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Resonant-tunneling transfer times between asymmetric GaAs/$Al_{0.35}Ga_{0.65}As$ double quantum wells

M. G. W. Alexander, M. Nido,* and W. W. Rühle
Max-Planck-Institut für Festkörperforschung, D7000 Stuttgart 80, Federal Republic of Germany

K. Köhler
Fraunhofer-Institut für Angewandte Festkörperphysik, D7800 Freiburg, Federal Republic of Germany
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Electron tunneling through the barrier in asymmetric double-quantum-well structures is investigated by time-resolved picosecond luminescence spectroscopy. Change from nonresonant to resonant tunneling is achieved with a perpendicular electric field. Energetic alignment of electron subbands in the two wells strongly enhances tunneling transfer rates. The resonant transfer times decrease strongly with barrier thickness. The wells are coupled at resonance by energy-conserving scattering processes between states localized in a single well. The buildup of delocalized coherent states at resonance would lead to much shorter transfer times.

Tunneling of carriers in semiconductor heterostructures has been extensively studied by transport measurements.[1] However, the dynamical aspects of tunneling, although important for the physical understanding as well as for device applications, have remained to a large extent unclear. Indirect measurements of the time constants by high-frequency oscillation[2] and time-of-flight[3] experiments show a change in the carrier dynamics due to a transition from *nonresonant* to *resonant* tunneling, i.e., from a situation where initial and final states are nonenergetically aligned to the case of level alignment. However, it is still discussed whether the resonances between the various levels of the heterostructures are caused by *coherent* or *sequential* tunneling.[4] Recent experiments[5,6] support the sequential rather than the coherent picture.

A promising insight into this important question is provided by direct measurement of the carrier dynamics in semiconductor heterostructures by the means of time-resolved optical spectroscopy. The tunneling escape process from a single quantum well (QW) (Refs. 7 and 8) as well as the tunneling transfer process in an asymmetric double-quantum-well (ADQW) structure were studied. The ADQW has been considered to be an ideal structure to study the physics of nonresonant as well as resonant tunneling, since the carrier confinement can be tuned, by an electric field perpendicular to the QW's from intrinsic localization to strong coupling of the wells.[9,10] Nonresonant electron-tunneling transfer times were measured in ADQW's based on the GaAs/AlAs,[11] $In_{0.53}Ga_{0.47}As/InP$,[12] and $GaAs/Al_xGa_{1-x}As$ (Refs. 13–16) systems. Oberli *et al.*[17] have recently reported the observation of the transition from nonresonant to resonant electron tunneling by applying an electric-field perpendicular to a $GaAs/Al_xGa_{1-x}As$ ADQW sample. The resonant transfer time was of the order of picoseconds. This measurement led the authors to the assumption that longitudinal-optical (LO) phonon intersubband scattering limits their transfer times, which made it impossible to observe the faster coherent tunneling time. Monte Carlo simulations had been performed to support their conclusion.[18]

In this paper, we present results of time-resolved picosecond photoluminescence (PL) on ADQW's with varying barrier thicknesses d in a perpendicular electric field F. Our results demonstrate for the first time that resonant electron transfer in ADQW's can be observed up to remarkably thick barriers. The observed resonant transfer times are much longer than expected and depend strongly on barrier thickness. Our experiments yield two important conclusions: First, we show that LO phonon intersubband scattering out of a resonant coherent state extending over an entire ADQW structure is *not* the limiting factor of the measured transfer times at resonance. Second, a resonance without any coherent state formation requires electron scattering between states localized in adjacent wells for barrier transfer. The efficiency of such scattering has to depend strongly on the energetic separation of the quantized energy levels in the adjacent wells and has to be largest for level alignment. Scattering mechanisms meeting this claim will be suggested.

