Excitonic effects can lead to decreased intersubband oscillator strength

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We study intersubband transitions of an electron in a quantum well, in the presence of a hole. When the energy of one subband of the electron approaches the top of the well, the electron-hole attraction strongly influences the electronic state. We show that these excitonic effects lead to a reduction in oscillator strength for an intersubband transition of given energy. This finding should be considered in the design of quantum well devices, such as recent devices that produce midinfrared radiation from near-infrared laser structures.

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In recent years, there has been extensive and sustained interest in intersubband transitions in quantum wells (QW's).[1] These transitions are important both because of their role in fundamental research on semiconductors[2–5] and because of their potential technological importance in devices such as quantum well infrared photodetectors[6] and quantum cascade lasers.[7] In this work, the intersubband oscillator strength plays an essential role in determining QW behavior. In particular, a large intersubband oscillator strength is often favorable for these devices. This gives rise to an interesting theoretical question: one would like to know how to optimize a QW structure so that it possesses an intersubband transition of desired energy and maximum oscillator strength. The obvious approach is to increase the well width and depth, keeping the intersubband transition energy constant, to make the oscillator strength near unity (see Fig. 1). Unfortunately, this scheme is impractical since, in general, the well depth cannot be tuned at will; aside from the materials issues, devices often require the final state to be close in energy to the top of the well so that carriers can enter or exit.

Alternatively, one might imagine taking advantage of excitonic effects to enhance oscillator strength, and here we investigate this possibility. Consider, for instance, recent devices that produce midinfrared (MIR) radiation from near-infrared (NIR) QW lasers.[8,9] In these devices, an electron emits MIR radiation by making an intersubband transition between two QW conduction-band states. The electron then vacates the final QW conduction-band state by making a stimulated interband transition that destroys a hole in the QW valence band and produces NIR radiation. Since the initial intersubband transition proceeds in the presence of the hole, excitonic effects can influence the intersubband oscillator strength. This is not surprising, and we expect excitonic effects to be pronounced in many materials of contemporary interest such as in III–V semiconductors such as GaN and II–VI semiconductors such as ZnSe.[10] What is surprising is the sign of the result. We show that the excitonic transition can exhibit *reduced* oscillator strength in comparison to a transition of the same energy with no excitonic effects. This is certainly not obvious or intuitive; in fact, the opposite situation can occur when wells get sufficiently deep—excitonic effects can then lead to slightly *increased*, rather than decreased, intersubband oscillator strength. By modeling these effects, we develop here a physical understanding that is germane to quantum dot (QD) lasers involving intersubband transitions in relatively flat QD's,[11,12] interband Raman scattering,[13] some contemporary studies of semiconductor QW physics,[4,5] and, as mentioned above, MIR emission from NIR QW lasers.[8,9]

The calculation begins with the familiar model Hamiltonian for an exciton[14] in a QW,

$$H=-\frac{\hbar^2}{2m_e^*}\nabla_e^2-\frac{\hbar^2}{2m_h^*}\nabla_h^2-\frac{e^2}{\kappa|\vec{r}_e-\vec{r}_h|}+V(z_e)+V(z_h). \tag{1}$$

Here, m_e^* and m_h^* are the electron and hole effective masses, $\vec{r}_e$ and $\vec{r}_h$ are the three-dimensional position vectors of the electron and hole, and $V(z)$ is a potential well of depth V_o stretching from $-a$ to a in the z direction. The electron and hole are assumed to occupy a single parabolic band each, which is not realistic but should not influence our conclusions qualitatively.

To find the eigenstates of H, we separate out the z dependence of the Coulomb interaction,[15] writing $H=H_o+\delta H$ where,

$$H_o=-\frac{\hbar^2}{2m_e^*}\nabla_e^2-\frac{\hbar^2}{2m_h^*}\nabla_h^2-\frac{e^2}{\kappa\sqrt{(x_e-x_h)^2+(y_e-y_h)^2}}+V(z_e)+V(z_h), \tag{2}$$

and

$$\delta H=-\frac{e^2}{\kappa|\vec{r}_e-\vec{r}_h|}+\frac{e^2}{\kappa\sqrt{(x_e-x_h)^2+(y_e-y_h)^2}}. \tag{3}$$

