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Direct measurement of resonant and nonresonant tunneling times in asymmetric coupled quantum wells

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We have directly measured the tunneling times in an asymmetric coupled double-quantum-well structure using subpicosecond luminescence spectroscopy. We observe a strong dependence of the nonresonant tunneling times on the barrier thickness. The most striking feature of the data is a drastic reduction of the tunneling time and then a further increase as an applied electric field sweeps electron levels in the two wells through a resonance. The measured resonant tunneling time is considerably longer than the estimated coherent tunneling time.

Since the historical discovery of the tunnel effect in the α decay of radioactive nuclei, tunneling has intrigued scientists for both fundamental reasons and potential applications, e.g., tunneling electron microscopy. Following the seminal work by Esaki and Tsu,[1] many studies have been devoted to tunneling in semiconductor heterostructures.[2] The demonstration of very-high-frequency oscillations in semiconductor double-barrier diodes[3] has generated considerable excitement, and was followed by the observation of intrinsic bistability[4] and charge accumulation.[5] Electron transport by sequential resonant tunneling[6] and photoconductive gain due to effective-mass filtering[7] were observed in superlattice heterostructures.

Conceptually, the fundamental properties of tunneling are well understood,[8] although the significance of tunneling times and their magnitudes remain to be unraveled. Experimentally, the static aspects of tunneling have been explored in various structures; the dynamical aspects of tunneling, however, are only beginning to be addressed with the advance of ultrafast spectroscopic techniques. Recently, the escape time of electrons photoexcited into the quantum well of a double-barrier resonant-tunneling structure (DBRT) was measured by time-resolved luminescence,[9] and the transport of electrons perpendicular to the layers of a multiple-quantum-well structure was studied by a time-resolved photocurrent technique.[10] In this latter case, however, the tunneling of carriers cannot be isolated as it is intertwined with diffusion, energy-relaxation mechanisms, and carrier drift.

In this paper we report on a direct measurement of the resonant and nonresonant tunneling times across a single potential barrier separating two quantum wells of unequal thicknesses by subpicosecond time-resolved-luminescence spectroscopy. We demonstrate that the tunneling times out of the narrow well depend strongly on the barrier thickness and compare these with theoretical estimates of phonon-assisted tunneling times. The most exciting observation is a drastic reduction of the tunneling time in an asymmetrical double-quantum-well structure (DQWS) when the ground state of one well is resonantly coupled to the first-excited state of the adjacent well by applying an electric field. At resonance, the measured tunneling time is substantially larger than that estimated from the energy splitting of the coupled electronic states, suggesting that the tunneling process does not proceed in a fully coherent manner.

In an asymmetrical DQWS the energy eigenstates are localized in each well, and the wells are only weakly coupled. This is in contrast to the symmetric case for which the wells are strongly coupled. The extent of the localization, however, can be varied by applying an electric field perpendicular to the layers. Delocalization of the ground state in the narrow well is expected to occur, for example, at a value of the electric field such that the $n=1$ level in the narrow well coincides with the $n=2$ level in the wide well (Fig. 1). In this study we use the decay time of the narrow-well luminescence to probe the tunneling of electrons across the barrier separating the two quantum wells. The degree of localization is thus reflected in the tunneling rate.

The samples studied were grown at 600 °C by molecular-beam epitaxy on (100) n-type GaAs substrates. Eight periods of an asymmetrical double-quantum-well unit are placed inside the intrinsic region of a p-i-n diode structure; a 150-Å-thick layer of $Al_{0.3}Ga_{0.7}As$ separates each period. Each unit consists of two GaAs wells of different thicknesses, nominally 70 and 100 Å, and an $Al_{0.3}Ga_{0.7}As$ barrier. We investigated three different samples with $Al_{0.3}Ga_{0.7}As$ barrier thicknesses of 40, 55, and 65 Å. The layer thicknesses measured by transmission electron microscopy (TEM) were 10% smaller than the nominal values and were consistent with those derived from the luminescence measurements. The total width of the intrinsic region was 5600 Å. The sample was processed into an array of diode mesas (area 200×200 μm^2, height 4 μm) with gold electrical contacts on top of each mesa and a common contact on the substrate side. The current-voltage characteristics of the p-i-n diodes featured low reverse-bias current of less than 100 pA (in the dark) and breakdown voltages in excess of 20 V. The processed sample was mounted on a sapphire disk and placed in thermal contact with a cold finger

