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Electron-hole liquids and band-gap renormalization in short-period semiconductor superlattices

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In short-period superlattices, excited electrons and holes are subject to a one-dimensional perturbation with spatial dimension comparable to the bulk excitonic radius and strength comparable to the bulk excitonic binding energy. We study the stability of the electron-hole liquid and band-gap renormalization as a function of the perturbing potentials (band offsets) using the density-functional approach. We find that there is no universal band-gap renormalization in superlattices, that type-II staggered superlattices are the best candidates for observing the electron-hole liquid, and that a luminescence line in these systems should show a blue shift at intermediate electron-hole densities due to a positive differential band-gap renormalization.

The electron-hole liquid and electron-hole plasma in highly excited semiconductors have been thoroughly investigated.[1] Much of the success in our understanding of this complex many-body problem comes from the fact that the large excitonic radius a_0 and small excitonic Rydberg R allow us to treat electrons and holes as a homogeneous electron-hole plasma (EHP). Much the same is true for semiconductor quantum wells where a picture of a quasi-two-dimensional EHP holds.[2] However, no consensus on the stability of the electron-hole liquid (EHL) and band-gap renormalization has emerged as yet.[2,3] In short-period superlattices,[4] excited electrons and holes are subject to a one-dimensional perturbation of spatial dimension and strength comparable to the bulk excitonic radius and excitonic Rydberg. Moreover, superlattice potentials (band offsets) can be very different for electrons and holes: in type-I superlattices such as GaAs-$Ga_{1-x}Al_xAs$ both electrons and holes are localized in GaAs layers; in mixed superlattices such as CdTe-$Cd_{1-x}Mn_xTe$ electrons are localized but holes are essentially three dimensional (3D); in type-II superlattices such as InAs-GaSb and GaAs-AlAs electrons and holes are spatially separated.[5] The exciting possibility exists of selecting a type-I or a type-II superlattice in a single GaAs-AlAs superlattice sample by the application of a weak electric field.[6] In each case the effect of the superlattice can be varied by varying band offsets, i.e., one can go from a 3D to a 2D system in a controlled way. That this evolution is nontrivial will be shown here.

To study the ground-state properties of the electron-hole system we consider a simple model of interacting electrons and holes with densities n_1 and n_2 and effective masses m_1 and m_2. Each density couples to a 1D periodic potential $V(z)$ (band offset) with periodicity a. In the density-functional approach the ground-state energy is given by[1]

$$E(n_1,n_2)=\sum_{i=1}^{2}\sum_{j,\text{occupied}}\varepsilon_j^i-\frac{1}{2}\sum_{i=1}^{2}\xi_i\int d\mathbf{r}\,n^i(\mathbf{r})\phi(\mathbf{r})+E_{\text{XC}}(n_1,n_2)-\sum_{i=1}^{2}\int d\mathbf{r}\,n_i(\mathbf{r})v_{\text{XC}}^i(n_1,n_2)\,, \tag{1}$$

where E_{XC} is the exchange-correlation energy, $v_{\text{XC}}^i=\delta E_{\text{XC}}/\delta n^i$ is the exchange-correlation potential, $\xi_{i,j}=\pm 1$. ϵ is the background dielectric constant and ε_j^i are the eigenvalues of Kohn-Sham (KS) equations

$$\begin{aligned}
&\left[-\frac{1}{2m_i}\nabla^2+v_{\text{eff}}^i(\mathbf{r};n_1,n_2)\right]\psi_j^i(r)=\varepsilon_j^i\psi_j^i(\mathbf{r})\,,\\
&n_i(\mathbf{r})=\sum_{j,\text{occupied}}|\psi_j^i(\mathbf{r})|^2\,,\\
&v_{\text{eff}}^i=v^i(z)+\xi_i\phi(z)+v_{\text{XC}}^i\,,\\
&\phi(z)=\frac{e^2}{\epsilon}\sum_j\xi_j\int dr'\frac{n_j(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\,.
\end{aligned} \tag{2}$$

Here v_{eff}^i is the effective potential for the ith carrier and ϕ is the electrostatic potential.

