Coherent Oscillations of a Wave Packet in a Semiconductor Double-Quantum-Well Structure

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We present the first study of the dynamics of an extended electronic wave packet in a solid. The wave packet is created in a GaAs/AlGaAs double-quantum-well structure by ultrashort pulse excitation. We observe the oscillatory motion of the wave packet between the two wells by using time-resolved degenerate four-wave-mixing and pump-and-probe spectroscopy.

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The development of ultrafast laser sources has opened up new possibilities for investigating the dynamics of nonstationary states. Excitation with a spectrally broad pulse encompassing many optical transitions of a system creates a wave packet which is a linear superposition of the eigenstates of the system. The time evolution of such a wave packet in real or configuration coordinate space can be investigated by using a delayed probe pulse, and provides information about the relaxation dynamics of nonstationary states. Such experiments have been recently performed in atoms[1] and molecules.[2]

One of the simplest and most instructive systems for such an investigation is a double-well potential.[3] Recent advances in the growth of ultrathin layers by epitaxial techniques make it possible to realize such a double-quantum-well structure (DQWS) in semiconductors using alternate layers with different band gaps. An asymmetric DQWS, consisting of a narrow well (NW) and a wide well (WW), offers the unique possibility of tuning the eigenstates by applying an electric field. With increasing field of appropriate polarity, the splitting between the two lowest conduction electron levels ($\Delta E = E_1 - E_2$) decreases, goes through a minimum at resonance because of anticrossing, and then increases again. Correspondingly, the wave functions of the two levels, which are localized primarily in one well or the other for zero bias, become increasingly delocalized (with nearly equal probability in each well at resonance; Fig. 1, top), and then become localized again with further increase in bias. Another important feature of the asymmetric DQWS is that the lowest optical transition energies in the two wells are different. This allows one to selectively create an exciton wave packet only in the WW by resonant excitation with an ultrashort pulse of spectral width larger than ΔE. This wave packet subsequently oscillates between the WW and NW with a period equal to $h/\Delta E$, because the two constituent wave functions evolve at a different rate. As the electron levels are tuned out of resonance, the oscillation frequency increases because of increased energy separation, while the oscillation amplitude decreases because of increased localization of the wave functions. Such a structure thus offers many advantages for investigating the time evolution of a wave packet.

In this Letter, we present the first study of the dynamics of an extended wave packet in a solid. We created an electronic wave packet in the WW of an asymmetric GaAs/AlGaAs DQWS using an ultrashort laser pulse and traced the oscillatory motion of this wave packet in real time by degenerate four-wave mixing and by pump-and-probe spectroscopy. By varying the alignment of the levels, the properties of the wave packet are strongly

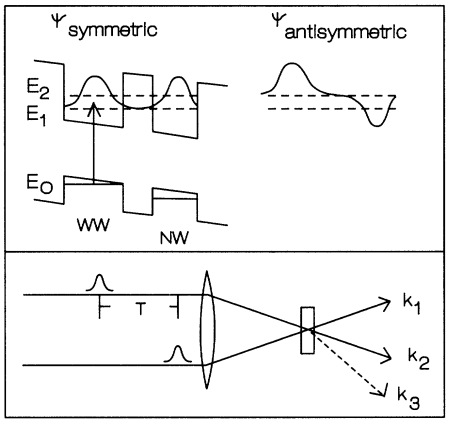

FIG. 1. Top: schematic of the double-well structure at resonance, showing the symmetric (left) and antisymmetric (right) delocalized eigenstates. The vertical arrow indicates the excitation conditions for creation of a wave packet in the wide well (WW). Bottom: geometry of the experiment.

influenced. We present a simple time-dependent theory which accounts for the main features of the experiments and shows that the two time-resolved experimental techniques give complementary information about the damping of the oscillation.

The basic sample structure used in our experiments (Fig. 1, top) consists of a 170-Å GaAs quantum well, followed by a 17-Å $Al_{0.35}Ga_{0.65}As$ barrier, and a 120-Å GaAs quantum well. Ten periods separated by 150-Å $Al_{0.35}Ga_{0.65}As$ barriers are grown on an n-doped GaAs substrate.[4] Part of the substrate is removed by wet etching to allow transmission experiments. The electric field is applied between a semitransparent Au-Cr Schottky contact on top of the sample and the doped back side. The excitation source is a tandem synchronously pumped LDS-751 dye laser system that produces pulses of about 500-fs duration and 4.2-meV spectral width. The laser is tuned close the heavy-hole (hh) exciton resonance of the WW.

