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Light-Induced Electron Transfer Counter to an Electric Field Force in an Asymmetric Double Quantum Well

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LIGHT-INDUCED ELECTRON TRANSFER COUNTER TO AN ELECTRIC FIELD FORCE IN AN ASYMMETRIC DOUBLE QUANTUM WELL

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Electron transfer counter to an electric-field force is predicted for an asymmetrical double quantum well subjected to a dc bias in response to optical (far ir) excitation of an intersubband electronic transition. This transfer exhibits a resonance enhancement when the bias electric field aligns the excited levels in the wide and narrow wells. The transfer effect is driven by the quantum-mechanical delocalization caused by the coherent resonant tunneling which prevails over the electric force. The effect brings about photoinduced increase of the potential difference at the double well and a transient electric current opposite to the direction favored by the bias.

1. Introduction and qualitative description of effect

The aim of this Letter is to predict an effect which manifests itself as transfer of electrons counter to an electric-field force in response to the optical excitation of an intersubband transition in an asymmetric quantum well. A quantum well is a semiconductor heterostructure (see, e.g., [1]), whose potential confines electrons to a small region. Such confinement brings about quantum splitting of the electron energy bands into subbands separated by excitation energy on the order of $\hbar^2/m^*a^2$, where m^* is the electron effective mass and a is a confinement size (width of the well). In what follows, we will assume that the conduction band states are populated due to a modulation doping of the barrier regions and/or an incoherent optical excitation from the valence band, and consider purely electronic transitions between subbands of the conduction band [which are often called QWEST (Quantum Well Electronic (inter)Subband Transitions)].

To explain the essence of the effect, let us consider an asymmetric double quantum well with an electric field applied perpendicular to the well plane. The schematic of the confining potential and electron levels (subbands) is shown in Fig. 1 with $|1\rangle$ and $|2\rangle$ as the ground states in the narrow (N) and wide (W) wells, respectively. Let us assume that the electric field aligns the excited levels in the coupled wells, and denote as $|+\rangle$ and $|-\rangle$ the upper and lower components of the excited level doublet. The $|\pm\rangle$–state wave functions are extended over both the N and W wells due to resonant tunneling. In contrast, the lower levels are nor aligned, and the $|1\rangle$ state is basically localized in the N well and $|2\rangle$ in the W well. Since the subband splitting of the W well is smaller, the overall ground state is $|1\rangle$ in the N well (see Fig. 1). We assume both the electron density and temperature to be not very high, so that only the $|1\rangle$ state is considerably populated.

Suppose that ir light excites an intersubband transition in the N band, i.e. one of the transitions of the type $|1\rangle \to |\pm\rangle$ shown in Fig. 1 by a wavy arrow. The electron excited to either of the $|\pm\rangle$ states is quantum-mechanically delocalized over both the wells. Subsequent relaxation brings about electron transitions to the ground states $|1\rangle$ and $|2\rangle$ shown in Fig. 1 by dashed arrows. The transition rates are proportional to the probabilities for an electron to be localized in the corresponding wells and, for aligned levels, are on the same order of magnitude. Thus, with an appreciable probability, the electron comes to the state $|2\rangle$, which is mainly localized in the W well. Note that an electron lives in the $|2\rangle$ level, before undergoing transition back to $|1\rangle$, for a comparatively long time due to two reasons: first, the transition $|2\rangle \rightarrow |1\rangle$ is not resonantly enhanced, and second, the rate of tunneling between the coupled wells is smaller in the state $|2\rangle$ as compared to that for $|\pm\rangle$, since the tunneling probability is strongly reduced with decrease of the excitation energy.

Summarizing, a net result of the photoexcitation of the intersubband transition in the N well is a transfer of the electron from the N well to the W well in the direction of the potential increase (cf. Fig. 1), i.e. *against* the direction of of the field force. Indeed, the energy needed for such a transfer is taken from the exciting radiation. The effect is based on the quantum delocalization of electron prevailing over the field force. If the bias field is too strong, then the electron distribution in the excited states is shifted toward the N well and the effect may virtually disappear. Also, the dephasing, relaxation, destroying the quantum coherence, diminishes effect. The theory presented below addresses these factors.

