OFFICE OF NAVAL RESEARCH

Grant N00014-90-J-1193

TECHNICAL REPORT No. 91

Photoinduced Electron Transfer Counter to the Bias Field in Coupled Quantum Wells

by

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Prepared for publication

in

Photo-Induced Space Charge Effects in Semiconductors: Photoconductivity, Spectroscopy and Electro-optics
Edited by K. W. Goossen, N. M. Haegel and D. D. Nolte
Materials Research Society Symposium Proceedings, Volume 261-Q2

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August 1992

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE	3. REPORT TYPE AND DATES COVERED
	August 1992	Interim

4. TITLE AND SUBTITLE

Photoinduced Electron Transfer Counter to the Bias Field in Coupled Quantum Wells

5. FUNDING NUMBERS

Grant
N00014-90-J-1193

6. AUTHOR(S)

Mark I. Stockman, Leonid S. Muratov, Lakshmi N. Pandey and Thomas F. George

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)

Departments of Chemistry and Physics
Washington State University

8. PERFORMING ORGANIZATION REPORT NUMBER

WSU/92/91

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)

Office of Naval Research
800 N. Quincy Street
Arlington, Virginia 22217

10. SPONSORING/MONITORING AGENCY REPORT NUMBER

11. SUPPLEMENTARY NOTES

Prepared for publication in Photo-Induced Space Charge Effects in Semiconductors: Photoconductivity, Spectroscopy and Electro-optics (Materials Research Society Symposium Proceedings, Volume 261-Q2)

12a. DISTRIBUTION/AVAILABILITY STATEMENT

Approved for public release; distribution unlimited

12b. DISTRIBUTION CODE

13. ABSTRACT (Maximum 200 words)

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14. SUBJECT TERMS

COUPLED QUANTUM WELLS
BIAS FIELD
PHOTOINDUCED ELECTRON TRANSFER
TUNNELING
COHERENT
DENSITY MATRIX

15. NUMBER OF PAGES

7

16. PRICE CODE

NTIS

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
Unclassified	Unclassified	Unclassified	

Photo-Induced Space Charge Effects in Semiconductors: Photoconductivity, Spectroscopy and Electro-optics
Edited by K. W. Goossen, N. M. Haegel and D. D. Nolte
Materials Research Society Symposium Proceedings, Volume 261-Q2

PHOTOINDUCED ELECTRON TRANSFER COUNTER TO THE BIAS FIELD IN COUPLED QUANTUM WELLS

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ABSTRACT

Optical excitation of electrons in an asymmetric double quantum well is theoretically examined. The well is biased to align the excited levels and permit resonant electron tunneling. Emphasis is made on the photoinduced transfer of electrons counter to the bias electric field force. A density-matrix approach is developed to describe optical excitations in the presence of an arbitrary dephasing. The excitation profiles obtained for cases of different dephasing reveal the full range of tunneling coupling between the wells from completely coherent to incoherent (stepwise).

I. INTRODUCTION

The aim of this paper is to consider theoretically processes of optical excitation and electron transfer in an asymmetric double quantum well, i.e., in a system consisting of two different quantum wells coupled by electron tunneling. We will concentrate on intersubband electronic transitions, which are excited by far ir radiation and consider both optically linear and nonlinear effects. We will focus on the effect of the light-induced transfer of electrons from one well to the other one in the double quantum well. It is important that in biased quantum wells such a transfer can occur against the electric field force and with high quantum yield (up to 0.5).

We will assume that the conduction band states in the quantum well 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. We will also assume the electron density to be small enough to exclude excitonic and other many-body effects.