The exchange-correlation energy can only be treated approximately and we use the local-density approximation

$$E_{\text{XC}}=\int d\mathbf{r}[n_1(\mathbf{r})e_{\text{XC}}(n_1)+n_2(\mathbf{r})e_{\text{XC}}(n_2)]\,, \tag{3}$$

where e_{XC} is the exchange-correlation energy per electron or hole. We consider two approximations: (1) the Vashishta and Kalia (VK) approximation where e_{XC} is the energy per electron (hole) in a homogeneous electron-hole plasma as parametrized in Ref. 1 and (2) an approximation in which e_{XC} is the exchange correlation per electron (hole) in a single-component plasma as parametrized by Hedin and Lundqvist.[7] The approximation (1) overestimates while approximation (2) underestimates correlation energies, while exchange energies are treated on the same footing. We next proceed to solve self-consistently the set of coupled Kohn-Sham equations and determine ground-state energy as a function of electron-hole density for

different superlattice potentials. Since the external potential is a function of z, we write KS wave functions and eigenvalues in the form

$$\psi_j^i(r,z)=\frac{1}{\sqrt{Aa}}e^{i\mathbf{k}\cdot\mathbf{r}}e^{ik_z z}\sum_{l=-\infty}^{+\infty}\exp\left[-i\left(\frac{2\pi}{a}l\right)z\right]C_l^i(k_z)\,, \tag{4}$$

$$\varepsilon_j^i=k^2/2m_i+\varepsilon^i(k_z)\,.$$

Here k_z is the Bloch wave vector along the z axis and $\mathbf{r}$ is the position in the plane perpendicular to z axis. We expand all relevant quantities (potentials, densities) in the Fourier series, e.g., $n(z)=\sum_l n_l e^{-i(2\pi/a)lz}$. The KS equations are then cast in the form of matrix equations for coefficients $C_l^i(k_z)$ and eigenvalues $\varepsilon^i(k_z)$ which are solved to determine self-consistent densities, Fermi energies, Hartree and exchange-correlation potentials, and KS band structure. This procedure has been carried out for three sets of potentials.

Set (a):

$$V_1(z)=V_2(z)=0,\quad |z|<a/4\,,$$

$$V_1(z)=V_e,\quad V_2(z)=V_h,\quad a/4<|z|<a/2\,.$$

Set (b):

$$V_1(z)=0,\quad |z|<a/4\,,$$

$$V_1(z)=V_e,\quad a/4<|z|<a/2\,,$$

and

$$V_2(z)=0\ \text{ for }\ 0<|z|<a/2\,.$$

Set (c):

$$V_1(z)=0,\quad 0<|z|<a/4\,,$$

$$V_1(z)=V_e,\quad a/4<|z|<a/2\,,$$

and

$$V_2(z)=V_h,\quad 0<|z|<a/4\,,$$

$$V_2(z)=0,\quad a/4<|z|<a/2\,.$$

The masses have been taken to be $m_1=0.1$ and $m_2=0.4$, which are characteristic of electron and heavy-hole masses. The cases a, b, and c describe a type-I, mixed, and type-II superlattice. We define a band gap as the difference of the sum of the lowest KS eigenvalues for a given density of carriers and for the empty bands.[8] In a similar way the chemical potential (the sum of Fermi energies) is defined. The changes of the band gap with increasing carrier density (increasing power of radiation) are responsible for the shift of the luminenscence peak while the difference between the gap and the chemical potential determines the width of the peak. If an electron-hole liquid is formed, the position of the peak is insensitive to the intensity of radiation and to the changes in temperature. The EHL can be formed if the energy per electron-hole pair is equal to the chemical potential and is smaller than the binding energy of an exciton. In our model the exciton corresponds to the lowest bound state of an interacting electron-hole pair in the presence of superlattice potentials. The binding energy is obtained by solving an integral equation for the Fourier transform of the exciton wave function