In a first experiment, we investigate the dynamics of the photoexcited wave packet by time-resolved self-diffracted degenerate four-wave mixing (DFWM), which has been previously used to study the dephasing of excitons in semiconductors.[5,6] In this experiment, a probe signal is diffracted from an exciton grating into the background-free direction $2\mathbf{k}_2-\mathbf{k}_1$ [$\mathbf{k}_1$ and $\mathbf{k}_2$ are the wave vectors of two incident pulses forming the grating (Fig. 1, bottom)] and is measured as a function of the delay time, giving information about the exciton dynamics.

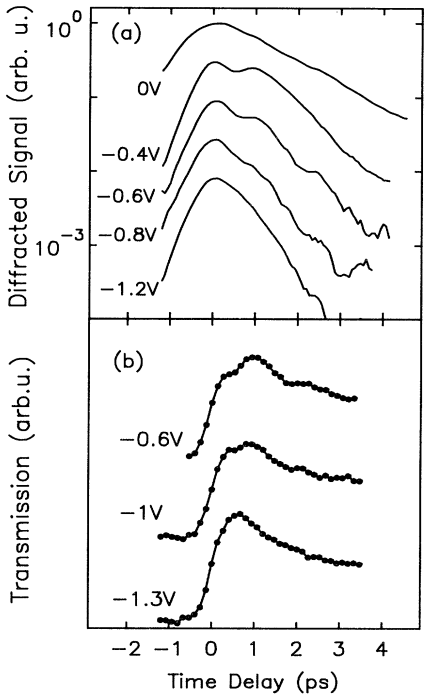

FIG. 2. (a) Diffracted signal and (b) change in transmitted probe intensity vs delay time for different bias fields.

Figure 2(a) shows the DFWM signal versus the delay time for the asymmetric DQWS. Lattice temperature (5 K) and excitation intensity are kept low to minimize phase relaxation by exciton-phonon or exciton-exciton scattering. If the electric field is above (0 V) or below (−1.2 V) the field necessary for resonance, the signal is exponential and reflects the free decay of the polarization, i.e., the loss of phase coherence of the excitons due to scattering processes.[5,6] A strikingly different signal is observed if the conduction electron levels are tuned in resonance (−0.6 V): The decay now shows a clear periodic modulation.

The modulation of the DFWM signal reflects the oscillatory behavior of the wave packet, which was photoexcited in the WW. The oscillation period of about 1.3 ps agrees well with that expected from the measured splitting $\Delta E \approx 3$ meV in the linear absorption spectrum at resonance (Fig. 3). Since the oscillation is significantly damped we observe only one pronounced peak and a weak second oscillation. If we tune the conduction electron levels slightly out of resonance (−0.4 and −0.8 V), the modulation becomes weaker due to the increasing localization of the eigenstates in the wells. If the laser is detuned below or above the hh exciton transition of the WW, the modulation vanishes because a wave packet is not created in this case.

The modulation also disappears for increased excitation intensity or lattice temperature, because dephasing processes become faster than the oscillation period. If we tune the laser close to the hh exciton resonance of the NW, we find a similar modulation and a similar dependence on the separation of the two levels. In this case,

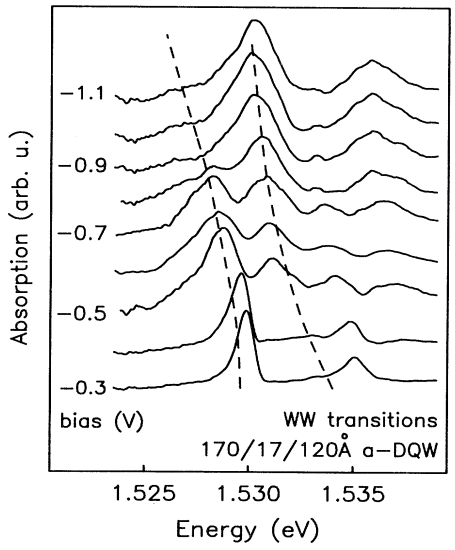

FIG. 3. Linear absorption spectra for different bias fields. The dashed lines are guides for the eye to indicate the two transitions.

however, we also create free carriers in the WW, and we observe a more pronounced damping with increasing intensity, due to the scattering of the wave packet when it is in the WW. This is yet another proof for the spatial oscillation of the wave packet.