Much work has been done on the electronic, optical and kinetic properties of semiconductor double quantum wells (see, e.g., Ref. 1 and references cited therein, and also recently published papers[2-15], which are relevant for the present work). A fundamental phenomenon, which is a subject of the study, is resonant tunneling between the quantum wells. A distinctive feature of this phenomenon is a considerable enhancement of the tunneling probability if the energies of the donor and acceptor levels are close enough. To describe theoretically this phenomenon or interpret experimental results, most of the above cited works rely on the use of the Schrödinger equation. In this approach[1], the wave functions of resonant levels in the wells are mixed due to tunneling, and these states repulse forming a doublet separated by the energy $2|\tau|$, where τ is the tunneling amplitude. The tunneling is described by the delocalization of the electron wave function. Such tunneling is often called coherent, and we will follow this terminology. It is well understood (see, e.g., Refs. 3, 4 and 7) that relaxation destroys coherence, and makes tunneling incoherent (stepwise). When the relaxation rate Γ becomes on order of the tunneling amplitude τ or greater, the incoherent (stepwise) tunneling takes place between non-mixed states.

To describe a general case of an arbitrary relaxation we will use the density-matrix technique, which allows one to fully take into account the relaxation, including the dephasing contribution to the polarization-relaxation rate. Such a contribution, which is usually neglected, may be important.

To explain the essence of the effect of electron counterfield transfer[16], 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(a) with $|1\rangle$ and $|2\rangle$ as the ground states in the narrow (N) and wide (W) wells, respectively. The excited state in the N well is $|3\rangle$, and in the W well $|4\rangle$. Let us assume that the electric field aligns the excited levels $|3\rangle$ and $|4\rangle$, so that tunneling from one excited level to the other one is resonantly enhanced.

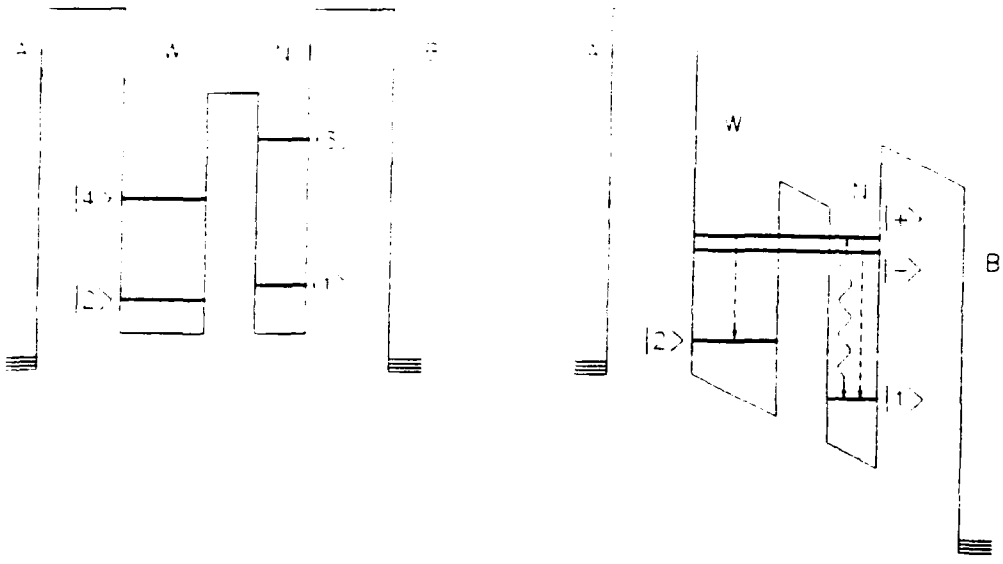

Fig. 1. Coupled wide (W) and narrow (N) quantum wells (a - in the absence of bias, b - in the presence, where the excited levels are aligned). Schematic of the confining potential, energy levels, and radiative (wavy arrow) and nonradiative (dashed arrows) transitions. The regions A and B containing a 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).

Qualitatively, the electron transfer effect is most pronounced in the coherent-tunneling case, where the aligned excited states form a doublet, the upper and lower components of which we denote as $|+\rangle$ and $|-\rangle$. The $|\pm\rangle$-state wave functions are delocalized over both the N and W wells due to resonant tunneling. In contrast, the lower levels are not 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(b)]. We assume both the electron density and the 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 well, i.e. one of the transitions of the type $|1\rangle \rightarrow |\pm\rangle$ shown in Fig. 1(b) by a wavy arrow. Since the splitting of the levels in the N well is assumed to be considerably greater, the radiation does not excite a transition in the W well. 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(b) 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.

