

Exciton coherence in symmetric coupled quantum wells and dots

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I calculate exciton states of laterally confined, symmetric, double-quantum-well structures in electric fields to study interwell tunneling and electron-hole Coulomb correlation effects. When correlation is dominant, the zero-field exciton ground state is a *coherent* state of direct excitons in *both wells* with enhanced oscillator strength. For increasing field, the ground state becomes direct in a *single well* and then indirect, and exciton peaks in the absorption spectra evolve asymmetrically.

Exciton states of quantum well systems are of great current interest. Exciton binding is enhanced by confinement, so excitons persist to room temperature, and large modulation of exciton energies and oscillator strengths by an applied field occurs without field-induced ionization. Exciton states in double-well structures depend critically upon electron-hole Coulomb correlation and single-particle interwell tunneling, which is important when single-particle states of one well are near resonance with states in the other well. In asymmetric double-well structures, excitons are direct or indirect. An exciton that is direct at low fields remains direct after a level crossing of single-particle states induced by increasing the field until the energy gain from intrawell Coulomb binding is less than the gain from interwell tunneling to an indirect state. In symmetric double-well (SDW) structures, interwell tunneling is resonant for electrons and holes at zero field. Excitons in SDW can have large mixing of the direct and indirect exciton states.

Extensive effort has been made to understand experimentally¹⁻⁶ and theoretically^{5,7-10} excitons in SDW. Excitons are easiest to understand when the wells are strongly coupled so that interwell tunneling dominates the Coulomb correlation.² In this limit, exciton states are electron-hole single-particle pair states. The single-particle states are symmetric and antisymmetric coupled-well states (see Fig. 1). At zero field, the exciton ground state is the symmetric exciton, made from symmetric electron and hole states $|s\rangle_{\text{ex}} = |s\rangle_e |s\rangle_h$, with a finite oscillator strength. The antisymmetric exciton, made from the antisymmetric electron and hole states $|a\rangle_{\text{ex}} = |a\rangle_e |a\rangle_h$, has higher energy but the same oscillator strength as the symmetric exciton. Forbidden excitons, made from one symmetric and one antisymmetric state of the electron and hole pair, occur at energies between the symmetric and antisymmetric excitons. At finite fields, Coulomb effects become more important and

mix these four states. At high fields the excitons become direct or indirect.

Coulomb effects are enhanced relative to interwell tunneling when the barrier between the two wells is thick so that the splitting between symmetric and antisymmetric single-particle states is small or when lateral confinement is imposed, by forming quantum dots or by applying a magnetic field perpendicular to the wells, to increase the electron-hole lateral overlap without changing the interwell tunneling.^{11,12} In this paper I calculate the exciton states of laterally confined SDW in an applied field to study these states when Coulomb effects are important. The key result of this paper is that the zero-field exciton states are coherent or incoherent states when Coulomb correlation is important. The exciton ground state in this limit is a coherent state of the direct excitons in both wells, $|cd\rangle_{\text{ex}} = (1/\sqrt{2})(|1\rangle_{\text{ex}} + |2\rangle_{\text{ex}})$. The incoherent direct state $|incd\rangle_{\text{ex}} = (1/\sqrt{2})(|1\rangle_{\text{ex}} - |2\rangle_{\text{ex}})$, has higher energy. This interwell coherence is analogous to the volume coherence of bulk excitons and the two-dimensional coherence of excitons in isolated quantum wells. With increasing field, the interwell coherence weakens until the ground state is direct in one well. At high fields the ground state is indirect. This identification of the exciton states is consistent with results presented by Dignam and Sipe.¹⁰ However, they did not discuss the coherence of the exciton states. It is critical that this coherence be clearly established. The coherent direct exciton has all of the oscillator strength of the direct excitons in the two wells while the incoherent exciton is forbidden. Large modulation of exciton properties is possible by applying a field to break the coherence. In this paper I calculate absorption spectra to show how interwell coherence effects can be observed.