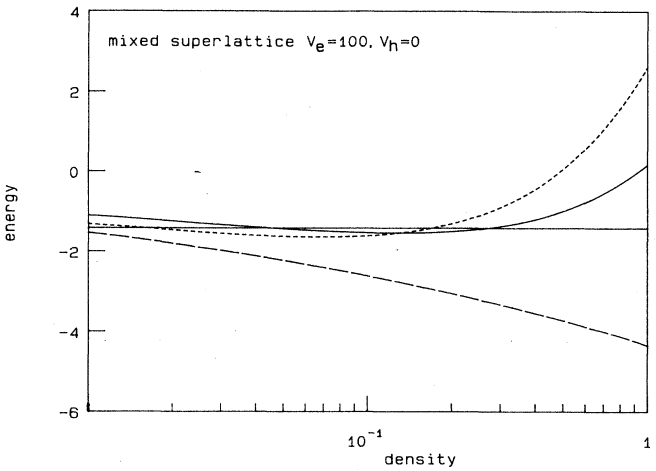

FIG. 1. The energy per electron-hole pair (solid line), the single exciton binding energy (horizontal solid line), the band gap (dashed line), and the chemical potential (dotted line) as a function of electron-hole density for a mixed superlattice. The energy is measured in excitonic Rydbergs and the density in units of a_0^{-3} where a_0 is the excitonic radius. The amplitude of the conduction-band offset is $V_e=100$ ($V_h=0$ for holes). Other parameters are $a=a_0$, $m_1=0.1m_e$, and $m_2=0.4m_e$.

$$[\varepsilon_1(\mathbf{k},k_z)+\varepsilon_2(\mathbf{k},k_z)-E]\phi(\mathbf{k},k_z)+\int\frac{d^2k'}{(2\pi)^2}\int_{-\pi/a}^{\pi/a}dk_z' v(\mathbf{k},k_z;\mathbf{k}',k_z')\phi(\mathbf{k}',k_z')=0\,,$$

$$v(\mathbf{k},k_z,\mathbf{k}',k_z')=\sum_{\substack{l_1l_2\\ l_3l_4}}v_{3D}\left(\mathbf{k}-\mathbf{k}';k_z-k_z'+\frac{2\pi}{a}(l_1-l_2)\right)C_{l_1}^1(k_z)C_{l_2}^1(k_z')C_{l_3}^2(-k_z)C_{l_4}^2(-k_z')\delta_{l_1-l_2,l_4-l_3}\,. \tag{5}$$

v_{3D} is the 3D Fourier transform of the Coulomb interaction. In writing Eq. (5) we have retained only states from the lowest conduction and valence subbands. The extension to higher subbands allows for the systematic approach to the exact solution of Eq. (5). We have solved Eq. (5) by transforming it into a symmetrical singular Fredholm equation which was posed as an eigenvalue problem for a charge on a hole and solved using modified quadrature method.[9]