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 [see Fig. 1(b)], i.e. *against* the direction of the field force. Indeed, the energy needed for such a transfer is taken from the exciting radiation. Note that if the transition in the wide well is excited, the electron transfer would occur in the direction of the field force.

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 Ref. 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 Ref. 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 Ref. 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.

II. EXCITATION AND ELECTRON TRANSFER IN THE DENSITY-MATRIX FORMALISM

A. Equations for density matrix

Below we present a general theory based on the density-matrix approach, which allows one to describe the full range of the interwell tunneling regimes from completely coherent in the case of small polarization-relaxation rate to the opposite case of completely stepwise for strong polarization relaxation. The simplifying feature of our approach is the use of the tight-binding model in the restricted basis of the states in isolated wells $|1\rangle, |2\rangle, |3\rangle, |4\rangle$, and of the relaxation-constant model for the relaxation term in the equation of motion for the density matrix (see below).

We start with the Hamiltonian of the system in the form $H = \sum_i \varepsilon_i a_i^\dagger a_i + \sum_{ij} V_{ij} a_i^\dagger a_j$, where ε_i are the subband energies, and $a^\dagger$ and a are the electron creation and annihilation operators, with $i, j = 1, 2, 3, 4$. The one-electron operator V describes interaction with the electromagnetic field and electron interwell tunneling, and its independent nonzero elements are $V_{31} = -\mathbf{d}_{31}\left(\mathbf{E}e^{-i\Omega t} + \text{c.c.}\right)$, $V_{43} = \tau$. The one-electron density matrix r is defined as $r_{ij} = \left\langle a_j^\dagger a_i \right\rangle$. Its diagonal matrix elements are the population probabilities $n_i \equiv r_{ii}$.

The equation of motion for r can be obtained in the usual way by commuting the pair operator $a_j^\dagger a_i$ with the Hamiltonian and adding the relaxation term. This has the well-known form (we use the system of units in which $\hbar = 1$):

$$\frac{\partial r}{\partial t} = i[r, \varepsilon + V] - R \quad . \tag{1}$$

where the one-electron energy operator ε is defined as $\langle i|\varepsilon|j\rangle = \varepsilon_i \delta_{ij}$, and R is the relaxation operator. In the low-temperature case, i.e. for neglect of thermal activation, the diagonal part of R describes spontaneous decays from higher- to lower-lying levels, and in the the model of relaxation constants has the form

$$R_{ii} = n_i \sum_{j\,(j<i)} \gamma_{ji} - \sum_{j\,(j>i)} \gamma_{ij} n_j \quad . \tag{2}$$

where γ_{ij} is the rate constant for spontaneous decay $|j\rangle \rightarrow |i\rangle$.

In what follows, we will neglect direct relaxation transitions that involve nonresonant interwell tunneling, $|3\rangle \rightarrow |2\rangle$ and $|4\rangle \rightarrow |1\rangle$, on the grounds of the small probability of nonresonant tunneling with respect to the resonant one. The rates of the above processes are negligible with respect to the rates of the collateral two-step processes involving the resonant tunneling, $|3\rangle \rightarrow |4\rangle \rightarrow |2\rangle$ and $|4\rangle \rightarrow |3\rangle \rightarrow |1\rangle$, which will be taken into account. However, the nonresonant-tunneling process $|2\rangle \rightarrow |1\rangle$ should be included despite its small rate, because there is no resonant process to compete with it. Thus, only the following decay rate constants should be taken into consideration: $\gamma_3 \equiv \gamma_{13}$, $\gamma_4 \equiv \gamma_{24}$, $\gamma_2 \equiv \gamma_{12}$.