The closest counterpart of the above described effect is the observation by Sauer, Thonke and Tsang [2] of photoinduced space-charge buildup due to asymmetric electron and hole tunneling in coupled quantum wells. The effect of [2] is similar to the present effect in regard to electron transfer against the electric-field force but, nevertheless, is essentially different in the following respects. First, there is no relaxation involved in charge buildup in [2], and, as a result, the electron buildup is *minimum* for the levels aligned, while in our case it is *maximum*. Also, for the aligned excited levels after switching off the optical excitation, the charge, which has been transferred between wells, disappears in a time on the order of the resonant tunneling time, while in our case the charge transferred is stable on this temporal scale. Second, the effect [2] is induced by interband transitions, and, therefore, the portion of the photon energy accumulated in the potential energy of a transferred electron is small, as distinct from the present effect based on intersubband transitions. Third, the charge transfer in [2] is based upon the difference in the tunneling time of the electrons in the conduction band and holes in the valence band, while no conduction-band holes participate and no such requirement is relevant for the present effect.

2. Quantitative description

We start the theory with the coherent tunneling regime, where the tunnelling amplitude τ is is much greater than the relaxation rates in the system, and there exists the doublet of the delocalized states $|\pm\rangle$. Note that at least partially coherent tunneling has been demonstrated experimentally [3, 4]. The maximum counter-field transfer effect occurs at low temperatures $T \ll \hbar^2/m^*a^2$. We assume that the light is resonant to one of the transitions $|1\rangle \to |\pm\rangle$ and, consequently, for an asymmetric double well, is not resonant to the transitions $|2\rangle \to |\pm\rangle$. Taking this into account, the rate equations describing the populations n_i of the states $|i\rangle$ have the form

$$\frac{\partial n_1}{\partial t} = -w_{\pm 1}n_1 + (\gamma_{1\pm} + w_{\pm 1})n_\pm + \gamma_{12}n_2 \quad , \qquad \frac{\partial n_2}{\partial t} = \gamma_{2\pm}n_\pm - \gamma_{12}n_2 \quad ,$$
$$\frac{\partial n_\pm}{\partial t} = w_{\pm 1}n_1 - (\gamma_{1\pm} + w_{\pm 1})n_\pm - \gamma_{2\pm}n_\pm \quad . \tag{1}$$

Here $\pm$ refer to either $|+\rangle$ or $|-\rangle$ states; $w_{\pm 1}$ is the rate of light absorption or stimulated emission for the radiative transition $|1\rangle \leftrightarrow |\pm\rangle$, i.e. $w_{\pm 1} = I\sigma_{\pm 1}$, where $\sigma_{\pm 1}$ is the corresponding cross section and I is the light intensity; and γ_{ji} is the rate of the relaxation transition $|i\rangle \to |j\rangle$, $i, j = 1, 2, +, -$. For simplicity, we do not include transitions between the doublet components $|+\rangle$ and $|-\rangle$, because these transitions essentially do not change the excitation kinetics (the electron transfer probability via each of these components is nearly the same at the resonance).

The stationary solution of Eq. (1) yields a population number n_2, which is the probability of electron transfer from the N to W well,

$$n_2 = \gamma_{2\pm}w_{\pm 1}\left[\gamma_{12}\gamma_\pm + w_{\pm 1}(2\gamma_{12} + \gamma_{2\pm})\right]^{-1} \quad , \tag{2}$$

where $\gamma_\pm \equiv \gamma_{2\pm} + \gamma_{1\pm}$. For the case of high intensities, $w_{\pm 1} \gg \gamma_{12}\gamma_\pm(2\gamma_{12} + \gamma_{2\pm})^{-1}$, one readily obtains from Eq. (2) the saturated transfer probability

$$n_2^{(s)} = (1 + 2\gamma_{12}/\gamma_{2\pm})^{-1} \quad . \tag{3}$$

For the opposite limiting case of nonsaturating light intensities, we assume the broad spectral band of the light, so that both transitions $|1\rangle \to |+\rangle$ and $|1\rangle \to |-\rangle$ occur. Note that in the nonsaturated case, Eq. (1) is valid irrespectively to the spectral selection, because only the transitions from the $|1\rangle$ state, as the overall ground state, and not the $|2\rangle$ state, are excited. An expression for the transfer quantum yield $\bar{Q}$ follows from Eq. (2),

$$\bar{Q} = \left(w_{+1}\gamma_{2+}/\gamma_{+} + w_{-1}\gamma_{2-}/\gamma_{-}\right)\left(w_{+1} + w_{-1}\right)^{-1} . \tag{4}$$