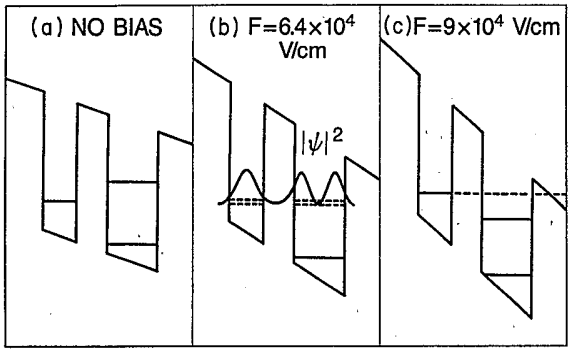

FIG. 1. Conduction-band edge of an asymmetric DQWS for three values of the applied electric field. Under flat-band conditions, subband energies are 61 meV ($n=1$) in a narrow QW, and 32 meV ($n=1$) and 143 meV ($n=2$) in a wide QW.

(~20 K) inside an optical cryostat.

Short optical pulses of 750 fs duration tunable over the range 7200–8000 Å were generated at a 82-MHz rate by synchronously pumping a dye laser (Styryl 8) with the compressed and frequency doubled output of a mode-locked, continuous-wave, neodymium-doped yttrium aluminum garnet laser. Time-resolved luminescence is realized by the energy up conversion in a $LiIO_3$ crystal of a photon from the photoexcited luminescence with a photon of a delayed pump-laser pulse.[11] The photoluminescence is excited using an areal energy density of approximately 500 nJ/cm^2 per pulse at a photon energy of either 1.713 or 1.675 eV. Under these conditions the photon energy changed from below to above the $n=2$ excitonic transition of the wide well without any influence on the time constants.

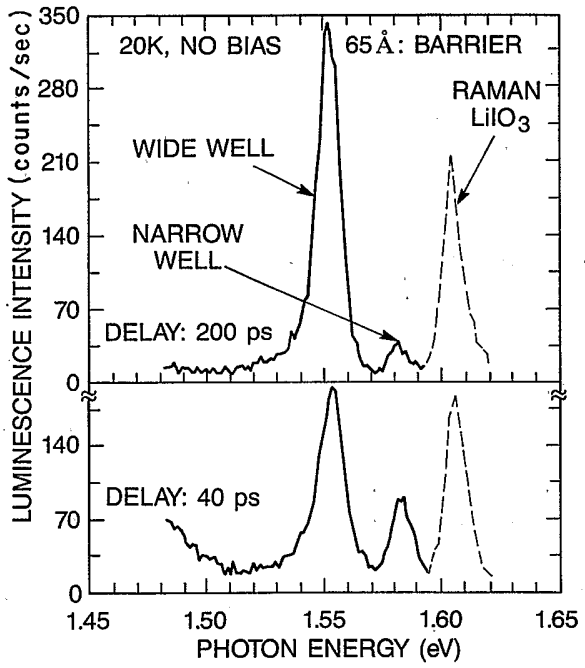

FIG. 2. Luminescence spectra at 20 K of an asymmetric coupled DQWS of 65 Å barrier thickness. The energy positions of the excitonic lines are associated with 63- and 90-Å well widths.