In order to treat excitons in QW's as two-dimensional objects,[14] one neglects δH. This should be reasonable provided the hole and the electron are sufficiently well bound in the QW. Then, the separability of H_o permits us to write its eigenstates $|N,M,\vec{K},n_e,n_h\rangle$ as $\Psi_{N,M,\vec{K},n_e,n_h}(\vec{r}_e,\vec{r}_h)=\chi_{N,M,\vec{K}}(x_e,y_e,x_h,y_h)\phi_{n_e}(z_e)\phi_{n_h}(z_h)$ in which the motion parallel to the QW plane is decoupled from the motion perpendicular to it. The energy of the state $|N,M,\vec{K},n_e,n_h\rangle$ is termed $E_{N,M,\vec{K},n_e,n_h}$. The χ are two-dimensional exciton

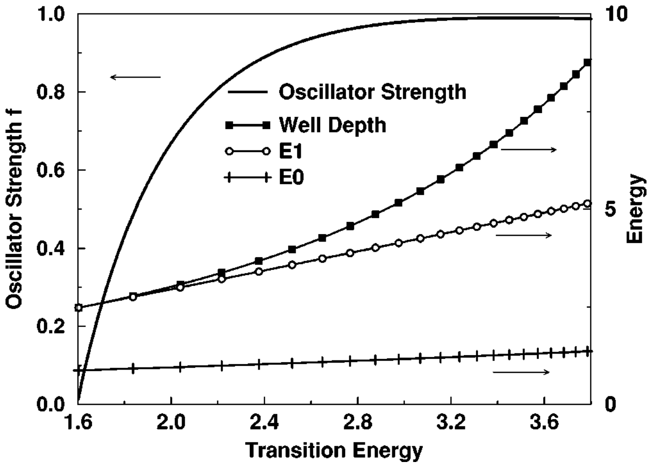

FIG. 1. Plot of intersubband oscillator strength as a function of transition energy in the two-dimensional exciton approximation ($H=H_o$). Well depth, excited-state energy E_1, and ground-state energy E_0 are also shown. Vertical distance between well depth and E_1 gives binding energy of excited state. Energies are measured in units of $\hbar^2/2m_e^*a^2$. The constant factor ϵ_z has been set to unity.

eigenfunctions[14] and the ϕ are eigenstates of a one-dimensional square potential well.[16] In experimental QW's, the square potential-well depth V_o often lies between $(\pi/2)^2$ and $(\pi)^2$ in units of $\hbar^2/2m_e^*a^2$, so that there are exactly two bound electronic states $\phi_{n_e}(z_e)$ with $n_e=0,1$. The energies of these two states are E_0 and E_1 in units of $\hbar^2/2m_e^*a^2$.

Electronic transitions between eigenstates of H_o are governed by the oscillator strength

$$f=2\frac{|\langle N',M',\vec{K}',n_e',n_h'|\vec{\epsilon}\cdot\vec{p}|N,M,\vec{K},n_e,n_h\rangle|^2}{m_e^*(E_{N',M',\vec{K}',n_e',n_h'}-E_{N,M,\vec{K},n_e,n_h})}. \quad (4)$$

Here, $\vec{\epsilon}$ is the polarization vector of light causing the transition (we neglect the momentum of this light) and $\vec{p}$ is the momentum of the electron. We focus on the experimentally relevant case of electronic intersubband transitions for which $n_e'=1$ and $n_e=0, n_h'=n_h=0$. In this case, the dipole matrix element appearing in f becomes

$$\begin{aligned}\langle N,M,\vec{K},1,0|\epsilon_z p_z|N,M,\vec{K},0,0\rangle\delta_{N',N},\delta_{M',M},\delta_{\vec{K}',\vec{K}}\\ =-\epsilon_z\frac{2\sqrt{E_0E_1}}{E_1-E_0}\frac{1}{\sqrt{1+1/(V_o-E_0)^{1/2}}}\\ \times\frac{1}{\sqrt{1+1/(V_o-E_1)^{1/2}}}\frac{\hbar}{i}\frac{1}{a}\delta_{N',N},\delta_{M',M},\delta_{\vec{K}',\vec{K}}\end{aligned} \quad (5)$$

since only the z component of the momentum can connect the initial and final states. Note the restrictive selection rules that require that the excitonic state remain unchanged in the transition and imply that the oscillator strength of an intersubband transition is independent of excitonic state. In Fig. 1, we plot the oscillator strength of an intersubband transition for the case $H=H_o$ as a function of transition energy.

