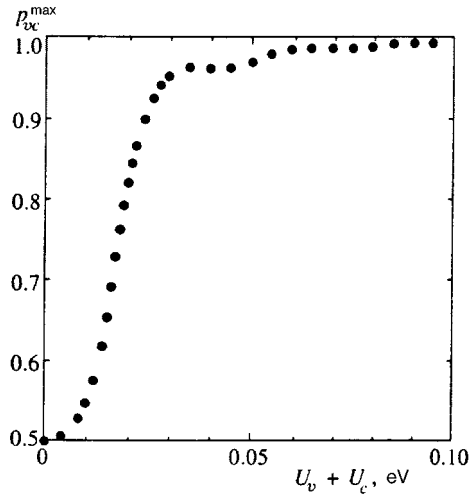

FIG. 3. Maximum probability $p_{vc}^{\max}$ of one-electron transition from the valence band to the conduction band due to a periodic perturbation with frequency $\Omega=\varepsilon_c-\varepsilon_v-U_v$ ($\delta=0$) as a function of the sum U_v+U_c of Coulomb repulsion energies in the valence band and conduction band; $\varepsilon_c-\varepsilon_v=1$ eV, $|\lambda|=0.01$ eV. The probability was calculated numerically using Eqs. (23)–(28).

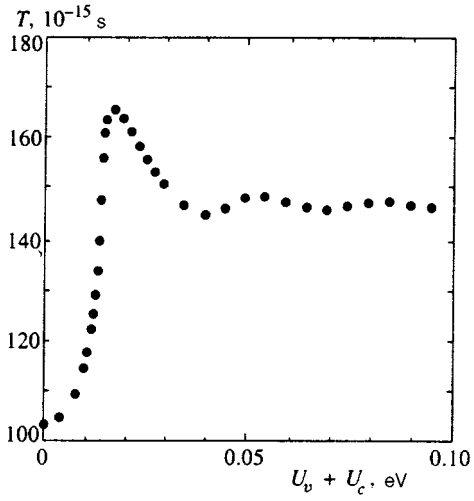

FIG. 4. Time T corresponding to the maximum probability $p_{vc}^{\max}$ of one-electron transition from the valence band to the conduction band due to a periodic perturbation with frequency $\Omega=\varepsilon_c-\varepsilon_v-U_v$ ($\delta=0$) as a function of the sum U_v+U_c of the Coulomb repulsion energies in the valence band and conduction band; $\varepsilon_c-\varepsilon_v=1$ eV, $|\lambda|=0.01$ eV. The time was calculated numerically using Eqs. (23)–(28).

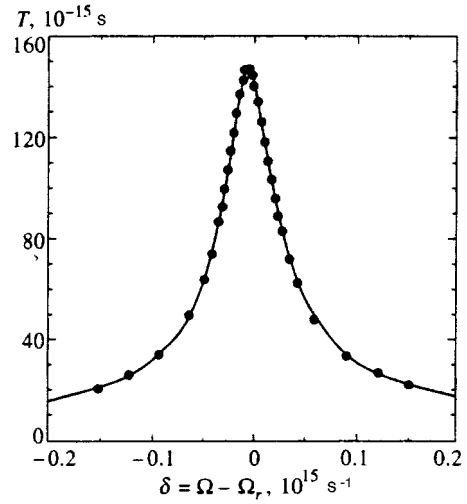

FIG. 6. Time T corresponding to the maximum probability $p_{vc}^{\max}$ of one-electron transition from the valence band to the conduction band due to a periodic perturbation with frequency Ω as a function of $\delta=\Omega-\Omega_r$, where $\Omega_r=\varepsilon_c-\varepsilon_v-U_v$ is the resonant frequency; $\varepsilon_c-\varepsilon_v=1$ eV, $U_v+U_c=0.1$ eV, $|\lambda|=0.01$ eV. The dots show numerical calculations using Eqs. (23)–(28). The analytic solution defined by Eqs. (30)–(32) in the ''three-level approximation'' is plotted as a solid line.

and $|\lambda|=0.0001$ eV. For $|\delta|\leqslant 10^{-10}$ s^{-1}, we have $p_{vc}^{\max}\geqslant 0.998$, i.e., the transition probability is essentially indistinguishable from unity. In this region, T varies between $1.4607\cdot 10^{-11}$ and $1.4622\cdot 10^{-11}$ s, i.e., by 0.1%.