Another way to observe the oscillation of the wave packet is a pump-and-probe transmission experiment. In this experiment, a "pump" pulse ($\mathbf{k}_2$) creates the wave packet and the sample transmission is probed with a weak "probe" pulse ($\mathbf{k}_1$) as a function of delay time, $-T$ (Fig. 1, bottom). Figure 2(b) shows the change in transmitted probe intensity versus delay time for the asymmetric DQWS. Temperature and intensity are identical to the DFWM experiment. The trace for a bias of -1.3 V is representative for nonresonant conduction electron levels: The signal has a steplike onset and decays subsequently. The trace for electronic resonance of the wells (-0.6 V) shows a weak oscillatory modulation, with a period in good agreement with the results of the DFWM experiment. If the conduction electron levels are detuned (-1 V), the modulation becomes weaker and the period shorter, as expected.

In a simple picture, these oscillations can be viewed as a change in bleaching of the excitons as the wave packet and the excitons change from spatially direct to indirect and back. Thus the observation of oscillations in pump-and-probe experiments gives further strong evidence for the spatial oscillations of the wave packet.

The linear absorption spectra of the sample obtained with a weak cw laser (Fig. 3) fully support these results and conclusions. For small applied field (-0.3 V), the experimental spectrum shows the first hh and light-hole (lh) exciton transitions of the WW. As the field is increased, a new peak appears above the hh exciton peak (-0.5 V). This new peak gains strength and the original hh exciton peak loses strength until they are about equal at resonance (-0.6 V). The splitting between the peaks is about 3 meV. With further increase in field, the lower-energy peak rapidly moves away and loses strength, while the higher-energy peak moves slowly and acquires the strength of the original hh exciton peak. The spectra reflect the eigenstate probability distribution in the wells: At resonance, both states are delocalized and have nearly equal probability in both wells; out of resonance, the delocalization gradually disappears, and only the transitions within a well are important.[7]

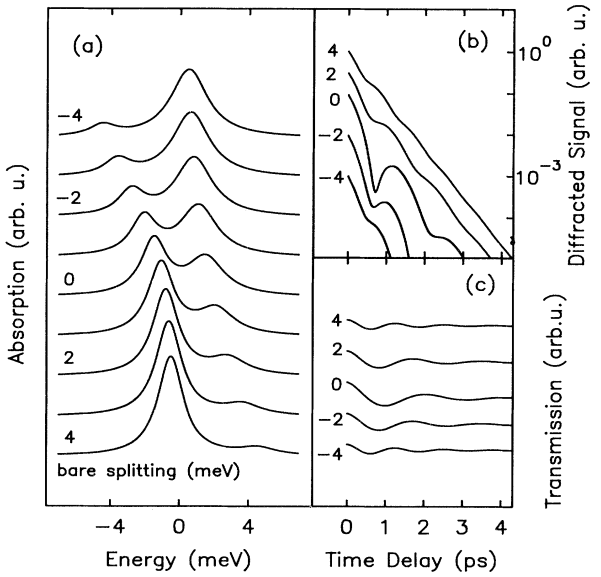

FIG. 4. Calculated (a) linear absorption spectrum, (b) diffracted signal, and (c) change in transmitted probe intensity vs delay time for different bare energy splittings, indicated by the numbers to the left of the traces.

The experimental observations can be qualitatively accounted for by considering a three-level system describing a ground state, $|0\rangle$, and two hybridized excited states, $|\mathrm{WW}\rangle$ and $|\mathrm{NW}\rangle$, only one of which ($|\mathrm{WW}\rangle$) is optically coupled to $|0\rangle$.[2] This translates into two *stationary* and optically coupled excited states, $|1\rangle$ and $|2\rangle$, with energies

$$E_{1,2}=\tfrac{1}{2}\{E_{\mathrm{WW}}+E_{\mathrm{NW}}\pm[(E_{\mathrm{WW}}-E_{\mathrm{NW}})^2+4V^2]^{1/2}\}\,, \quad (1)$$

and spectral weights

$$w_{1,2}=\frac{1}{2}\left(1\pm\frac{E_{\mathrm{WW}}-E_{\mathrm{NW}}}{[(E_{\mathrm{WW}}-E_{\mathrm{NW}})^2+4V^2]^{1/2}}\right), \quad (2)$$

where V is the tunneling rate ($\hbar=1$). In the short (δ function) pulse limit, the linear absorption spectrum (I_a), diffracted signal (I_b), and change in transmitted probe intensity (I_c) are calculated to be

$$I_a(\omega)\propto\frac{w_1\gamma_1}{(\omega-E_1+E_0)^2+\gamma_1^2}+\frac{w_2\gamma_2}{(\omega-E_2+E_0)^2+\gamma_2^2}\,, \quad (3a)$$

$$I_b(T)\propto\Theta(T)\left(\frac{w_1^2}{2\gamma_1}+\frac{w_2^2}{2\gamma_2}+\frac{2w_1w_2(\gamma_1+\gamma_2)}{(\gamma_1+\gamma_2)^2+\Delta E^2}\right)[w_1^2e^{-2\gamma_1T}+w_2^2e^{-2\gamma_2T}+2w_1w_2\cos(\Delta ET)e^{-(\gamma_1+\gamma_2)T}]\,, \quad (3b)$$

$$I_c(T)\propto\Theta(-T)\{w_1^2+w_2^2+w_1w_2[1+\cos(\Delta ET)e^{\gamma_3T}]\}\,. \quad (3c)$$