As one can see from Eqs. (1)-(4), the electron transfer kinetics is determined by the decay constants γ_{ji} and the rates $w_{\pm 1}$, which, in turn, depend on the wave function mixing between the individual wells. This mixing is described by the probabilities $P_{\pm}^{(N)}$ and $P_{\pm}^{(W)}$ for an electron in the mixed $|\pm\rangle$ state to be in the corresponding N or W well. To find γ_{ij}, we invoke a quantum-mechanical idea that the relaxation causes localization, and an electron localizes in the well in which it has experienced the relaxation. This assumption is valid if the nonresonant tunneling rate is small. From this we obtain

$$\gamma_{1\pm} = \gamma^{(N)} P_{\pm}^{(N)} \quad , \qquad \gamma_{2\pm} = \gamma^{(W)} P_{\pm}^{(W)} \quad , \tag{5}$$

where $\gamma^{(N)}$ and $\gamma^{(W)}$ are the decay rates for the excited states in the N and W wells.

Since the initial state $|1\rangle$ is mainly localized in the N well, the radiative transition couples this state only to the component of the $|\pm\rangle$ state localized in the N well, and, therefore, the transition probability $w_{\pm 1}$ is proportional to $P_{\pm}^{(N)}$. Assuming the spectral width of radiation to be much greater than the splitting between $|+\rangle$ and $|-\rangle$, we obtain

$$w_{+1}/w_{-1} = P_{+}^{(N)}/P_{-}^{(N)} . \tag{6}$$

From Eqs. (4)-(6), we get the transfer quantum yield in the form

$$\bar{Q} = \gamma^{(W)} \left[P_{+}^{(N)} P_{+}^{(W)}/\gamma_{+} + P_{-}^{(N)} P_{-}^{(W)}/\gamma_{-}\right] \left[P_{+}^{(N)} + P_{-}^{(N)}\right]^{-1} . \tag{7}$$

To determine the transfer probability (2), we need to estimate the interwell transition constant γ_{12}. Note that γ_{12} is proportional to the probability $P_{2}^{(N)}$ for an electron in the state $|2\rangle$, which is mainly localized in the W well, to be in the N well. Assuming the relaxation of all states in the well to occur with the same rate determined, e.g. by collisions, one can estimate $\gamma_{12} \approx \gamma^{(N)} P_{2}^{(N)}$, and obtain from Eq. (3) the saturated transfer probability

$$n_{2}^{(s)} = \left(1 + 2\gamma^{(N)} P_{2}^{(N)}/\gamma^{(W)} P_{\pm}^{(W)}\right)^{-1} . \tag{8}$$

3. Numerical results

We have numerically solved the Schrödinger equation (see [5] for details) for an electron in the potential shown in Fig. 1, the widths of the AW barrier/W well/WN barrier/N well/NB barrier equal to 100/19/8/14.5/100 nm, all the barriers being $Al_{0.1}Ga_{0.9}As$, which corresponds to the well depth $U_0 = 77.5$ meV.

The solution of the Schrödinger equation yields the aligning electric field $E_a = 6.4$ kV/cm; energies of the levels (meV): $E_1 = 4.7$, $E_2 = 15.9$, $E_- = 40.9$, and $E_+ = 43.9$; the tunneling amplitude $\tau = (E_+ - E_-)/2 = 1.5$ meV/$\hbar$; the dipole matrix elements: $d_{+1}/e = 2.4$ nm, $d_{-1}/e = -3.0$ nm with e as the elementary charge; and the localization probabilities: $P_1^{(N)} = 0.98$, $P_2^{(W)} = 0.99$, $P_-^{(N)} = 0.58$, $P_+^{(N)} = 0.42$ $[P_i^{(N)} + P_i^{(W)} = 1]$. From the last set of data we see that the state $|1\rangle$ is, in fact, localized in the N well and $|2\rangle$ in the W well, while the $|+\rangle$ and $|-\rangle$ states are almost evenly delocalized over both the wells, as assumed above.

