Wave packets in a semiconductor superlattice

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A theoretical study of hole wave packets in a GaAs, $Al_{0.3}Ga_{0.7}As$ superlattice is presented. Formation of the wave packet by short laser pulse excitation and the time-dependent nature of the wave packet are discussed, with particular attention given to the quasi-periodic nature of the time dependence. Phenomena analogous to those displayed in atomic and molecular wave packet systems are predicted. The wave packet formalism provides a powerful tool for studying coherent hole dynamics, including quantum beat phenomena, and could be useful in determining heavy and light hole relaxation rates.

When a short laser pulse excites a quantum system, a spatially localized wave packet is formed. This phenomenon has been observed in both atomic and molecular systems.[1] In this letter, we present a theoretical study of hole wave packets in a semiconductor superlattice, the unit cell of which consists of four semiconductor layers. The first and third layers are GaAs wells of width 9 and 14 monolayers, 25.4 and 39.6 Å. The second and fourth material layers are $Al_{0.3}Ga_{0.7}As$ barriers of 5 monolayers, 14.1 Å, thickness. This unit cell is then repeated so that, effectively, the material is a series of coupled quantum wells of alternating thicknesses. The hole states of interest are the first and second heavy hole and the first light hole states. The first heavy hole state lies 28.5 meV below the GaAs bulk band edge, while the first light hole state and the second heavy hole state are 40.3 and 52.7 meV, respectively, below the same point. This gives a separation of 11.8 meV between the HH1 and LH1 states and 12.4 meV between the LH1 and HH2 states, with a difference in the separations of about 5%. The band-gap discontinuity is split with 43% allotted to the valance band. Calculations are done for the superlattice at room temperature and the growth direction is [100]. The theoretical model is a $\mathbf{k}\cdot\mathbf{p}$, local pseudopotential method based on that of Smith and Mailhiot.[2]

The squared modulus of the envelope functions for the first two heavy and the first light hole states are shown in Fig. 1. The first heavy and light hole states are localized mainly in the wider well, while the second heavy hole state is localized in the narrower well. The heavy hole states, which split from the same single quantum well state due to coupling, are thus spatially separated. By exciting all three states simultaneously using a short laser pulse, a wave packet consisting of the coherent superposition of these states can be formed. The wave packet has the form

$$\Psi(t) = e^{i\omega_{\mathrm{HH1}}t}C_{\mathrm{HH1}}\Psi_{\mathrm{HH1}} + e^{i\omega_{\mathrm{LH1}}t}C_{\mathrm{LH1}}\Psi_{\mathrm{LH1}} + e^{i\omega_{\mathrm{HH2}}t}C_{\mathrm{HH2}}\Psi_{\mathrm{HH2}}, \quad (1)$$

where there is a harmonic time dependence for each subband, the Ψ's are the envelope functions of the individual eigenstates of the superlattice, and the coefficients are proportional to the square root of the interband electron transition rates. The frequency associated with each subband is given by $E_n/\hbar$, where E_n is the energy separation between the bulk GaAs valence band edge and the given subband. Only transitions to the first electron state, $C1$, need to be considered due to the large energy separation between the first and second electron states and, for simplicity, the transition rates are normalized to the HH1–$C1$ transition rate. For comparison to experiment, the absolute value of the transition rates can be calculated using this theoretical formalism. The transition rate from a hole subband n to the first conduction subband is proportional to

$$W_n \propto \frac{I_1(n)}{\hbar\omega_1}\frac{|p_{cv}(n)^2|}{2m} m_r^{*3/2}E(n)^{1/2}, \quad (2)$$

where I_1 and ω_1 are the laser intensity at the transition energy and the center frequency of the laser and m_r^* is the