4. EFFECT OF NONRESONANT COMPONENTS

In the resonant approximation we neglected several ''nonresonant'' harmonics of the applied periodic field (see Sec. 3). To assess their effect on the final result, we numerically solved the time-dependent Schrödinger equation (13) with Hamiltonian (9) using the explicit Runge–Kutta method of order 7(8) based on the equations of Dormand and Prince with automatic selection of the step size.[22] We calculated the coefficients $A_i(t)$ in Eq. (11) with initial conditions $A_i(0)=\delta_{i1}$, and determined the value of $p_{vc}(t)=|A_2(t)|^2+|A_3(t)|^2$. As follows from our calculations, the function $p_{vc}(t)$ determined in the nonresonant approximation in the parameter range of interest (see Eq. (35)) coincides with that found analytically in the ''three-level resonant approximation'' based on Eqs. (30)–(32) to within a fraction of a percent. It is noteworthy, however, that when nonresonant terms are taken into account, the probability $p_{vc}^{\max}$ peaks at a very small but finite offset δ. The good agreement between the numerical and analytic results sug-

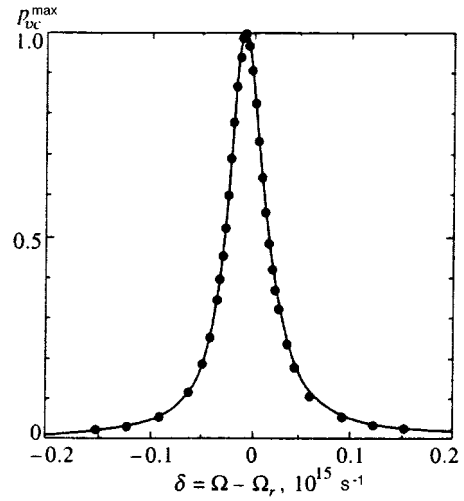

FIG. 5. Maximum probability $p_{vc}^{\max}$ of a one-electron transition from the valence band to the conduction band due to a periodic perturbation with frequency Ω as a function of $\delta=\Omega-\Omega_r$, where $\Omega_r=\varepsilon_c-\varepsilon_v-U_v$ is the resonant frequency; $\varepsilon_c-\varepsilon_v=1$ eV, $U_v+U_c=0.1$ eV, $|\lambda|=0.01$ eV. The dots show numerical calculations using Eqs. (23)–(28). The analytic solution of Eqs. (30)–(32) in the ''three-level approximation'' is plotted as a solid line.

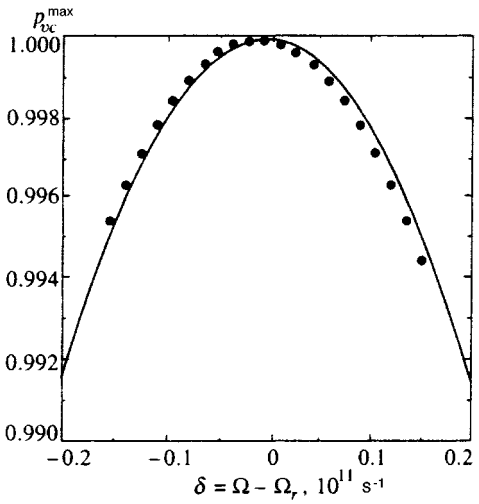

FIG. 7. Same as Fig. 5 with $U_v+U_c=0.01$ eV, $|\lambda|=0.0001$ eV.