The transition rate from the W to N well $\gamma_{12} \approx \gamma^{(\mathrm{N})} P_2^{(N)} = 0.01\gamma^{(\mathrm{N})}$ is very small with respect to $\gamma^{(\mathrm{N})}$, which ensures high saturated probability (8) $n_2^{(s)} = 0.99$, low optical excitation rates $w^{(s)}$ needed to achieve saturation of the $|2\rangle$ state, $w^{(s)} \sim \gamma_{12} \ll \gamma^{(\mathrm{N})}$, and comparatively long lifetime of the transferred charge after switching off the radiation $t_l = \gamma_{12}^{-1}$. In practical terms, the typical decay rate of the excited states $\gamma^{(\mathrm{N})} \sim 10^{12}\ \mathrm{s}^{-1} = 0.66$ meV/$\hbar$, which yields $t_l = 0.1$ ns. For the linewidth of ~ 2 meV characteristic of QWEST and the dipole elements given above, the saturation light intensity $I_s = (E_\pm - E_1)\gamma_{12}/\sigma_\pm$ can be estimated as $I_s \sim 60\ \mathrm{kW/cm^2}$.

Besides the data shown above, the computation provides escape rates γ_{Bi} from the $|i\rangle$ states in the double well to the B region (Fig. 1), i.e. in the direction of the potential drop. As expected, the largest of the obtained rates are those for the excited states: $\gamma_{B-} = 1.8\ \mu$eV/$\hbar$ and $\gamma_{B+} = 1.1\ \mu$eV/$\hbar$. These escape rates play the role of the rate constants for the parasitic process of the light-induced leak from the quantum well. However, comparing $\gamma_{B\pm}$ to the tunneling amplitude $\tau = 1.5$ meV/$\hbar$ and also to the rates $\gamma_{2\pm} \sim 0.3$ meV/$\hbar$ of the population of the $|2\rangle$ state from $|\pm\rangle$, we arrive at the conclusion that the escape current is negligibly small with respect to the interwell tunneling current, and can cause only a very small positive charging of the well system as a whole without affecting the counter-field electron transfer.

The transfer quantum yield $\bar{Q}$ as a function of the bias electric field E calculated from Eqs. (7) and (9) for a simplest case $\gamma^{(N)} = \gamma^{(W)}$ is shown in Fig. 2 with the solid line. As one can see, $\bar{Q}$ has a rather sharp resonance at the field $E = 6.4$ kV/cm, which exactly corresponds to alignment of the excited levels in the two coupled wells. The maximum value $\bar{Q}_{max} \approx 0.55$, and, as the computations show, essentially does not depend upon the barrier width, which is a consequence of the coherency of the tunneling. The high magnitude of $\bar{Q}_{max}$ and the localization probabilities $P_{\pm}^{(W)}$ and $P_{\pm}^{(N)}$ being equal to approximately 50% clearly demonstrate that, for the aligning field, the quantum delocalization prevails over the field force.

4. Analytical results and discussion

To find an analytical expression for the transfer quantum yield, we, following [1], solve the Schrödinger equation in the restricted basis of the two non-overlapping excited states in the wells, with the nondiagonal matrix element of Hamiltonian equal to τ (τ coincides with the transfer integral t in the notation of [1]). Such approach is valid for a not very high bias (cf. above). This gives the localization probabilities in the familiar form

$$P_{+}^{(N)} = P_{-}^{(W)} = 1-P_{+}^{(W)} = 1-P_{-}^{(N)} = 4\,|\tau|^2 \left\{ \left[\delta + \left(\delta^2 + 4\,|\tau|^2 \right)^{1/2} \right]^2 + 4\,|\tau|^2 \right\}^{-1} , \quad (9)$$

with δ as the frequency mismatch between the excited states in the coupled wells. From Eqs. (5)-(7) and (9), we obtain

$$\bar{Q} = |\tau|^2 \left(\gamma^{(N)} + \gamma^{(W)} \right) \gamma^{(W)} \left\{ |\tau|^2 \left(\gamma^{(N)} + \gamma^{(W)} \right)^2 + \gamma^{(N)} \gamma^{(W)} \delta^2 \right\}^{-1} . \quad (10)$$

For the system considered above, the numerical calculation gives $\delta = 2.6(E - 6.4$ kV/cm) meV/$\hbar$. With this and for $\gamma^{(W)} = \gamma^{(N)}$, Eq. (10) predicts the dependence $\bar{Q}(E)$ shown in Fig. 2 with the dashed line. Both in shape and magnitude, it agrees with the result of the complete solution of the Schrödinger equation (solid line).

