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POSSIBILITY OF A DIRECT OBSERVATION OF THE TIME EVOLUTION IN HETEROSTRUCTURE BARRIER TUNNELING

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By varying *two* design parameters, e.g., the width of a quantum well (QW) and its chemical composition, or by applying an external field, it is possible to implement two QW's with identical ground-state levels in the conduction band – but different in the valence band. This allows a selective "preparation" of an initial electron state by interband photoexcitation. In a coupled QW system the electron will oscillate between the two wells, giving rise to an oscillating luminescence signal with a period directly related to the tunneling time.

TIME RESOLVED LUMINESCENCE spectroscopy has provided important insight into the dynamical behavior of many physical systems. With the advent of femtosecond lasers and improved frequency up-conversion techniques the time resolution of luminescence spectroscopy has moved into subpicosecond domain.[1,2] Recently, Deveaud et al.[3] applied these techniques to the study of perpendicular transport in GaAs/AlGaAs heterojunction superlattices – observing directly the electron motion through Bloch-type miniband states. The purpose of this Communication is to propose an experiment which should be capable of a direct observation of the time development of resonant tunneling in a coupled quantum-well system.

The idea will be described, assuming its possible implementation within the GaAs/AlAs heterojunction technology. (This specific choice of material is made mainly for clarity of the presentation; other material combinations, especially with a lower effective electron mass m_e, may be used advantageously.) Consider the heterostructure, illustrated in Fig. 1. The structure consists of two quantum wells (QW) separated by an $Al_xGa_{1-x}As$ barrier, $x \gtrsim 0.4$, of thickness L. One of the wells (QW-2) represents a pure GaAs layer of thickness $d_2 \simeq 30$ Å, the other is made of an $Al_yGa_{1-y}As$ alloy with a small fraction of aluminum $y \ll x$. The structure is designed so that the isolated-well ground-state electron levels coincide: $E_1 = E_2 \equiv E_0$. This, however, is achieved by adjusting *two* parameters of the epitaxial growth, not just one. For example, we can increase the width d_1 of QW-1 (which would lower its level E_1 approximately $\propto 1/d_1^2$), and at the same time increase the classical conduction-band energy in that well, approximately in proportion to its aluminum content y.

Inasmuch as the heavy-hole mass m_{hh} in the QW is much larger than m_e, the designed degeneracy of the electronic level is not accompanied by a similar degeneracy in the valence-band QW's. Therefore, the interband transition energies $h\nu_1$ and $h\nu_2$ are different in the two wells, which permits us to selectively excite electrons in one or the other well. These transitions are indicated by the arrows in Fig. 1; for clarity of the picture, small confinement effects in the heavy-hole subband of the QW are ignored.

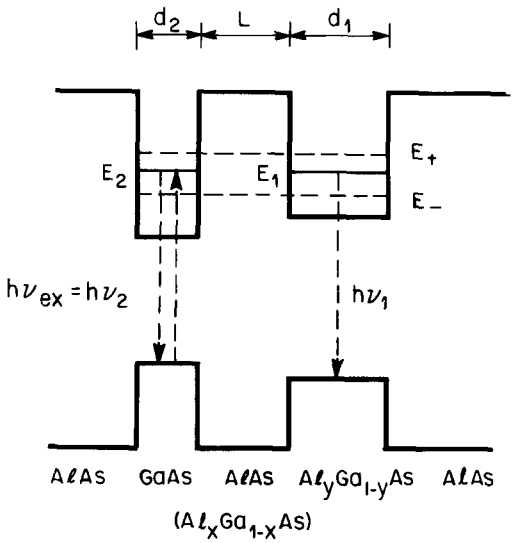

Fig. 1 Schematic illustration of an experimental structure. The tunnel barrier, separating two quantum wells, can be implemented either as a thin ($L \lesssim 30$ Å) AlAs layer or a slightly thicker $Al_xGa_{1-x}As$ layer with $x \simeq 0.4$. The wells can be undoped or lightly p-type; one can also use a modulation-type p doping in the outside AlAs layers.

As is well known, in the presence of a tunnel coupling, the levels E_1 and E_2 are not stationary. True

eigenstates of an ideal two-well system are given by the symmetric and antisymmetric combinations of the single QW states $|1>$ and $|2>$:

$$|\pm> = \frac{1}{\sqrt{2}}\,(\,|1> \pm |2>) \qquad (1)$$

$$\hat{H}\,|\pm> = E_{\pm}\,|\pm>\,, \qquad E_{+} - E_{-} \equiv \hbar\omega > 0\,.$$