The nondiagonal part of R describes the polarization relaxation and in the model under consideration is given by

$$R_{ij} = \Gamma_{ij} r_{ij} \quad , \quad \Gamma_{ij} = \frac{1}{2}(\gamma_i + \gamma_j) + \bar{\Gamma}_{ij} \quad , \tag{3}$$

where $\bar{\Gamma}_{ij} \geq 0$ is the pure dephasing term.

Below we adopt the resonant approximation, which is also synonymously called the Rotating-Wave Approximation (RWA). Applicability of this approximation is well established for optical that are fields not very strong. Technically, the RWA is equivalent to neglecting multiple harmonics of the light frequency Ω in the equations. Doing so, we can explicitly determine the temporal dependence of the polarizations,

$$\rho_{13} = \bar{\rho}_{13} \exp(\imath\Omega t) \quad , \quad \rho_{14} = \bar{\rho}_{14} \exp(\imath\Omega t) \quad , \quad \rho_{43} = \bar{\rho}_{43} \quad , \tag{4}$$

where $\bar{\rho}_{13}$, $\bar{\rho}_{14}$, and $\bar{\rho}_{43}$ are slowly varying amplitudes. From Eq. (1) we find the system of equations with constant coefficients for the density-matrix elements

$$\begin{aligned}
&\frac{\partial n_1}{\partial t} = -2\mathbf{Im}(\bar{\rho}_{13} G) + \gamma_2 n_2 + \gamma_3 n_3 \quad , && \frac{\partial n_2}{\partial t} = -\gamma_2 n_2 + \gamma_4 n_4 \quad , \\
&\frac{\partial n_3}{\partial t} = 2\mathbf{Im}(\bar{\rho}_{13} G) - \gamma_3 n_3 + 2\mathbf{Im}(\bar{\rho}_{43} \tau^*) \quad , && \frac{\partial n_4}{\partial t} = -2\mathbf{Im}(\bar{\rho}_{43} \tau^*) - \gamma_4 n_4 \quad , \\
&\frac{\partial \bar{\rho}_{13}}{\partial t} = \imath G^* (n_1 - n_3) + \imath\tau \bar{\rho}_{14} - g_{13} \bar{\rho}_{13} \quad , && \frac{\partial \bar{\rho}_{14}}{\partial t} = -\imath G^* \bar{\rho}^*_{43} + \imath\tau^* \bar{\rho}_{13} - g_{14} \bar{\rho}_{14} \quad , \\
&\frac{\partial \bar{\rho}_{43}}{\partial t} = \imath\tau (n_4 - n_3) + \imath G^* \bar{\rho}^*_{14} - g_{43} \bar{\rho}_{43} \quad ,
\end{aligned} \tag{5}$$

where the notations

$$g_{13} \equiv \Gamma_{13} + \imath(\Omega - \varepsilon_{31}) \quad , \quad g_{14} \equiv \Gamma_{14} + \imath(\Omega - \varepsilon_{41}) \quad , \quad g_{43} = \Gamma_{43} + \imath\varepsilon_{43} \tag{6}$$

are introduced, $G = -\mathbf{dE}$, and $\varepsilon_{ij} = \varepsilon_i - \varepsilon_j$ are the intersubband transition frequencies.

B. Stationary solutions

The stationary solution of Eq. (5) can be obtained in a straightforward manner. The expressions for the population probabilities of the excited levels are

$$\begin{aligned}
n_3 = &\Big[c(b+\gamma_4) - a(a+\gamma_4)\Big] \Big\{ c\left[b(3+\gamma_4/\gamma_2) + 2\gamma_4\right] + \\
&\gamma_3\gamma_4 + b(\gamma_3+\gamma_4) + a\left[\gamma_3(1+\gamma_4/\gamma_2) - a(3+\gamma_4/\gamma_2) - 3\gamma_4\right] \Big\}^{-1} \quad , \\
n_4 = &\Big[a(\gamma_3 - a) + cb\Big] \Big\{ c\left[b(3+\gamma_4/\gamma_2) + 2\gamma_4\right] + \\
&\gamma_3\gamma_4 + b(\gamma_3+\gamma_4) + a\left[\gamma_3(1+\gamma_4/\gamma_2) - a(3+\gamma_4/\gamma_2) - 3\gamma_4\right] \Big\}^{-1} \quad ,
\end{aligned} \tag{7}$$