The samples used in our experiments are all grown by molecular-beam epitaxy (MBE) on Si-n^+-doped (100) GaAs substrate. First, a buffer layer is grown, consisting of 600-nm GaAs, 20-nm $Al_{0.35}Ga_{0.65}As$, 1-nm GaAs, 10-nm $Al_{0.35}Ga_{0.65}As$, and five periods of a 2-nm GaAs/3-nm $Al_{0.35}Ga_{0.65}As$ superlattice. The ADQW structure itself, depicted schematically in Fig. 1, consists of a 100-nm $Al_{0.35}Ga_{0.65}As$ layer, a wide quantum well (QWW) of GaAs (width w_w of 10 nm), a barrier layer of $Al_{0.35}Ga_{0.65}As$ (thicknesses d of 6 nm, 8 nm, or 20 nm), a narrow quantum well (QWN) of GaAs (width w_n of 5 nm), and a 100-nm $Al_{0.35}Ga_{0.65}As$ layer. A 4-nm GaAs cap layer covers the structure. In contrast to the structures used by all other groups in previous experiments, our samples contain only *one* ADQW. Such a single ADQW avoids artifacts due to averaging vertically over thickness fluctuations of several barriers and inhomogeneous field distributions. The thickness of the layers and the aluminum composition are checked by reflection high-energy electron-diffraction (RHEED) oscillation measurements on reference samples. Transmission-electron microscopy (TEM) at near-atomic resolution and PL measurements

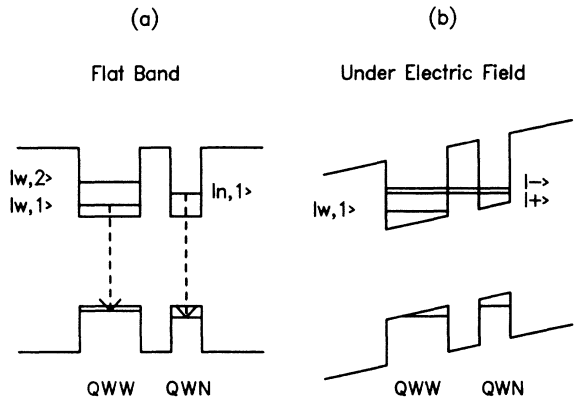

FIG. 1. Ideal, one-dimensional band diagram of ADQW's (a) under flat band conditions and (b) under resonance conditions. Also shown are the eigenstates of the system, which are discussed in the paper. The labeling is the same as in the text. The dashed lines depict the observed optical transitions.

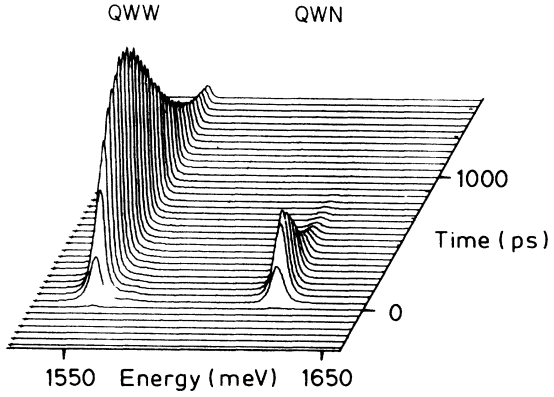

FIG. 2. Typical streak-camera spectrum of an ADQW sample. This spectrum is taken from a sample with a 6-nm barrier at 5 K. The electric field is zero, corresponding to flat band conditions as shown in Fig. 1(a).

show that the nominal data are correct to within at least 10%. A conventional Ohmic contact of AuGe/Ni is alloyed into the substrate side of the samples. Semitransparent 5-nm-thick NiCr-Schottky contacts of 1.0 mm diam are evaporated on the front side. The Schottky diodes are reverse biased in the experiments, showing low reverse dark currents $\leq 5\ \mu$A and breakdown voltages of more than 6 V. The samples are fixed on a cold finger in a helium cryostat with a temperature tuning range from 5 to 300 K. Excitation is performed with pulses of about 5 ps width, obtained from a tunable Styryl 8 dye laser, synchronously pumped by a mode-locked Ar^+-ion laser with a repetition rate of 80 MHz. The excitation energy is tuned either to 50 meV above the ground state $|n,1\rangle$ of QWN to excite both QW's simultaneously (high excitation energy) or to just below $|n,1\rangle$ to excite only QWW (low excitation energy). Measurements are performed with high excitation energy, if not mentioned otherwise, with an excitation density of about 10^{10} cm^{-2}. The PL from the samples is dispersed by a 0.32-m monochromator and detected by a two-dimensional synchroscan streak camera with an S-20 cathode. The time- and wavelength-resolved PL spectra are thus obtained with a temporal resolution of 10 ps and a spectral resolution of 0.5 nm.