The maximum quantum yield $\bar{Q}_{max}$, as given by Eq. (10) for $\delta = 0$, is

$$\bar{Q}_{max} = \left(1 + \gamma^{(N)} / \gamma^{(W)} \right)^{-1} . \quad (11)$$

This expression follows also directly from the general expression (7) under the condition of the maximum electron delocalization $P_{\pm}^{(W)} = P_{\pm}^{(N)} = 1/2$. For equal decay rates in the two wells, $\gamma^{(N)} = \gamma^{(W)}$, we have $\bar{Q}_{max} = 1/2$; for heavily doped wide well it is possible to obtain $\gamma^{(W)} \gg \gamma^{(N)}$, in which case $\bar{Q}_{max}$ approaches unity.

To briefly discuss, the results presented above indicate that the transfer of electrons against the electric field is quantum-mechanical effect based on the electron delocalization over the resonant states prevailing over the field force. The high quantum yield (11) $\bar{Q}_{max} = 0.5-1$, independent from the barrier thickness, is a consequence of the coherency of the transfer. The phase relaxation would destroy quantum-mechanical coherence and diminish the transfer. To explicitly demonstrate this, we invoke an expression for $\bar{Q}$, which we have obtained in a density matrix approach (will be published elsewhere) in the case of strong dephasing, $\Gamma_{WN} \gg \tau$, where Γ_{WN} is the polarization relaxation rate for the interwell tunneling. This expression has the form

$$\bar{Q} = 2\left|\tau\right|^2 \gamma^{(W)}\Gamma_{WN}\left[\gamma^{(N)}\gamma^{(W)}\left(\Gamma_{WN}^2+\delta^2\right)+2\left|\tau\right|^2\Gamma_{WN}\left(\gamma^{(N)}+\gamma^{(W)}\right)\right]^{-1} \quad . \qquad (12)$$

In contrast to Eq. (11), the quantum yield $\bar{Q} \to 0$ for $\Gamma_{WN} \to \infty$, or $|\tau| \to 0$. In a realistic case, $\tau \sim \Gamma_{NW}$, and, consequently, $\bar{Q}_{max} \sim 1$.

5. Possible experiments

Let us discuss possible experimental observation. The counter-field transfer of the electrons can be detected optically by monitoring changes of the intersubband absorption in the double well. The electrical detection of the transfer is also possible. We believe that the most reliable is the detection based on the capacitance coupling of the well to an external circuit. Such coupling is achievable even with the thick barriers AW and NB, thus excluding photoinduced leakage from the N well to B region, as discussed above.

For instance, the regions A and B (Fig. 1) containing a dense electron gas may play the role of the capacitor plates with which external conductors are in contact. For the regime of zero current in the external circuit, the counter-field transfer of electrons induced by switching on of the light brings about an *increase* of the potential difference U_{AB} by the amount

$$\Delta U_{AB} = 4\pi e\rho n_2 \Delta x/\varepsilon \quad , \qquad \Delta x \equiv \int_{-\infty}^{\infty}\left[\left|\Psi_1(x)\right|^2 - \left|\Psi_2(x)\right|^2\right] x\,\mathrm{d}x \quad . \qquad (13)$$

where ε is the mean dielectric constant of the well material. ρ is two-dimensional density of the electron gas in the well. Ψ_i is the wave function of the $|i\rangle$ state. and $\Delta x = 24$ nm for the example considered. In contrast to Eq. (13), if the photocurrent inside the well were directed along the potential drop. then U_{AB} would decrease.

For the applicability of the theory presented above, the potential increase ΔU_{AB} should be small which can always be achieved by reducing the electron density ρ. However, we should mention qualitative effects of the potential increase which. via changing the electric field inside the well. affects the photoexcitation and electron transfer. This is a feedback which can produce enhanced nonlinear optical responses, similar to ones observed in [6] for the interband transitions. and, possibly, an intrinsic optical bistability. We shall address these effects elsewhere.