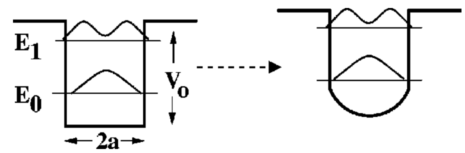

FIG. 2. Schematic of the effective potential experienced by the electron. The electron-hole interaction supplements the QW confinement and introduces parabolic curvature into the potential. The size of the change is dependent upon the electron-hole overlap $|\chi_{N,M,\vec{K}}(0)|^2$, and it vanishes in our model if this overlap vanishes.

To see how excitonic effects influence this picture, we calculate the effect of δH (3) on the oscillator strength. The form of δH (3) is cumbersome for calculation, so we approximate it as

$$\begin{aligned}\delta H&\equiv-\frac{e^2}{\kappa|\vec{r}_e-\vec{r}_h|}+\frac{e^2}{\kappa\sqrt{(x_e-x_h)^2+(y_e-y_h)^2}}\\ &=\frac{e^2}{\kappa}\int\frac{d^2k}{(2\pi)^2}\exp\{i\vec{k}\cdot[(x_e-x_h)\hat{x}+(y_e-y_h)\hat{y}]\}\\ &\quad\times\frac{2\pi}{k}(1-e^{-k|z_e-z_h|})\\ &\approx\frac{e^2}{\kappa}\int\frac{d^2k}{(2\pi)}\exp\{i\vec{k}\cdot[(x_e-x_h)\hat{x}+(y_e-y_h)\hat{y}]\}|z_e-z_h|\\ &=\frac{e^2}{\kappa}|z_e-z_h|(2\pi)\delta^{(2)}[(x_e-x_h)\hat{x}+(y_e-y_h)\hat{y}].\end{aligned} \quad (6)$$

We have taken the small $|z_e-z_h|$ limit of the Fourier transform of δH, which is appropriate when the QW width is much less than the exciton radius.

In our case of interest $n_h=0$, we find

$$\begin{aligned}&\langle N,M,\vec{K},1,0|\delta H|N,M,\vec{K},n_e',0\rangle\\ &\approx\frac{e^2}{\kappa}(2\pi)|\chi_{N,M,\vec{K}}(0)|^2\int dz_e\phi_1(z_e)^*\phi_{n_e'}(z_e)[z_e^2/2a\\ &\quad+a/2-a/\pi^2-a\cos(\pi z_e/a)/\pi^2]\\ &\approx\frac{e^2}{\kappa}(2\pi)|\chi_{N,M,\vec{K}}(0)|^2\int dz_e\phi_1(z_e)^*\phi_{n_e'}(z_e)\\ &\quad\times\left\{\begin{matrix}(z_e^2/2a+a/2) & |z_e|<a\\ a, & |z_e|\geqslant a\end{matrix}\right\},\end{aligned} \quad (7)$$

where we make the abbreviation $|\chi_{N,M,\vec{K}}(0)|^2$ for the density $|\chi_{N,M,\vec{K}}(x_e,y_e;x_h,y_h)|^2$ when the electron and hole sit on top of one another. (This density satisfies $|\chi_{N,M,\vec{K}}(0)|^2=\delta_{M,0}/[\pi a_B^2(N+1/2)^3]$ where $a_B=\kappa\hbar^2(1/m_e^*+1/m_h^*)/e^2\equiv\kappa\hbar^2/\mu^*e^2$ is the exciton Bohr radius[14]). Numerical studies[17] confirm that the physically plausible approximations (6) and (7) lead to qualitatively correct results. In addition to being reasonably accurate and analytically convenient, the approximation (7) gives a clear physical picture. It shows that the Coulomb attraction described by δH supplements the QW confining potential experienced by the electron. As shown schematically in Fig. 2, the net potential experienced by the electron roughly changes from a square well to a well with a parabolic bottom whose curvature depends upon the excitonic state.