I determine the properties of laterally confined excitons in SDW by use of the same model used to study laterally confined, asymmetric, double-well structures.^{11,12} I assume that the two wells are sufficiently narrow that the electron and hole occupy the lowest-energy single-particle subbands in each well. All four configurations for the electron and hole in the two wells can contribute to the exciton states. Typically, the length L of the lateral confinement (dot size or cyclotron radius) is much greater than the well width, and the intrawell motion of the electron-hole pair in the plane of each well is correlated. Exciton states must be modeled carefully to account for intrawell and interwell correlation and subband mix-

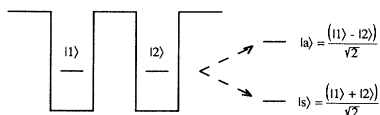


FIG. 1. Single-particle level of each well in an SDW and the level splitting to form symmetric and antisymmetric single-particle states.

ing when Coulomb correlation and interwell tunneling are both important.¹⁰ The correlation may be different for each configuration of the electron and hole in the two wells. I use the following wave function for low-energy exciton s states,^{10–13}

$$\Psi_{\text{ex}} = C_e(x_e, y_e) C_h(x_h, y_h) S(x_e, y_e, x_h, y_h; z_e, z_h), \quad (1)$$

where $C_{e(h)}$ is the single-particle electron (hole) ground-state wave function for lateral (x and y) confinement, and S accounts for electron-hole correlation in the lateral direction and the tunneling between wells (the z direction)

$$S(x_e, y_e, x_h, y_h; z_e, z_h) = \sum_{i,j=1,2} \varphi_{ei}(z_e) \varphi_{hj}(z_h) \chi_{ij}(x_e, y_e, x_h, y_h). \quad (2)$$

$\varphi_{e(h)i}$ is the electron (hole) ground state for well i and χ_{ij} is the lateral correlation when the electron is in well i and the hole in well j . Implicit in Eq. (1) is the simplifying assumption that the lateral confinement is the same in the two wells and in the barriers. For specific calculations, I model the lateral confinement for square quantum dots with infinite lateral confining potentials.¹³ For the correlated lateral motion, I use

$$\chi_{ij}(x_e, y_e, x_h, y_h) = \sum_n c_{ij,n} \exp\{-a_n[(x_e - x_h)^2 + (y_e - y_h)^2]\}. \quad (3)$$

Accurate energies and wave functions are obtained by use of 5–10 Gaussians.¹³

The SDW is modeled with an infinite barrier, the first well, the middle barrier with height determined by the band discontinuity, the second well, and another infinite barrier. A flat band profile is assumed for each region. For a bias applied between the two wells, I assume that the bands remain flat with the first well band edge fixed, the middle barrier shifted by half the bias, and the second well band edge shifted by the bias. These assumptions allow analytic wave functions for the well states to be used. The ground state $\varphi_{e(h)i}$ for well i is found by assuming that the middle barrier extends across the other well. Accurate states for the two lowest electron and two lowest hole one-dimensional levels in the coupled wells are obtained when $\varphi_{e(h)1}$ and $\varphi_{e(h)2}$ are the basis functions that are coupled by interwell tunneling. The effective-mass Hamiltonian is solved with the wave function defined by Eqs. (1)–(3). The electron-hole interaction is the screened Coulomb interaction. GaAs effective masses and dielectric constant are used for the wells and barriers. The barriers are 250 meV for the conduction band and 100 meV for the valence band corresponding to $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$. In this paper I consider SDW with 10-nm wells. Calculations done for SDW with thinner wells show reduced Coulomb effects because carriers leak out of thin wells more.

The character of exciton states in SDW is revealed by considering how the heavy-hole exciton ground-state oscillator strength (normalized to be near unity for an unconfined two-dimensional direct exciton, and shown in Fig. 2) varies with the applied bias between the wells. When Coulomb effects are dominant (wide tunnel bar-

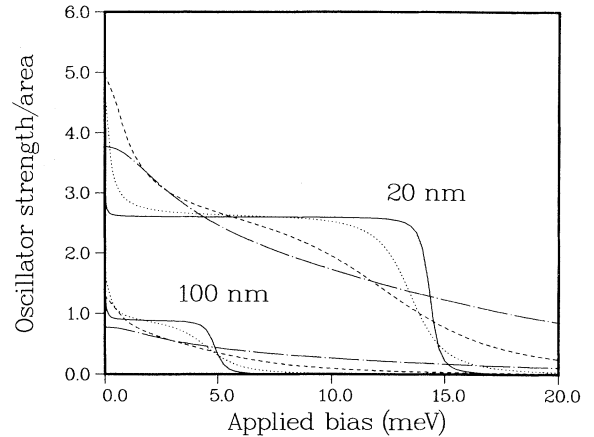


FIG. 2. Exciton ground-state oscillator strength in a laterally confined SDW (10-nm wells) normalized by dot area L^2 . Dependence on applied bias for $L = 20$ and 100 nm and tunnel barrier width of 1 (dashed-dotted line), 3 (dashed line), 5 (dotted line), and 7 nm (solid line).