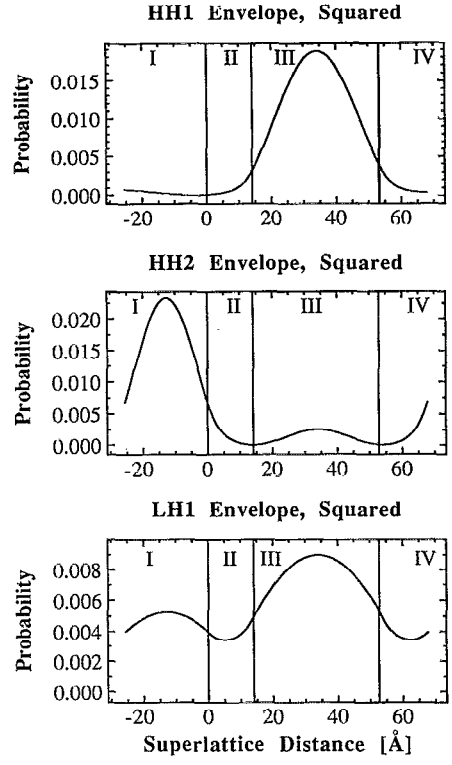

FIG. 1. Absolute square of each of the envelope functions, $\Psi^*\Psi$, of the three hole states combined to form the wave packet. Layers I and III are GaAs wells. Layers II and IV are $Al_{0.3}Ga_{0.7}As$ barriers. This material labeling scheme is used in all the figures. Envelope functions are continuous over a four-layer, superlattice period. The first well covers the negative distance values.

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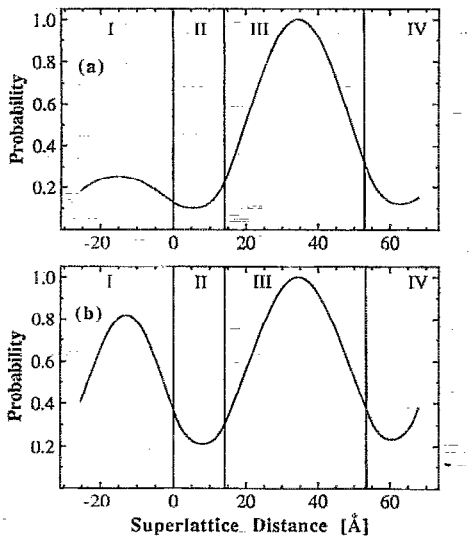

FIG. 2. (a) Probability function of a wave packet immediately after excitation. The wave packet is a linear combination of approximately equal amounts of each of the three hole subbands. It is well localized in the wider well. (b) The initial wave packet for different linear combinations of the hole states. The ratios of the HH1 to LH1 contribution and the HH1 to HH2 contribution are 1 to 2 and 1 to 3, respectively.

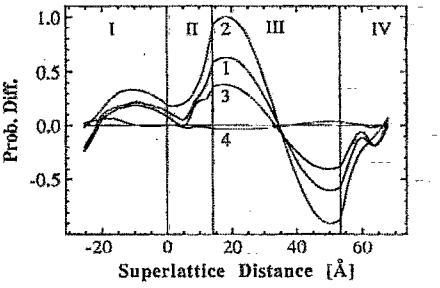

FIG. 3. Difference between the initial wave packet probability function and the probability function at later times, i.e., $\Psi(t)^*\Psi(t) - \Psi(0)^*\Psi(0)$. The wave packet is that illustrated in Fig. 2(a). The wave packet shifts to the left and then back to approximately its initial shape at $t = 333$ fs. The times are (1) $t = 83.5$ fs, (2) $t = 166$ fs, (3) $t = 249$ fs, (4) $t = 333$ fs.

reduced effective mass of the hole state being considered. The matrix element p_{cv} is between the first electron state and the hole state being considered and the laser light is linearly polarized. The polarization direction is normal to the superlattice growth direction. $E(n)$ is the transition energy minus the band-gap energy. The three-dimensional density of states, $m_r^{*3/2}E(n)^{1/2}$, was used in W_n. This approximation is reasonable because the coupling between the wells is strong. The effective masses were found from the theoretical formalism and follow directly from the band structure calculation. Exciton corrections to the energy were not used since exciton energies are similar for the three hole states.

The excitation of the wave packets is achieved using a short laser pulse which has the appropriate center wavelength and total pulse width in frequency space. The laser pulse is tuned to couple the three hole bands to the first conduction band. All three hole bands are coupled to the same conduction band because the short laser pulse has a large bandwidth in frequency space. Thus, by varying the width and center wavelength of the laser pulse, different amounts of each of the hole bands are combined to form the wave packet. For example, if a Gaussian laser pulse centered at 814 nm, with a pulse length of 65.8 fs, full width at the $1/e$ point of the pulse, is used to excite the first three hole states, the wave packet shown in Fig. 2(a) is created. The squared modulus of the wave packet envelope function is displayed. This wave packet has equal contributions from each of the hole states, to within 2%. The concentration of the wave packet in the wider well follows from the form of the HH1 and LH1 envelope functions, which are localized mainly in the wider well. The portion of the wave packet in the narrower well arises largely from the HH2 state. Although it is difficult to see on the plots, the wave packet peak in the wider well is narrower than the corresponding peak of either the HH1 or LH1 envelope function. This is a result of hole localization beyond that caused by the superlattice alone. This additional localization arises from the coherent combination of the hole eigenstates, that is, from the nature of the wave packet itself. It is analogous to the localization seen in Rydberg atomic wave packets.[3] In Fig. 2(b) we see the initial wave packet that results when a different laser pulse is used for excitation. In this case, the Gaussian laser pulse is centered at 810 nm and has a full width at the $1/e$ point of the pulse of 77 fs. The ratio of the HH1 contribution to the LH1 contribution is 1 to 2 while the HH1 to HH2 contribution ratio is 1 to 3. Thus, since the relative amount of the HH2 eigenstate in the wave packet is much greater than in the previous example, the probability is more balanced between the two wells. This follows directly from the form of the HH2 envelope function. From these two examples, it is seen that wave packets can vary greatly in form as a result of variations in the exciting laser pulse.