To compute the influence of δH on the intersubband oscillator strength, we employ first-order perturbation theory. The matrix elements (7) imply the following shift in the transition energy that appears in the denominator of f [Eq. (4)]:

$$\langle N,M,\vec{K},1,0|\delta H|N,M,\vec{K},1,0\rangle-\langle N,M,\vec{K},0,0|\delta H|N,M,\vec{K},0,0\rangle \approx \frac{e^2}{\kappa a_B}\frac{a}{a_B}a_B^2(2\pi)|\chi_{N,M,\vec{K}}(0)|^2\left(\frac{2/3+1/\sqrt{V_o-E_1}-1/4E_1+1/2V_o-\sqrt{V_o-E_1}/4E_1V_o}{1+1/\sqrt{V_o-E_1}} - \frac{2/3+1/\sqrt{V_o-E_0}-1/4E_0+1/2V_o-\sqrt{V_o-E_0}/4E_0V_o}{1+1/\sqrt{V_o-E_0}}\right). \tag{8}$$

The shift in energy is positive since the parabolic potential in Fig. 2 affects the excited state, which explores the edges of the well, more than the ground state. To compute the effect of δH on the intersubband oscillator strength, we also need to evaluate its effect on the dipole matrix elements. The first-order shifts in the wave functions add the following terms to the dipole matrix element:

$$\langle N,M,\vec{K},1,0|\delta H \times\sum\frac{|N',M',\vec{K}',n_e',n_h'\rangle\langle N',M',\vec{K}',n_e',n_h'|}{E_{N,M,\vec{K},1,0}-E_{N',M',\vec{K}',n_e',n_h'}} \times p_z|N,M,\vec{K},0,0\rangle \tag{9}$$

and

$$\langle N,M,\vec{K},1,0|p_z \times\sum\frac{|N',M',\vec{K}',n_e',n_h'\rangle\langle N',M',\vec{K}',n_e',n_h'|}{E_{N,M,\vec{K},0,0}-E_{N',M',\vec{K}',n_e',n_h'}} \times\delta H|N,M,\vec{K},0,0\rangle. \tag{10}$$

Note that the operators p_x and p_y still make no contribution to our intersubband transition since the matrix elements of δH vanish between states with $n_e=0$ and $n_e=1$. The sums (9) and (10) should proceed over $N',M',\vec{K}',n_e'$, and n_h', with the term that would lead to a zero denominator excluded from each sum. Because the operator p_z cannot change the state of the excitonic wave function or the state of the hole in the z direction, though, the sums actually need only proceed over n_e'. We, therefore, need to compute the sum

$$\sum_{n_e'\neq n_e}\frac{\phi_{n_e'}(z_e)\phi^*_{n_e'}(z_e')}{E_{n_e}-E_{n_e'}} = \lim_{E\to E_{n_e}} G(z_e,z_e',E)-\phi_{n_e}(z_e)\int dz\,\phi^*_{n_e}(z)G(z,z_e',E), \tag{11}$$

which we have related to the Green's function of a one-dimensional finite well. This Green's function can be obtained using the usual methods of one-dimensional differential equations.