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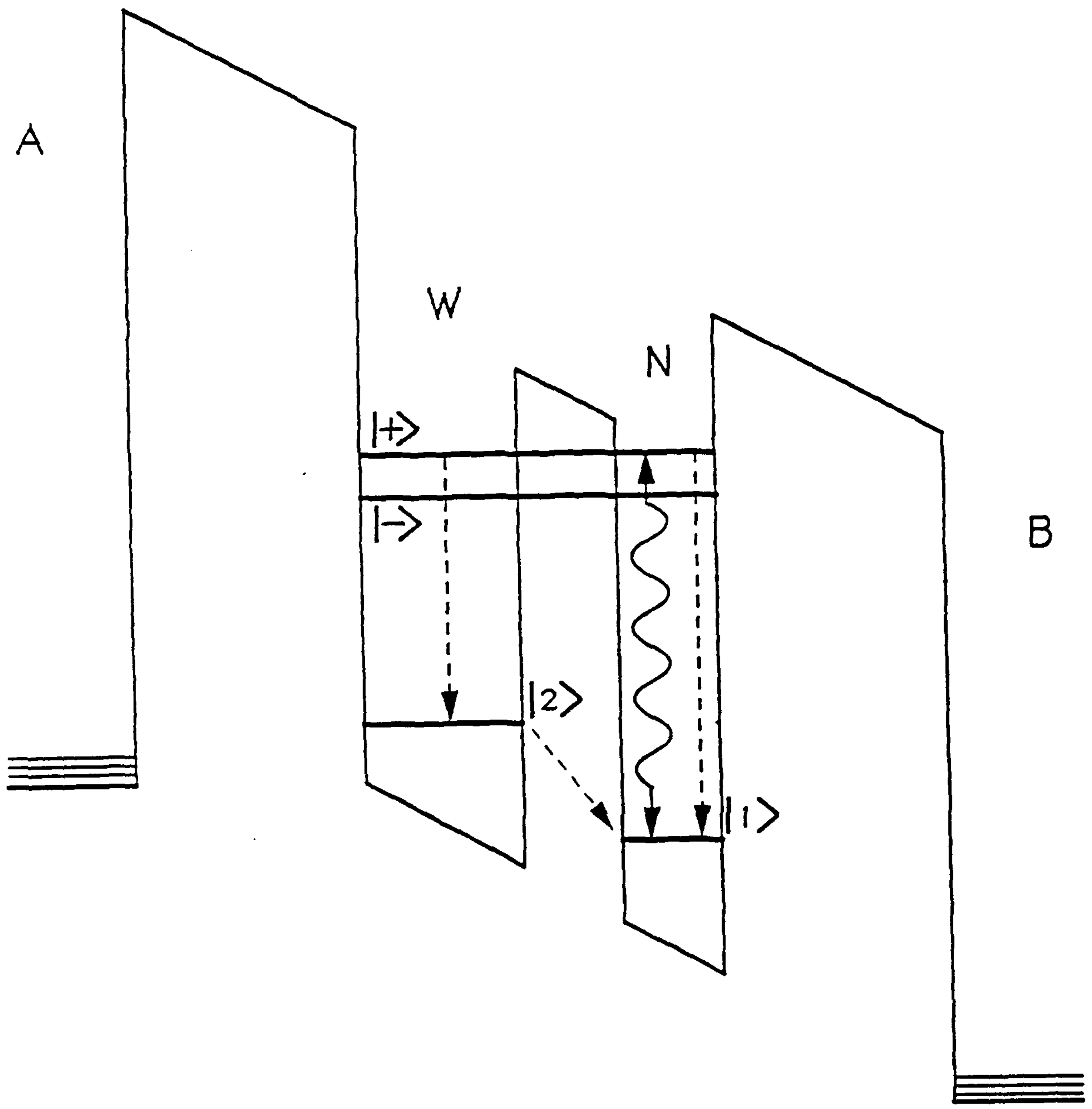
CAPTIONS TO FIGURES

Fig. 1. Coupled wide (W) and narrow (N) quantum wells subjected to a dc bias. Schematic of the confining potential, energy levels, and radiative (a wavy arrow) and nonradiative (dashed arrows) transitions. The regions A and B containing a dense electron gas serve as electrodes for the capacitance coupling of the double well to an external circuit. The insulating barriers AW and NB are supposed to be thick and high enough to exclude considerable tunneling through them (see the text).

Fig. 2. Quantum yield $\bar{Q}$ of the electron transfer counter to the field force as a function of the electric field E applied to the double well. Obtained by numerical calculation according to Eq. (7) (solid line) and from the analytical formula (10) for $\tau = 1.5$ meV$/\hbar$ (dashed line).



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