Photoluminescence spectra of a 65-Å asymmetrical DQWS under no applied bias are shown in Fig. 2 for two different delay times measured relatively to the excitation-laser pulse. As the delay time increases, the narrow-well luminescence decreases, and the luminescence from the wide well becomes the dominant feature of the spectrum. A time-resolved measurement reveals a large difference between the narrow- and wide-quantum-well luminescence decay times: 150 and 620 ps, respectively. Furthermore, the decay-time constant in the narrow well is strongly correlated to the thickness of the $Al_xGa_{1-x}As$ layer: 45- and 75-ps values were measured for 40- and 55-Å barrier thicknesses, respectively; the wide-well decay times remained much larger (520 and 1500 ps, respectively).

The radiative and nonradiative recombination times in good-quality quantum-well structures are known to be a few nanoseconds, and thermal ionization of the carriers out of the narrow well is not likely at 20 K. Furthermore, the large mass of the heavy holes makes their tunneling rates extremely small, and the photoexcited light holes are expected to thermalize to the heavy holes very rapidly. Therefore, the short decay times of the narrow-well luminescence are a direct consequence of the tunneling of electrons to the wide well.

We now consider the influence of an electric field applied perpendicular to the layers on the dynamics of photoexcited electrons in the asymmetrical DQWS. The effect of the electric field on the decay-time constant of the narrow-well luminescence is illustrated in Fig. 3 for three values of the reverse bias. Within the experimental accuracy of our measurements, the time evolution remains exponential over an order of magnitude. These decay times are directly identified with the tunneling times of electrons out of the narrow well. The dependence of these tunneling times, as the applied bias sweeps through resonance [Fig. 1(b)] is shown in Fig. 4. The tunneling times, after remaining nearly constant between the flat-band condition (1.8 V) and −2.0 V, decrease abruptly

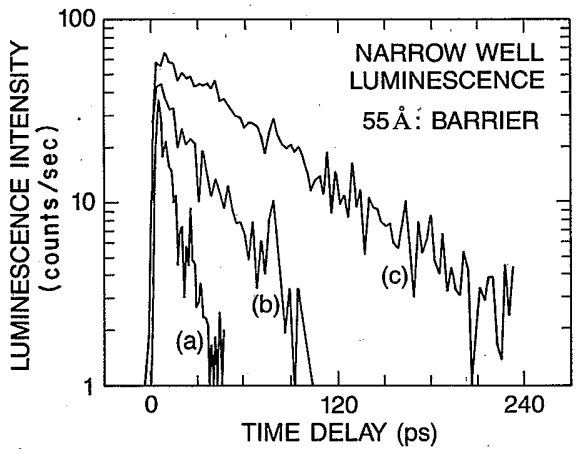

FIG. 3. Luminescence intensity in narrow quantum well vs time for an asymmetric coupled DQWS of 55 Å barrier thickness; temperature is 20 K, carrier density 2×10^{10} cm^{-2}, and laser excitation energy 1.713 eV: (a) −3.5 V, (b) −2.4 V, and (c) −2.0 V.

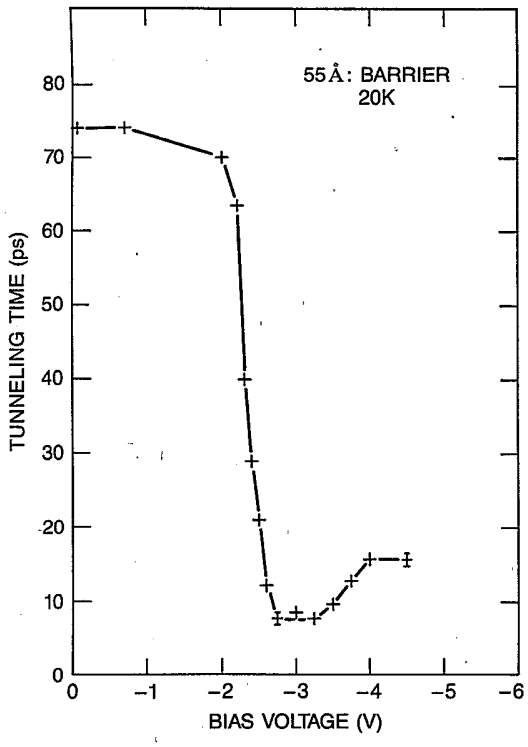

FIG. 4. Tunneling time vs applied voltage of an asymmetric coupled DQWS of 55 Å barrier thickness. Tunneling times are identified with the decay-time constants of the narrow-well luminescence. By definition, reverse-bias voltages have negative values.