Suppose the excitation energy is tuned to the lower of the two frequencies: $h\nu_{ex} = h\nu_2$. In this case, immediately upon the excitation, electrons will be "prepared" in state $|2>$. Subsequent evolution of this state in time is then given by[4]

$$|t> = e^{E_0 t/i\hbar}\,[\,|2>\cos(\omega t/2) - i\,|1>\sin(\omega t/2)]\,, \quad (2)$$

so that the electron densities in wells 1 and 2 oscillate with the frequency ω:

$$|<t\,|\,1>|^2 = \sin^2(\omega t/2) = \frac{1}{2}\,(1 - \cos\omega t)\,, \qquad (3)$$

$$|<t\,|\,2>|^2 = \cos^2(\omega t/2) = \frac{1}{2}\,(1 + \cos\omega t)\,,$$

In the absence of scattering, the luminescence signals at frequencies ν_1 and ν_2 will oscillate according to eqs. 3; the oscillation period T can be estimated as follows:

$$T \equiv \frac{2\pi}{\omega} = \frac{\lambda L m_e}{\hbar}\,e^{2\pi L/\lambda}\,, \qquad (4)$$

where $\lambda \equiv h/\sqrt{2m_e(\Phi - E_0)}$ is the de Broglie wavelength of the tunneling electron, Φ is the heterojunction barrier height. In eq. (4) I used a quasi-classical estimate for the tunnel splitting.

In order to picture the time evolution of luminescence signals in the presence of scattering, it is convenient to separate the time scales involved. Besides the oscillation period T, which is controlled mainly by the barrier phase area $L\sqrt{(\Phi - E_0)}$, the important times are the coherence time τ_{coh}, describing relaxation of the electron phase, and the recombination time τ_{rec}. The latter depends on the material purity and the intensity of the excitation signal. It should be expected that the condition $\tau_{rec} \gg T$ will be achieved easily; perhaps it will be most practical to work with a lightly doped p-type sample. On the other hand, fulfillment of the condition $\tau_{coh} \gg T$ will require a high-quality sample with smooth QW interfaces. In a perfect sample one can expect the electron phase relaxation to be dominated by the processes of acoustic phonon emission. In what follows, it will be assumed that $T \ll \tau_{coh} \ll \tau_{rec}$.

Figure 2 shows the predicted time variation of the luminescence signals upon a short ($\Delta t < T$) excitation pulse. Oscillations will be observed for $t \lesssim \tau_{coh}$ after which the excited electrons will be settled in the metastable "ground" state E_- of the two-well system. The luminescence signal will then continue until all the excited electrons decay via recombination ($t \sim \tau_{rec}$, cf. Fig. 2a). Figure 2b shows, on an expanded time scale, the expected oscillatory luminescence signals at frequencies ν_1 and ν_2; these signals are 180° out of phase.

It may appear that realization of the proposed experiment is inordinately difficult, because in the process

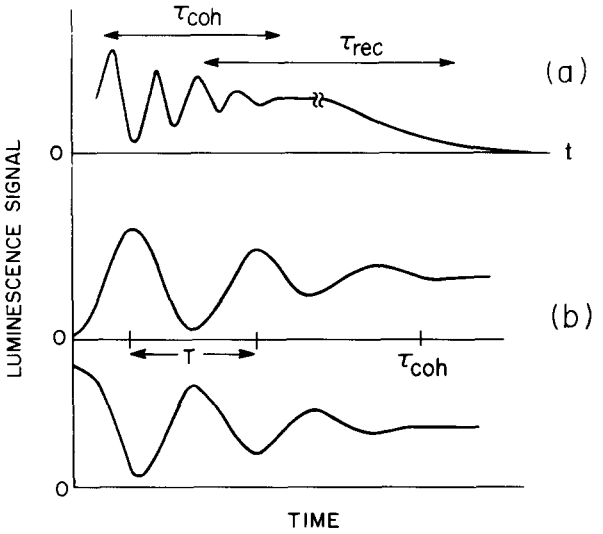

Fig. 2 Expected time evolution of the luminescence signals.
(a) Overall time evolution, showing the three characteristic time scales, assumed to satisfy the inequalities: $\tau_{rec} \gg \tau_{coh} \gg T$;
(b) Signals at $h\nu_1$ and $h\nu_2$ for $t \ll \tau_{rec}$; These signals are shifted by $T/2$ with respect to each other.

of crystal growth one cannot expect to be able to adjust two structure parameters so as to obtain the degeneracy of single-well levels E_1 and E_2 with a desired accuracy. One way of circumventing this difficulty is to place the double-well structure in the lightly-doped or intrinsic region of a *pin* junction – with the n contact on the side of QW-1 – as illustrated in Fig. 3. This permits fine-tuning of the level energies by applying a reverse-bias electric field.[5] (Moreover, introduction of this additional means of control allows one to design both wells to have the same chemical composition.) Of course, in this situation there will be no steady-state holes in the QW's apart from the photo-excited holes, trapped in QW-2. In order to obtain luminescence, one can then pump holes into QW-1 by an auxiliary illuminating of the sample – from the side of QW-1 – with photons above the fundamental threshold of the wide-gap cladding material. For this purpose, it may be advantageous to use an $Ga_zAl_{1-z}As$ layer with $z \ll 1$ in the immediate vicinity of QW-1, as shown in Fig. 3.