The time-dependent nature of the wave packet is illustrated in Fig. 3. The difference between the wave packet at some time after the excitation and the initial wave packet is shown. The wave packet considered is the one shown in Fig. 2(a), that is, the wave packet with approximately equal amounts of each of the hole eigenstates mixed together. The additional peak in the first material layer, leading to a slight loss of probability conservation, and the discontinuity in the wave functions between layers four and one that are observable in Fig. 3, arise from round-off error and numerical noise in the calculation. It is seen that the wave packet shifts in the wells in an almost periodic manner. If the energy separation between the three hole states were equal, the motion would be exactly periodic. The results shown in Fig. 3 occur at times equal to 1/4, 1/2, and 3/4 of one "period." The wave packet reaches its greatest deviation from its initial form at a time equal to 1/2 of a period, and then returns to approximately its initial form after a full period, about 333 fs. This motion then repeats itself except for small variations due to the unevenly spaced eigenstates. In this example, the deviation of the wave packet from its initial form is less than 10% of the total probability. However, by choosing different het-

erostructure systems and exciting them properly, a larger time-dependent modulation of the wave packet would be obtained. One method for obtaining a greater change in the wave packet with time would be to combine more eigenstates in the wave packet. This would require either a shorter excitation pulse for this system, which would be stretching experimental limits, or a system with subbands closer together in energy. Systems of this type are currently being studied.

It should be noted that this quasi-periodic motion of the wave packet will disappear as the holes relax out of their initially excited states. The wave packet will exist only when a coherence, a fixed phase relationship, is maintained between the three hole states. This coherence is established by the excitation of all three states with the same laser pulse. As the holes relax out of their initially excited states, this coherence is lost. The wave packet will break apart, causing all phenomena associated with the wave packet to cease. This breakup of the wave packet could be a useful tool in studying hole relaxation rates. The time scale over which the coherent nature of the hole dynamics disappears would be a direct measure of a hole's intraband phase relaxation rate, T_2. A measurement of the decay of the phase coherence of photoexcited heavy hole and light hole excitons has been made using a two-pulse self-diffracted transient grating technique.[4] In this work, quantum beats between a heavy hole exciton and a light hole exciton, arising from a coherent excitation of the two states, were observed. A similar technique could be used to measure the coherent wave packet dynamics and the eventual breakup of the wave packet. There are difficulties to be dealt with in any experiment intended to measure the wave packets. Rapid scattering mechanisms, such as carrier-carrier scattering, and similar phenomena could cause a destruction of the wave packet on an extremely fast time scale. Also, the existence of, effectively, a continuum of states in the plane of the well could cause the wave packet to disperse rapidly by allowing the holes to move in that plane. Due to the complexity of these effects, they are not dealt with in detail in the theory presented here, but could be for any actual experimental system. It should be noted that quantum beat phenomena have been described within a wave packet formalism in atomic physics.[5] A wave packet description applied to a superlattice or quantum well system could, thus, supply a more flexible and complete model of coherent hole dynamics than a quantum beat model alone. This added flexibility would contribute to a fuller understanding of hole relaxation rates and coherent hole dynamics in general.

In conclusion, we have presented a theoretical model for hole wave packets in a superlattice. The wave packets are formed by the simultaneous excitation of several hole subbands by a single laser pulse, thus establishing an initial coherence between the subbands. The initial form of the wave packet and its time-dependent behavior depend strongly on the excitation pulse and the band structure of the heterostructure being considered. Systems in which a large number of states are included in the wave packet would give a strong time-dependent modulation of the wave packet. Systems in which all the states are evenly spaced, such as parabolic quantum wells, are of particular interest since the time dependence would be periodic. Such systems are currently being studied. The wave packet model of coherent hole dynamics provides additional insight into quantum beat and related phenomena and could be a powerful tool in determining hole relaxation rates.

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