at a bias voltage centered at −2.4 V. Moreover, after reaching a minimum, the tunneling times increase again at lower bias voltages. The *nonmonotonic* behavior of the tunneling times is a definite signature of *resonant tunneling,* since other processes (e.g., thermionic emission at higher temperature) would exhibit a monotonic decrease of the decay-time constants with increasing electric field. This conclusion is supported by the observation of an increase of the photocurrent at the same voltage that characterizes the decrease of the tunneling time constants.[12] We show below that the behavior shown in Fig. 4 is a result of resonant coupling between the $n=1$ electron level of the narrow well and the $n=2$ level of the wide well. At a bias smaller than −3 V the luminescence decay time clearly increases, which indicates that the two wells are again weakly coupled. Nevertheless, it does not recover to the low-field value because the energy-potential profile of the DQW structure becomes extremely asymmetric at high fields [Fig. 1(c)]. This situation is analogous to a DBRT structure in which the electron escapes from a confined state of the QW into an energy continuum of states.

Before discussing the magnitude of the tunneling times, we compare the position of the resonance against the value of the electric field necessary to align the $n=1$ level of the narrow well with the $n=2$ level of the wide well. Based on the matrix-transfer technique described by Ricco and Azbel,[8] we estimate that resonant coupling occurs at a field[13] of 6.4×10^4 V/cm. In order to calibrate the internal electric field at −2.7 V, the voltage at which the tunneling times reach a minimum, we have measured the photocurrent-versus-wavelength characteristics of the diode for various values of the bias voltage;[14] the energy shift of the heavy-hole exciton is then checked against the theory of the quantum-confined Stark effect developed by Miller *et al.*[15] This comparison measures the internal field to be 7.0×10^4 V/cm for a −2.7-V bias. One explanation for the difference between the calculated and measured fields for resonant tunneling is that in the luminescence experiment the injected carrier density is sufficiently high to change the internal field because of some space-charge buildup from the holes left in the wells. This effect was experimentally confirmed by the observation of a shift of the resonance to higher voltage as the photoexcited carrier density was decreased from 4×10^{10} to 1×10^{10} cm^{-2}, the minimum value of the tunneling time was not affected. A rough estimate, assuming that all the electrons ($\sim2\times10^{10}$ cm^{-2}) have tunneled from the narrow to the wide well, leads to a worst-case estimate of the opposing electric field of $\sim3\times10^3$ V/cm. In reality, the buildup is a dynamical process which takes place simultaneously with the tunneling of electrons. There may be some additional effect from electrons leaving the wells altogether.

In the following section we discuss the magnitudes of the tunneling times. At resonance, the tunneling times reaches a minimum value of 7.5 ps for a 55-Å-thick barrier. In an ideal case, the resonant coupling of two energy subbands across a potential barrier produces extended states whose energy splitting is primarily defined by the barrier thickness and an effective energy-potential discontinuity. Under resonant conditions, tunneling is then a completely elastic and coherent process. In the absence of any damping (caused either by inelastic processes introducing friction in a classical sense or by elastic processes destroying the phase coherence), the particle oscillates back and forth across the barrier; half of that period represents a characteristic time associated with resonant tunneling ($\tau_{tu}=h/2\Delta E$). The coherent tunneling time, τ_{tu}, is calculated to be 0.67 ps for the 55-Å-barrier sample and 1.2 ps for a 65-Å barrier. Even though we have described the coupling of electronic states under an applied electric field, the basic features of tunneling are similar for an electron and an exciton.[16]