One should not underestimate the remaining difficulty in the implementation of an oscillatory luminescence experiment – producing a sufficiently perfect structure, in which the coherence time extends over several oscillation periods. Of course, by making the tunnel barrier more transparent one can always force the condition $T \ll \tau_{coh}$ (cf. the recent observation of a miniband conduction in GaAs/AlGaAs superlattices[3]), but since T has to exceed the pulse duration τ_{pulse}, in practice one is limited by the state of the subpicosecond luminescence art (experiments with $\tau_{pulse} \approx 65$ fsec are in progress, J. Shah, private communication). Taking $L \sim 50$ Å and $(\Phi - E) \sim 0.2$ eV, and using eq. (4), one

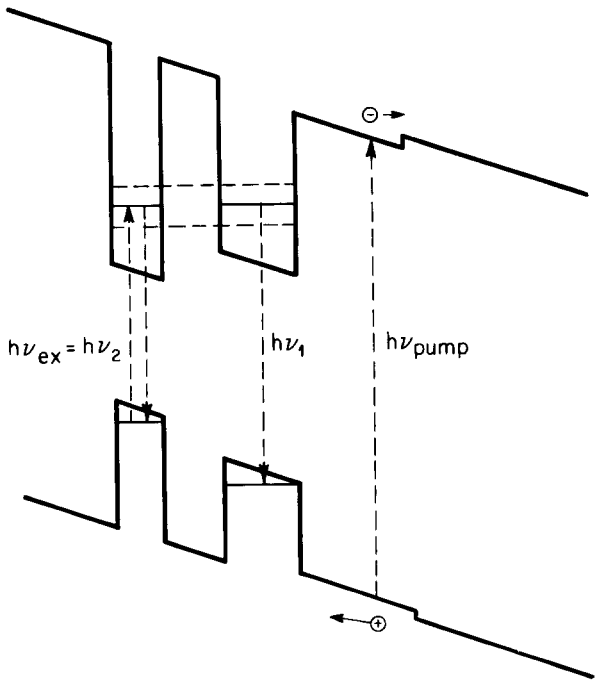

Fig. 3 Modified structure, which allows "fine-tuning" of the single-well levels E_1 and E_2 by the electric field of a reverse-biased *pn* junction. A non-equilibrium population of holes, necessary for the radiative recombination in the quantum wells, is maintained by an auxiliary pumping of interband transitions at frequency ν_{pump} in the cladding layer on the side of the *n* contact.

obtains an estimate $T \sim 0.6$ psec, which means that samples with the low temperature QW mobility of order 10^5 cm^2/V·sec (corresponding to a scattering time ~4 psec) should be adequate. Special attention must also be paid to the inhomogeneous broadening of QW energy levels. Intuitively, it can be expected that one should be able to tolerate spatial fluctuations of the QW and barrier widths with a characteristic length larger than the lateral diffusion length in the QW (which for the above example at 4.2 K is of order 10^{-5} cm). The effect of such inhomogeneities would be to add a spread in the tunneling period and somewhat diminish the oscillation amplitude. Shorter-length fluctuations can be more troublesome.

Observation of the predicted oscillatory luminescence would be of great scientific interest. As pointed out by Kane,[4] the oscillation period T of the electron exchange between two coupled quantum wells represents one of the few instances when the tunneling time can be defined unambiguously. The proposed experiment can be regarded as a solid-state analog of such phenomena as the dipole-moment oscillations of an ammonia molecule used in NH_3 masers,[6] energy exchange between coupled optical dielectric waveguides,[7] and oscillations of neutral K-mesons between states of different strangeness.[8] Potentially, this experiment contains a great deal of information about the microscopic quantum processes in quantum-well structures.

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REFERENCES

1. Jagdeep Shah, T. C. Damen, B. Deveaud, and Dominique Block, *Appl. Phys. Lett.* **50**, 1307 (1987).
2. Recent developments in ultrafast luminescence spectroscopy using sum-frequency-generation techniques have been reviewed by Jagdeep Shah, *IEEE J. Quant. Electron.* (February 1988).
3. Benoit Deveaud, Jagdeep Shah, T. C. Damen, Bertrand Lambert, and Andre Regreny, *Phys. Rev. Lett.* **58**, 2582 (1987).
4. E. O. Kane, in *Tunneling in Solids*, ed. by C. B. Duke, Suppl. 10 of *Solid State Physics: advances in research and applications*, Academic Press (New York, 1969).
5. After this paper had been completed, Jagdeep Shah informed me that similar designs were used in other experiments currently in progress.
6. Murray Sargent III, Marlan O. Scully, and Willis E. Lamb, *Laser Physics*, Addison-Wesley (Reading, Mass., 1974), Chap. 5.
7. Hermann A. Haus, *Waves and Fields in Optoelectronics*, Prentice-Hall (Englewood Cliffs, NJ, 1984), Chap. 7.
8. Kurt Gottfried and Victor F. Weisskopf, *Concepts of Particle Physics*, Oxford University Press (New York, 1984), pp. 148-154.