riers and strong lateral confinement), the exciton ground state changes abruptly twice as the bias is varied. At high bias, the ground state is indirect. At intermediate bias, the ground state is the lowest-energy direct exciton. At zero bias, the ground state is the coherent superposition of the direct excitons in both wells with nearly twice the oscillator strength of a direct exciton, f_d . When Coulomb effects are dominant, this transition occurs rapidly and the sharp rise in oscillator strength to nearly $2f_d$ is not clearly visible in Fig. 2. Coherence persists, with the oscillator strength greater than f_d , for a wide range of biases, tunneling barriers, and lateral widths. However, the abrupt transitions broaden and the zero-bias oscillator strengths are less than $2f_d$ when interwell tunneling becomes important. Results are shown in Fig. 2 for 100-nm lateral confinement. At this dot size, lateral confinement has a negligible effect on the exciton ground

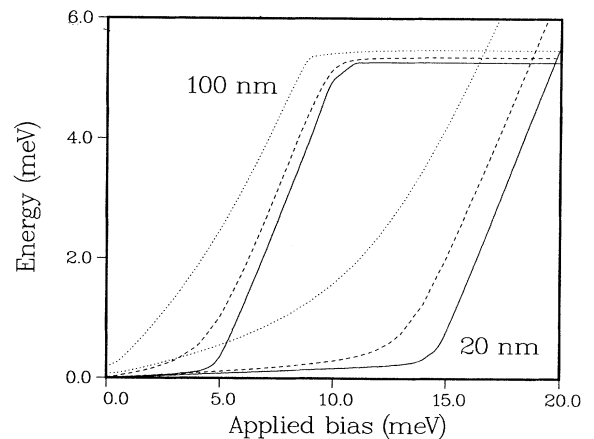


FIG. 3. Energy splitting between the ground and first excited exciton in a laterally confined SDW (10-nm wells). Dependence on applied bias for $L = 20$ and 100 nm and tunnel barrier width of 3 (dotted line), 5 (dashed line), and 7 nm (solid line).

state and only a weak effect on excited states. Thus, coherence effects are present even when lateral confinement is unimportant. Coherence is critical for enhancing oscillator strengths. For example, the ground-state oscillator strength in a SDW with a 3-nm tunnel barrier, 20-nm lateral confinement, and 2-meV applied bias is enhanced by 40% even though the ground state has more than 90% probability of being in the lowest direct state.

The energy splitting between the first excited and ground states is shown in Fig. 3. At zero field, the first excited state is the incoherent superposition of the two direct states, is nearly degenerate with the ground state, and is forbidden when Coulomb effects are dominant. The sharp increase in level splitting with increasing applied bias occurs when the ground state becomes indirect while the first excited state remains direct. A second transition, shown in Fig. 3 for SDW with 100-nm lateral confinement, occurs when the first excited state becomes an excited indirect state. Increasing the lateral confinement increases the bias needed for direct-to-indirect transitions.¹²

To determine how coherence effects can be observed, I calculate the absorption spectra for SDW with 3-nm tunnel barriers. The spectra include contributions from the four lowest-energy heavy-hole excitons in the SDW with the symmetry allowed by Eqs. (1)–(3). Each exciton peak is broadened by 1 meV. Contributions from light-hole excitons will give similar results. Spectra found by using uncorrelated single-particle pair states for the excitons are shown in Fig. 4. In this paper, I focus on features of the spectra that reveal the Coulomb correlation and coherence. Other details will be discussed elsewhere. The key feature of the spectra determined without Coulomb effects is the symmetry between the