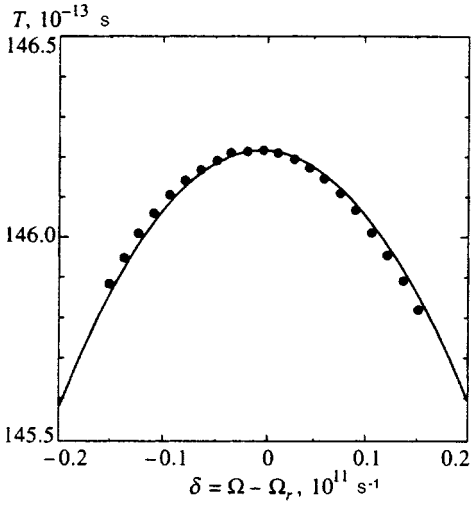

FIG. 8. Same as Fig. 6 with $U_v+U_c=0.01$ eV, $|\lambda|=0.0001$ eV.

gests that the nonresonant summands in the Hamiltonian (9) have little effect on system evolution. This result, however, was predictable for $|\lambda|\ll\Omega$.[14]

5. DISCUSSION OF RESULTS AND CONCLUSIONS

Thus, we have solved the problem of interaction between an hf electric field and a quantum dot with two size-quantized levels (one in the valence band and the other in the conduction band) and two electrons, taking into account Coulomb repulsion between the electrons. In the parameter range of interest, the problem has an easily interpretable analytic solution (30)–(32) in the so-called resonant three-level approximation (Sec. 3). We have shown that neither taking account of all two-electron levels of the quantum dot in the resonant approximation (Sec. 3) nor numerically solving the time-dependent Schrödinger equation with due account of ''nonresonant'' harmonics of the applied electric field (Sec. 4), have any perceptible effect on the time dependence of the energy-level populations.

With regard to the form of the model Hamiltonian (9), it does not include terms corresponding to the Coulomb interaction U_{vc} between two electrons in different levels $|v\rangle$ and $|c\rangle$ (if the valence band states are described in terms of holes, this interaction corresponds to excitonic effects). If the wave functions of the one-electron states $|v\rangle$ and $|c\rangle$ have different symmetry properties (as in GaAs/AlGaAs), the interaction energy U_{vc} should be significantly less than U_v and U_c. But even if it is not, it can be shown[18] that U_{vc} leads only to a shift in the resonant frequency, which becomes $\Omega_r=\varepsilon_c-\varepsilon_v-U_v+U_{vc}$, while the system dynamics remain unchanged. The frequency Ω_r of a specific quantum dot should be determined empirically.

Thus, we have shown that it is possible, in theory, to select the parameters of an external perturbation (Ω, $|\lambda|$, and T) and of a quantum dot ($U_v+U_c\approx e^2/\varepsilon a$) so that the probability of driving one electron from the valence band to the conduction band is extremely close to unity. Significantly, the time of this transition, $T=10^{-13}$–10^{-11} s, is several orders of magnitude shorter than the recombination time $\tau=10^{-10}$–10^{-9} s. Therefore, after the applied field is off (at $T\geqslant t$), the quantum dot is in a quasistationary excited state $|v\uparrow,c\downarrow\rangle$ or $|v\downarrow,c\uparrow\rangle$ with lifetime τ, after which the electron–hole pair will annihilate with generation of a one-photon wave packet $|1\rangle_f$ (Eq. (3)) with spectral distribution $f(\omega)$ centered at the frequency $\omega_0=(2\varepsilon_c+U_v)-(\varepsilon_c+\varepsilon_v)$, i.e., near the resonant frequency $\Omega_r=\varepsilon_c-\varepsilon_v-U_v$ of the external field applied to the quantum dot. The process of photon generation is certainly probabilistic, so we cannot give the exact time when the photon is emitted. An important point is that if the recombination is radiative, only one photon will be emitted. This source of one-photon states can be used in quantum systems for data transmission and processing, including quantum cryptosystems.

In conclusion, note that the estimates of the required parameters of the quantum dot and external perturbation provide evidence in favor of the feasibility of such a source of one-photon states. Moreover, if the external field frequency cannot be varied over a broad band, the source resonance condition can probably be achieved by varying the energy difference between the size-quantized levels in the valence and conduction bands via application of static electric and magnetic fields to the quantum dot.

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