The result for Eq. (9) is found to equal

$$R\left[\frac{\sqrt{E_0}}{E_1-E_0}\frac{1}{\sqrt{1+1/(V_o-E_0)^{1/2}}}\frac{1}{\sqrt{1+1/(V_o-E_1)^{1/2}}}\right] \times\left[-\frac{\cos(\sqrt{E_1}-\sqrt{E_0})}{(\sqrt{E_1}-\sqrt{E_0})^2}+\frac{\cos(\sqrt{E_1}+\sqrt{E_0})}{(\sqrt{E_1}+\sqrt{E_0})^2}+\frac{\sin(\sqrt{E_1}-\sqrt{E_0})}{(\sqrt{E_1}-\sqrt{E_0})^3}-\frac{\sin(\sqrt{E_1}+\sqrt{E_0})}{(\sqrt{E_1}+\sqrt{E_0})^3}-\frac{\sin(2\sqrt{E_1})}{1+1/\sqrt{V_o-E_1}}\left(\frac{1}{4E_1}-\frac{3}{16E_1^2}\right) -\frac{3\cos(2\sqrt{E_1})}{(2\sqrt{E_1})^3(1+1/\sqrt{V_o-E_1})}+\frac{1}{1+1/\sqrt{V_o-E_1}}\left(-\frac{1}{3}-\frac{1}{4E_1}+\frac{1}{2V_o}-\frac{\sqrt{V_o-E_1}}{4E_1V_o}\right)\left(\frac{2\sqrt{V_o-E_0}\sqrt{E_1}}{V_o(\sqrt{V_o-E_1}+\sqrt{V_o-E_0})} +\frac{\sin(\sqrt{E_1}-\sqrt{E_0})}{(\sqrt{E_1}-\sqrt{E_0})}-\frac{\sin(\sqrt{E_1}+\sqrt{E_0})}{(\sqrt{E_1}+\sqrt{E_0})}-\frac{1}{2\sqrt{E_1}}+\frac{\sqrt{E_1}}{2(V_o-E_1)(\sqrt{V_o-E_1}+1)}\right)\right], \tag{12}$$

where

$$R\equiv\left[\frac{\hbar}{i}\frac{1}{a}\frac{2m_e^*a^2}{\hbar^2}\frac{e^2}{\kappa a_B}\frac{a}{a_B}a_B^2(2\pi)|\chi_{N,M,\vec{K}}(0)|^2\right]$$

is a dimensionless measure of the importance of excitonic corrections. The result for Eq. (10) is

$$\langle N,M,\vec{K},1,0|p_z\sum\frac{|N,M,\vec{K},n_e',0\rangle\langle N,M,\vec{K},n_e',0|}{E_{N,M,\vec{K},1,0}-E_{N,M,\vec{K},n_e',0}}\delta H|N,M,\vec{K},0,0$$

$$=-R\left[\frac{\sqrt{E_1}}{E_1-E_0}\frac{1}{\sqrt{1+1/(V_o-E_0)^{1/2}}}\frac{1}{\sqrt{1+1/(V_o-E_1)^{1/2}}}\right]$$

$$\times\left[-\frac{\cos(\sqrt{E_1}-\sqrt{E_0})}{(\sqrt{E_1}-\sqrt{E_0})^2}-\frac{\cos(\sqrt{E_1}+\sqrt{E_0})}{(\sqrt{E_1}+\sqrt{E_0})^2}+\frac{\sin(\sqrt{E_1}-\sqrt{E_0})}{(\sqrt{E_1}-\sqrt{E_0})^3}+\frac{\sin(\sqrt{E_1}+\sqrt{E_0})}{(\sqrt{E_1}+\sqrt{E_0})^3}-\frac{3\cos(2\sqrt{E_0})}{(2\sqrt{E_0})^2(1+1/\sqrt{V_o-E_0})}\right.$$

$$-\frac{\sin(2\sqrt{E_0})}{1+1/\sqrt{V_o-E_0}}\left(\frac{1}{4E_0}-\frac{3}{16E_0^2}\right)+\frac{1}{1+1/\sqrt{V_o-E_0}}\left(-\frac{1}{3}-\frac{1}{4E_0}+\frac{1}{2V_o}-\frac{\sqrt{V_o-E_0}}{4E_0V_o}\right)$$

$$\left.\times\left(-\frac{2\sqrt{V_o-E_1}\sqrt{E_0}}{V_o(\sqrt{V_o-E_1}+\sqrt{V_o-E_0})}+\frac{\sin(\sqrt{E_1}-\sqrt{E_0})}{(\sqrt{E_1}-\sqrt{E_0})}+\frac{\sin(\sqrt{E_1}+\sqrt{E_0})}{(\sqrt{E_1}+\sqrt{E_0})}+\frac{1}{2\sqrt{E_1}}-\frac{\sqrt{E_0}}{2(V_o-E_0)(\sqrt{V_o-E_0}+1)}\right)\right].\quad(13)$$

These shifts in the dipole matrix elements, together with the energy shift (8) enable us to compute the excitonic correction to the oscillator strength (4). The fractional change in the oscillator strength $\delta f/f$ is plotted in Fig. 3 as a function of transition energy. Several curves are shown, with various values of the parameter R defined above. In terms of the hydrogen atom radius a_o and the electron mass m_e, we find $R=4(m_e^*\mu^{*2}/\kappa^3 m_e^3)(a/a_o)^3[\delta_{M,0}/(N+1/2)]^3$. For a 40-Å-wide GaAs QW ($a=20$ Å), for instance, $R=0.2$, while $R=0.7$ for a 20-Å-wide ZnSe QW and $R=1.1$ for a 20 Å wide GaN well.