The measured resonant tunneling time (7.5 ps, Fig. 4) is significantly longer than the coherent time calculated above for several reasons. In a real system the coherence of the oscillating wave packet cannot be sustained for times exceeding some other characteristic time, e.g., the momentum-relaxation time or the impurity-scattering time.[17] Another mechanism of importance is the LO-phonon intersubband scattering process acting upon an electron on the first-excited subband. If the scattering time constant is long compared to the coherent tunneling time, an electron reaching the wide well will have a finite probability to tunnel back to the narrow well. In view of a recent experiment by Seilmeier *et al.*,[18] an upper-bound value of the intersubband scattering time is 3 ps. On that basis, the measurement of the tunneling time at reso-

nance is presumably controlled by the larger intersubband scattering time. The inhomogeneous broadening of the excitonic line is another factor to consider in this context. In our structure we have measured a linewidth of 4.3 meV in the narrow QW in excess of the calculated energy splitting (3.1 meV). The minimum value of the decay time then is an average over separate regions of the well that exhibit a resonance at a slightly different value of the internal field. Finally, the inhomogeneity in the electric field, which is caused by the background impurity level of $p \sim 3 \times 10^{15}$ cm^{-3} deduced from a C-V measurement, leads to a broadening of the resonance by about 0.5 V, in agreement with the data of Fig. 4. Any broadening mechanisms of the resonance increase the measured tunneling time at resonance. The observation of a region (-2.7 to -3.0 V) over which the resonant tunneling time remains constant is a consequence of the inhomogeneous broadening of the exciton line and of an inhomogeneous electric field. Of all the possible mechanisms affecting the decay time of the narrow-well luminescence, the intersubband scattering process is, in this system, the main intrinsic limitation to an absolute measurement of the coherent tunneling time.

Finally, we discuss the magnitudes of the off-resonance tunneling time. In contrast to the case of a DBRT structure, tunneling requires a collision in order to conserve the electron parallel momentum. Phonon-assisted transitions are expected to contribute to tunneling if the subband energy separation in adjacent wells exceeds one-LO-phonon energy [Fig. 1(a)]. An estimate of this tunneling time[19] in the limit of thick enough barrier yields a value of 210 ps for a 50-Å barrier separating a 60- and a 88-Å QW. It should be noted that bulk-phonon modes were used in this calculation. The relevance of confined and interface phonon modes to this calculation has to be reassessed since their inclusion could lead to an increase of the phonon-assisted tunneling rates[20] large enough to explain a shorter luminescence decay time of 75 ps. However, the present study cannot distinguish whether phonon-assisted tunneling dominates over impurity-related tunneling.

In this study we have demonstrated the existence of a large resonant enhancement of the tunneling rates by time-resolved-luminescence spectroscopy in an asymmetrical DQWS. This enhancement results from the resonant coupling of the ground state of an electron in the narrow well with the first-excited state in the wide well. A range of dynamical effects were shown to affect the exact position of the resonance as well as the tunneling time at resonance. The measured resonant tunneling time is considerably longer than the estimated coherent tunneling time. Away from resonance, a strong dependence of the tunneling times on the barrier thickness is demonstrated.

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[12]A peak in the photocurrent is not expected and not observed because the tunneling times are much shorter than the carrier lifetime.

[13]This value of the electric field can be obtained more simply by assuming that the applied electric field causes a rigid shift of the ground-state energy of the narrow QW with respect to the first-excited–state energy of the wide QW. This approximation amounts to neglecting the energy Stark shift of the two states. In the absence of any internal electric field, the difference in confinement energies is equal to 82 meV (ΔE). The value of the electric field is then derived from $|\mathbf{F}| = 2\Delta E/q(d_{w1} + 2d_b + d_{w2})$.

[14]These results will be reported elsewhere.

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[16]Because of the Coulomb interaction between an electron and a hole, the voltage at which the resonance occurs could be slightly shifted. In view of the relatively small changes of the binding energies with well thickness, this shift is negligible compared to the magnitude of the electron-confinement energies; furthermore, the energy splitting ΔE is identical to first order for both electronic and excitonic states.

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