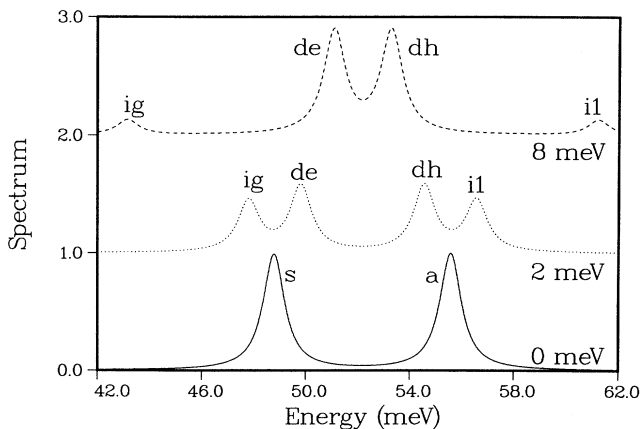


FIG. 4. Absorption spectra of a laterally confined SDW (10-nm wells, 3-nm tunneling barrier, and 100-nm lateral width) for applied bias of 0, 2, and 8 meV. Exciton states are found without Coulomb effects. The energy is relative to the band gap. Peaks are for excitons that are symmetric (*s*), antisymmetric (*a*), direct in the well with the lower-energy electron or hole state (*de, dh*), and indirect ground and first excited state (*ig, il*).

low- and high-energy peaks. At zero bias, the symmetric and antisymmetric exciton have equal absorption. At high bias, the direct excitons have the same absorption. Spectra found with and without Coulomb effects are similar at high bias. When Coulomb effects are included and dominant (spectra are shown in Fig. 5 for 20-nm lateral confinement; similar results are found for 100-nm lateral confinement), the symmetry of the peaks is broken at low bias. At zero bias, absorption is concentrated in one peak, with nearly twice the magnitude of either absorption peak at high bias, because the coherent direct state has nearly all the oscillator strength. The weak high-energy peak due to the coherent indirect exciton is observable because interwell tunneling mixes in a component of the direct states. When bias is applied, absorption by the coherent direct state weakens while the incoherent direct state, which is nearly degenerate with the coherent direct state (as shown in Fig. 3), becomes allowed. The asymmetric splitting of the main peak with increasing bias is a key signature of interwell coherence. If the excitons were strictly direct in one well or the other at zero bias, then all the absorption would still be in one main peak at zero bias, but the peak would split symmetrically with increasing bias.

In conclusion, exciton states in symmetric double-well structures have interwell coherence when Coulomb effects are dominant. This coherence concentrates the oscillator strength in the ground-state exciton and makes other transitions forbidden. Peaks in absorption spectra evolve asymmetrically with increasing bias due to interwell coherence. The asymmetry may be difficult to observe because the near degeneracy of the ground and first excited exciton states must be split by increasing the applied bias without destroying the coherence. Interestingly, photoluminescence exciton spectra of SDW obtained by Chen *et al.*² show asymmetric peak splittings with increasing applied bias that can be explained as coherence

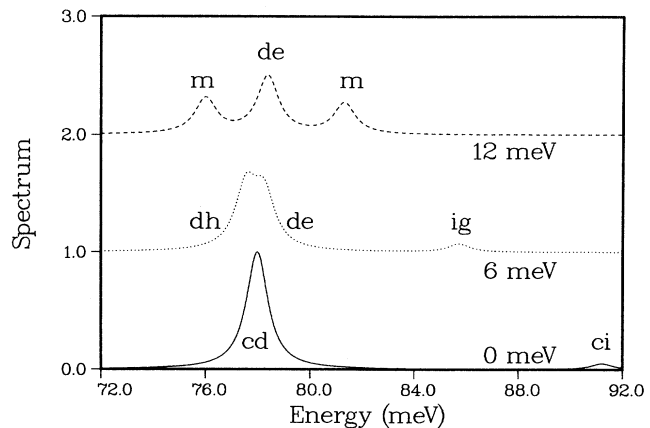


FIG. 5. Absorption spectra of a laterally confined SDW (same structure as in Fig. 4, but a 20-nm-wide dot) for applied bias of 0, 6, and 12 meV. Exciton states are found including Coulomb effects. Coherent direct and indirect (*cd, ci*) exciton peaks are shown. Peaks *m* are a mixture of *dh* and indirect states. Other peaks are labeled as in Fig. 4.

effects. However, these structures show stronger asymmetry than expected, especially for SDW with thin barriers and wells. Additional studies should be done to clearly identify interwell coherence. Photoluminescence excitation studies of SDW designed with thick barriers

and wells to enhance Coulomb effects, and studies with low-temperature time-resolved spectroscopy,⁵ to probe the ground-state dynamics, and nonlinear spectroscopy, where coherence effects should be more important, are needed.

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