A typical result is shown as a three-dimensional plot in Fig. 2. Luminescence decay times τ^* are determined by spectral integration of the luminescence of each QW and fitting the obtained temporal decays to single exponential decays. We have shown previously[16] that the measured decay time of the QWN luminescence τ_n^* is given by

$$1/\tau_n^* = 1/\tau_T + 1/\tau_R\ , \qquad (1)$$

where τ_T is the tunneling transfer time and τ_R is the radiative recombination time, which to first approximation is given by τ_w^*, the decay time of the luminescence of QWW measured with low excitation energy. Changes in the decay time τ_n^* of QWN directly map variations in the tunneling transfer time τ_T, since τ_R depends only weakly on perpendicular electric fields.[19]

The PL peak energies exhibit a shift to lower values with increasing field caused by the quantum confined Stark effect (QCSE).[20] The shift of the PL from QWW is used to determine the internal electric field F in the ADQW's; results of a transfer-matrix calculation[21] are fitted with appropriate parameters[22] to the PL peak energy of QWW at zero field. A transfer-matrix calculation using the derived parameters gives the peak energy versus field dependence. The internal field value is thus determined from the measured PL peak energy. This *internal* field calibration excludes such artifacts as nonlinear field distributions at the edges of the intrinsic region of the heterostructures, which can strongly vary from one sample to another.

The main result of our experiments can be seen in Fig. 3. The PL decay time τ_n^* of QWN is shown as a function of the electric field for 6-, 8-, and 20-nm-thick barriers at

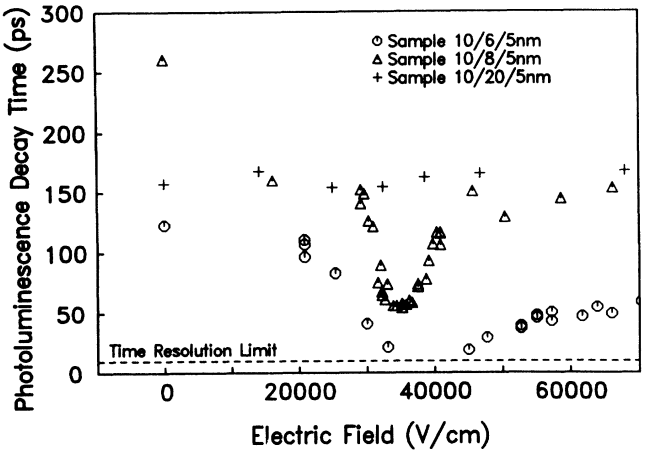

FIG. 3. Decay times of the luminescence from QWN vs electric field perpendicular to the layers, taken at 5 K. Resonances at about 35 kV/cm for the 8-nm barrier and about 40 kV/cm for the 6-nm barrier sample can be clearly seen.

5 K. For the sample with a 6-nm barrier, the decay time, already fast at $F=0$ due to efficient nonresonant tunneling,[16] rapidly decreases up to field values of 40 kV/cm, where the decay time becomes faster than our time resolution. Above 45 kV/cm, the decay time becomes longer again, but does not recover to its initial values. The decay time τ_n^* of the sample with an 8-nm barrier exhibits a clear resonance with a decrease of τ_n^* from 220 ps at zero field to 50 ps at 35 kV/cm and a recovery to 150 ps for higher field values. For the 20-nm barrier sample, we do not observe any significant change in the decay times τ_n^*. Notice that in the case of the 20-nm barrier sample at $F=0$ the absolute values of τ_n^* are reduced according to Eq. (1) by a factor of 2 compared with the value of the 8-nm sample, since the radiative recombination time τ_R is reduced due to better sample quality.[23] This reduction of τ_R is found by measuring τ_ω^* as mentioned above. Measurements of the decay times τ_n^* versus field for all samples at 50 K show that the values of τ_n^* at the resonance minima do not change with temperature, but the resonance broadens considerably.

The theoretical field values necessary to align the first electron level $|n,1\rangle$ of QWN and the second electron level $|w,2\rangle$ of QWW are calculated with the transfer-matrix formalism mentioned above. We obtain critical fields of 38 kV/cm for the 8-nm and of 44 kV/cm for the 6-nm barrier. These values coincide within the error of our field calibration of ± 5 kV/cm with the measured positions of the resonances, giving evidence to the fact that we observe resonances due to the alignment of the considered electron levels. The incomplete recovery of the decay times above the resonances are explained by the fact that at high fields the electrons cannot only relax to the ground state $|w,1\rangle$, but also into the $|w,2\rangle$ state.