where the notations

$$\begin{aligned}
a \equiv 2|\tau|^2 |G|^2 \mathbf{Re}\,(f^{-1}) \quad , \quad b \equiv 2|\tau|^2 \mathbf{Re}\left[(g_{13} g_{14} + |\tau|^2) f^{-1}\right] \quad , \\
c \equiv 2|G|^2 \mathbf{Re}\left[(g^*_{43} g_{14} + |G|^2) f^{-1}\right] \quad , \quad f \equiv g^*_{43}(g_{13} g_{14} + |\tau|^2) + g_{13}|G|^2
\end{aligned} \tag{8}$$

are introduced. As follows from from the second equation of the Eq. (5), the probability n_2 of the electron transfer is simply related to n_4 by

$$n_2 = \gamma_4 \gamma_2^{-1} n_4 \quad . \tag{9}$$

Note that he expressions (7) are exact.

C. Numerical illustrations

The effect of the polarization relaxation on the electron excitation and transfer can be traced in Fig. 2. We can see in Fig. 2(a) that in the case of a weak dephasing ($\Gamma_{13} = \Gamma_{14} = 0.1$ meV), the excitation contours are two almost separate peaks, which, as can easily be verified, are positioned at the transitions frequencies of the split doublet levels. The asymmetry of the excitation contour is due to nonzero level mismatch ϵ_{43}.