The cause of the negative shift shown in Fig. 3 is as follows. If we fix the well depth, we find that the supplemental confinement of the electron by its interaction with the hole (Fig. 2) produces (i) an increase in the intersubband transition energy and (ii) an increase in the intersubband oscillator strength. However, effect (i) is relatively large and effect (ii) is relatively small. To understand Fig. 3, we compare this to an intersubband transition of the same energy but with no hole present. Such a transition would have to occur in a QW with a larger V_o, to compensate for excitonic effect (i). But, as is evident from Fig. 1, a QW with a larger V_o would have a significantly larger oscillator strength, surpassing the small excitonic effect (ii). Thus, for a given energy transition, one gets a higher oscillator strength for a well without excitonic corrections than for a well with excitonic corrections. Essentially, adding an excitonic effective potential to a shallow square well does a good job of increasing transition energy but a bad job of enhancing oscillator strength. To enhance intersubband oscillator strength in a shallow well, it is better to increase the well depth. The effect is particularly pronounced when the transition energy is small, and one is dealing with a shallow well whose first excited state is approaching the well top. (The situation can change for deep wells, where adding an excitonic effective potential can do a slightly better job of enhancing oscillator strength than simply increasing the well depth further.)

In summary, intersubband transition oscillator strength at fixed energy can actually decrease as a result of the electron-hole interaction. This result is interesting in its own right, and it needs to be considered in QW's designs for fundamental and applied science.

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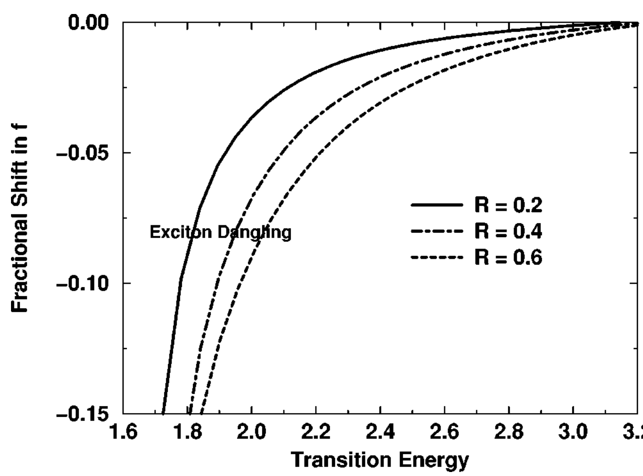

FIG. 3. Plot of fractional change in intersubband oscillator strength $\delta f/f$ as a function of intersubband transition energy. The energy is measured in units of $\hbar^2/2m_e^*a^2$. Curves are given for several values of the dimensionless quantity $R\equiv(2m_e^*a^2/\hbar^2)(e^2/\kappa a_B)(a/a_B)a_B^2(2\pi)|\chi_{N,M,\vec{K}}(0)|^2$. The perturbative approach of this paper is most reliable when the well is deep enough to prevent the exciton from "dangling."

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[17] We have checked the accuracy of these approximations by comparing (Ref. 17) the approximate potential in Eq. (7) to the exact potential that results from Eq. (3). When the excitonic wave function has $M=0$, the approximate potential in Eq. (7) is certainly within a factor of 2 of the exact potential even for wide wells. For the $M>0$ case, Eq. (7) gives zero since the excitonic wave function $|\chi_{N,M,\vec{K}}(0)|^2$ vanishes. This is quite accurate since the exact potential is found to be very small in the $M>0$ case, suppressed by at least a factor of 10 relative to exact potential in the $M=0$ case.