Here, $\Theta(T)$ is the Heaviside step function, $\gamma_{1(2)}$ the dephasing rate of the 1-0 (2-0) transition, and γ_3 that of the 1-2 transition. Inhomogeneous broadening and dephasing of the 0-0, 1-1, and 2-2 transitions have been ignored.

Figure 4 shows the results of evaluating Eq. (3) for various bare energy splittings, $E_{\mathrm{WW}}-E_{\mathrm{NW}}$, and the following parameters: $\gamma_1=0.9$, $\gamma_2=1.3$, $\gamma_3=0.7$, and $V=1.5$ meV. γ_1 and γ_2 are chosen such that the upper excited state to $|0\rangle$ transition has a faster dephasing rate than the lower one, consistent with its larger width in the linear absorption spec-

trum (Fig. 3) and the faster decay of the DFWM signal [Fig. 2(a)] for large bias. Physically, this corresponds to subband-to-subband scattering, e.g., due to (interface or barrier) disorder on *short* length scales.

The simple theory reproduces the main features of the experiments, except around zero time delay, where the actual pulse profile is important. In agreement with experiment, the modulation and oscillation period in I_b and I_c decrease, as the bare energy splitting increases and the eigenstates become localized. The oscillation in I_b is damped because of the different dephasing of the 1-0 and 2-0 transitions and that in I_c because of the dephasing of the 1-2 transition, reflecting their different physical origin (polarization effects in the former case and population effects in the latter). This shows that the DFWM and the pump-probe experiments give *complementary* information about the wave-packet dynamics.

In conclusion, we report the first experimental observation of coherent wave packet oscillations in a solid. A comparison with a simple theoretical model gives good qualitative agreement and yields new information about the dephasing of the extended states. The period of oscillation and the various damping processes are fundamental quantities that determine the tunneling transfer time between two quantum wells.[8] Recently, DQWS were proposed as a novel scheme of generating terahertz radiation.[3] It would be very interesting to search for this radiation, which should be emitted by the oscillating carriers.

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[1]J. A. Yeazell and C. R. Stroud, Jr., Phys. Rev. Lett. **60**, 1494 (1988); A. ten Wolde, L. D. Noordam, A. Lagendijk, and H. B. van Linden van den Heuvell, Phys. Rev. Lett. **61**, 2099 (1988).

[2]M. J. Rosker, F. W. Wise, and C. L. Tang, Phys. Rev. Lett. **57**, 321 (1986); P. C. Becker, H. L. Fragnito, J. Y. Bigot, C. H. Brito Cruz, R. L. Fork, and C. V. Shank, Phys. Rev. Lett. **63**, 505 (1989); H. L. Fragnito, J. Y. Bigot, P. C. Becker, and C. V. Shank, Chem. Phys. Lett. **160**, 101 (1989).

[3]S. Luryi, Solid State Commun. **65**, 787 (1988); IEEE J. Quantum Electron. (to be published).

[4]All thicknesses are nominal. Various optical measurements indicate that the real thicknesses are within 10% of the design values.

[5]L. Schultheis, J. Kuhl, A. Honold, and C. W. Tu, Phys. Rev. Lett. **57**, 1635 (1986); **57**, 1797 (1986); E. O. Göbel, K. Leo, T. C. Damen, J. Shah, S. Schmitt-Rink, W. Schäfer, J. F. Müller, and K. Köhler, Phys. Rev. Lett. **64**, 1801 (1990).

[6]T. Yajima and Y. Taira, J. Phys. Soc. Jpn. **47**, 1620 (1979).

[7]A similar anticrossing is observed for the NW hh and lh transitions.

[8]D. Y. Oberli, J. Shah, T. C. Damen, C. W. Tu, T. Y. Chang, D. A. B. Miller, J. E. Henry, R. F. Kopf, N. Sauer, and A. E. DiGiovanni, Phys. Rev. B **40**, 3028 (1989).