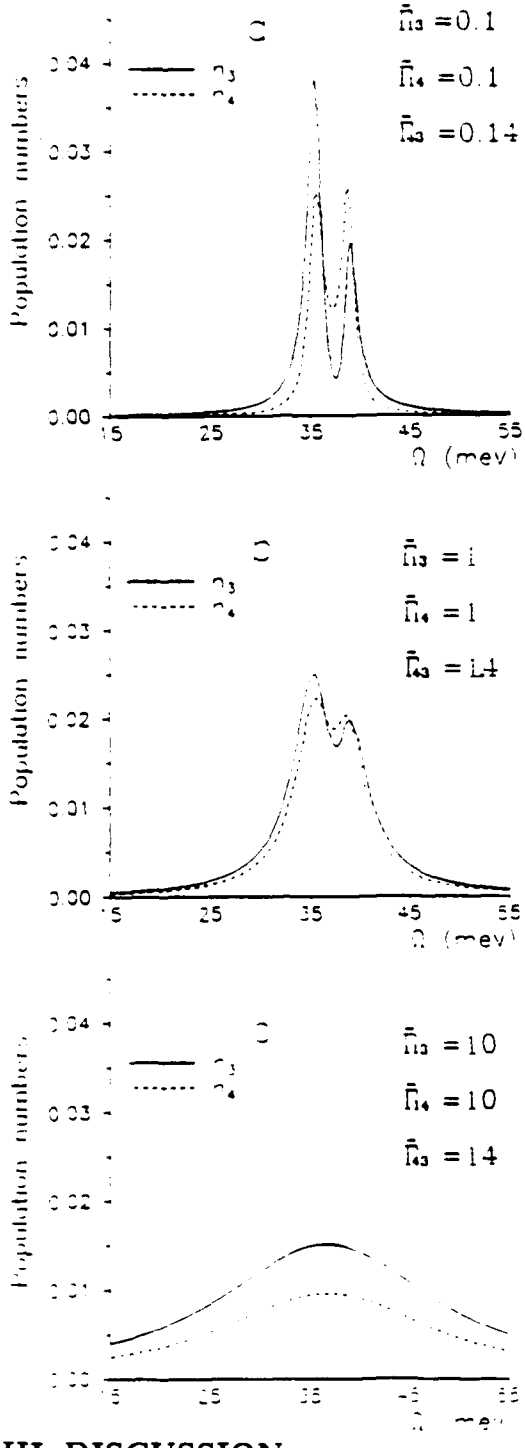

Fig. 2. Population numbers n_3 (solid line) and and n_4 (dashed line) of the excited levels as functions of the exciting light frequency Ω (meV) for the dephasing constants shown in the graphs. The dephasing relaxations in the two coupled wells are assumed equal. $\bar{\Gamma}_{13} = \bar{\Gamma}_{14}$. The other relevant parameters are $|G| = 0.2$ meV, $\gamma_3 = \gamma_4 = 0.2$ meV, $\Gamma_2 = 0.006$ meV, and $\epsilon_{43} = 1$ meV.

We emphasize that near the peak maxima we have $n_4 \sim n_3$, which means that the electron transfer counter to the electric field force occurs with a high probability, an electron is excited by the light to the $|3\rangle$ state, but appears with close or even greater probability in the $|4\rangle$ state localized in the other well. Using Eq. (9), we can make sure that in the spectral maxima $n_2 \approx 1$, i.e. the populations are saturated. At the same time, the parameter that governs the polarization saturation, $|G|^2/\Gamma_{13}^2 \ll 1$. This means, in particular, an absence of field broadening and a low probability of excitation into the continuum.

With an increase of the dephasing rate [see Fig. 2(b,c)], the spectral peaks are broadened and overlap. However, the population number n_4 and, consequently, the transfer probability n_2 (9) do not considerably diminish. This shows that the counterfield transfer effect persists even for relatively strong dephasing. For $\Gamma_{13} = \Gamma_{14} = 10$ meV, the doublet structure is completely absent, and the absorption contour is symmetrical and centered at the frequency ϵ_{31} of the transition in the isolated narrow well [Fig. 2(c)]. This means that the electron is first excited within the N well and then tunnels into the W well, i.e. the tunneling is incoherent..

III. DISCUSSION

In the stationary regime, closed analytical expressions (7) and (9) for the populations n_3 and n_4 of the excited states, and for the counterfield-transfer probability n_2 are obtained. The full range of the transition from a nearly coherent-tunneling regime to an almost incoherent one can be traced in Fig. 2. For a weak polarization relaxation, the excitation profiles reveal a two-peak structure typical for the coherent tunneling. In this case, the optical wave plays the role of a probe field exciting the system to the doublet

states $|\pm\rangle$. As the dephasing increases, the stationary states $|\pm\rangle$ are no longer a good zeroth-order approximation, and the interwell coupling becomes incoherent (stepwise): the first step is the excitation from the state $|1\rangle$ to $|3\rangle$ of the W well, followed by the second step of the tunneling into the N well. In qualitative agreement with the above picture, with an increase of the dephasing, the double-peak structure disappears, replaced by a single peak centered at the transition frequency ϵ_{31}. If the dephasing in the two coupled wells is increased in the same proportion, the excitation profiles are broadened [see Fig. 2(c)].

Let us discuss possible experimental observation of the counterfield electron transfer. This effect can be detected optically by monitoring changes of the intersubband absorption in the double well: as the transfer proceeds, the absorption band shifts to a lower frequency by the amount ϵ_{21}. Electrical detection of the transfer is also possible. In this case, external conductors should be in contact with the regions A and B in Fig. 1. However, achieving the regime of a stationary current in the external circuit is problematic, because in this case the barriers AW and NB should be penetrable for electrons. If so, the optical excitation, apart from bringing about the counterfield electron transfer, would also increase the rate of the electron escape from the N well to the B region, i.e. in the direction favored by the bias. Thus the counterfield transfer may be completely masked by this leak current. We believe that the most reliable observation is the detection of the counterfield transfer 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 the B region discussed above.

ACKNOWLEDGMENTS

The authors are grateful to B. D. McCombe for useful discussions. We acknowledge use of facilities of the Pittsburgh Supercomputing Center under grant PHY890020P. This work has been supported by the Office of Naval Research.

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