In a coherent quantum-mechanical picture, the alignment of $|n,1\rangle$ and $|w,2\rangle$ would lead to the buildup of a bonding and an antibonding state

$$|\pm\rangle = 1/\sqrt{2}(|n,1\rangle \pm |w,2\rangle) . \tag{2}$$

The levels would repel each other with an energy ΔE_{rep}, and a wave packet could oscillate between the wells with a time constant $\tau_{\text{coh}} = \hbar/(2\cdot\Delta E_{\text{rep}})$. We calculate for these time constants values of 1.2 and 5.6 ps for the 6- and 8-nm barrier sample, respectively. Bonding and antibonding states could decay into the ground state $|w,1\rangle$ by LO phonon intersubband scattering.[17] The scattering time would be given by

$$\tau_T^{-1} = 2\pi/\hbar\,|\langle w,1|H_F|\pm\rangle|^2\rho , \tag{3}$$

with the Fröhlich interaction Hamiltonian H_F and the density of the final states ρ. By inserting Eq. (2) into Eq. (3), we easily see that the LO phonon intersubband scattering time in an ADQW could at most increase by a factor of 2 (Ref. 24) with respect to the case of a single QW. The intersubband scattering time for a single QW was recently measured to be 1 ps (Ref. 25), i.e., we expect a maximum of 2 ps for the intersubband scattering time in the ADQW. The prediction of this coherent model is in clear contradiction to our observations, which show a value of even 50 ps in the case of an 8-nm barrier and a strong dependence of the transfer time on barrier thickness. The application of a coherent model is also very doubtful with respect to measured dephasing times,[26] which are much faster than the time constants observed in our experiments.

To explain our results, we have to consider *even at energetic alignment* of $|w,2\rangle$ and $|n,1\rangle$ a sequential model of two single QW's coupled by a resonant scattering process which depends strongly on barrier thickness. The strong influence of the barrier thickness on the decay time τ_n^* at the resonance minimum means that we measure the time constant of this resonant scattering rather than the LO phonon intersubband scattering time if we look at τ_n^*. This interpretation of the decay times at resonance immediately explains the results of Oberli *et al.*,[17] who measured a time of 7.5 ps at resonance, which is much longer than the expected value of 2 ps. However, since the coupling depends strongly on the barrier thickness, it could probably be possible to observe the LO phonon intersubband scattering in samples with barriers thinner than those used up to now.

We discuss in the following possible resonant scattering processes leading to tunneling transfer at the resonance position. Our experimental results lead to the conclusion that the scattering process has to depend strongly on the energy-level separation $\Delta E = E_{w,2} - E_{n,1}$, where $E_{w,2}$, $E_{n,1}$ are the energy eigenvalues of $|w,2\rangle$ and $|n,1\rangle$. The scattering efficiency must reach a maximum for the case of energy conservation, i.e., $\Delta E = 0$, and has to depend strongly on the barrier thickness. The absolute value of the scattering times should not depend strongly on temperature. The scattering from $|n,1\rangle$ to $|w,1\rangle$ by LO phonons in the barrier would show resonances at $\Delta E = 36$ meV or $\Delta E = 50$ meV, the energies of the GaAs- and AlAs-like phonon,[27] in contradiction to our observations. Following Sawaki *et al.*,[28] the condition of a strong dependence on ΔE and a maximum at $\Delta E = 0$ would be satisfied by a scattering process involving ionized impurities in the barrier. The same theoretical argument could hold for alloy scattering in the ternary barrier as well as for interface scattering. All three scattering processes on the one hand depend on the penetration of the wave function $|n,1\rangle$ into the barrier, i.e., strongly on barrier thickness. Since on the other hand all these processes depend only weakly on temperature, we cannot distinguish between them in our experimental results.

In summary, we show results of picosecond time-resolved luminescence measurements on asymmetric double-quantum-well structures with a varying perpendicular field. Resonances are detected if the first electron energy level in the narrow QW and the second level in the wide QW align. The transfer times at resonance are found to be strongly dependent on barrier thicknesses, in contradiction to predictions of a coherent description of the system. Scattering processes with energy conservation are necessary to explain the transfer from the narrow to the wide well. Our measurements exclude LO phonon scattering processes, leaving impurity-assisted, interface or alloy scattering as the possible scattering mechanisms.

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*On leave from Opto-Electronics Research Laboratories, NEC Corporation, 4-1-1, Miyazaki, Miyamae-ku, Kawasaki, Kanagawa 213, Japan.

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