We now turn to the discussion of results. In Fig. 1 we

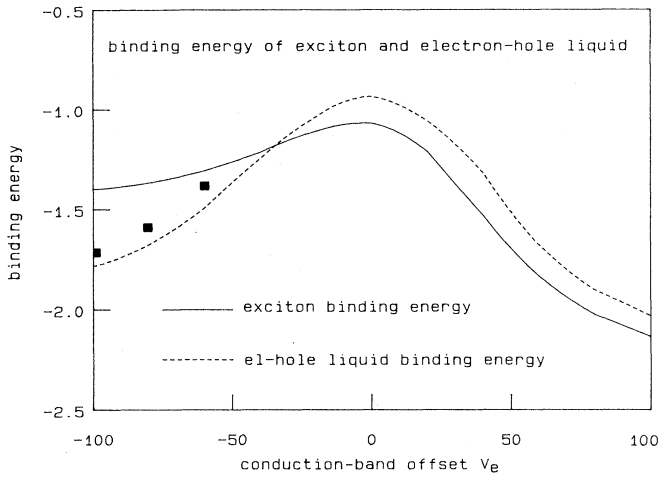

FIG. 2. The minimum energy of the EHL and the exciton binding energy as a function of the conduction-band offset V_e in the VK approximation. The valence-band offset is $V_h = V_e/4$ and other parameters as in Fig. 1. The negative values of the band offset correspond to a type-II and positive ones to a type-I superlattice. Squares indicate results in the second approximation which neglects correlations between electrons and holes.

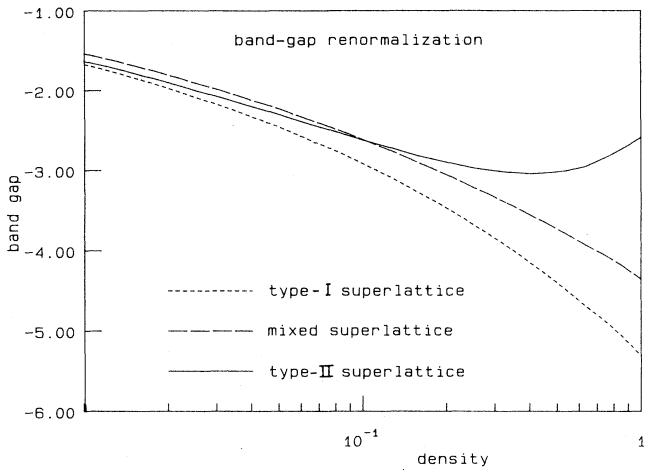

FIG. 3. The band gap as a function of the electron-hole density for the type-I, type-II, and mixed superlattice. Band offsets are $V_e = 100$, $V_n = 25$. Other parameters as in Fig. 1. Note the nonmonotonic dependence of the band gap on density in a type-II superlattice.

show the energy of the EHL, the binding energy of an exciton (solid lines), the chemical potential (dotted line), and the renormalized band gap (dashed line) as a function of electron-hole density for a mixed superlattice. The minimum EHP energy and the excitonic binding energy in type-I (positive V_e) and type-II superlattices (negative V_e) are shown in Fig. 2. The band gap as a function of density for a type-I, type-II, and mixed superlattice is shown in Fig. 3. We find that the excitons have larger binding energy than the EHL in type-I superlattices (see Fig. 2) irrespective of the band offset, but for band offsets larger than a critical value, EHL becomes stable in type-II superlattices. This effect is true even if we neglect entirely correlations between electrons and holes (approximation 2) which decreases EHL binding energies. The uncertainty in correlation energy does not affect the stability of EHL. For strong confinement of electrons ($V_e = 100R$) we find that EHL is unstable in a type-I superlattice but stable in type-II superlattice ($V_e = 100R, V_h = 25R$) and weakly stable in a mixed superlattice ($V_e = 100R, V_h = 0$) as seen in Fig. 1. We note that in a strictly two-dimensional layered electron-hole liquid the exciton gas is the ground state.[10] This demonstrates that the superlattice effects are important. We now turn to the band gap as a function of density as shown in Fig. 3. At low densities the largest reduction in the gap is for a type-I superlattice, followed by a type-II and mixed superlattices. This can be understood as being due to confinement of electrons and holes, strong in type-I and type-II superlattices but much weaker in a mixed superlattice. As the density increases both type-I and mixed superlattices show a gap shrinkage at different rates and quite different from an approximate 2D relation $\sim n_{2D}^{1/3}$.[2] The behavior of the gap in a type-II superlattice is quite different. While initially the gap starts to decrease it reaches a minimum value and then starts to increase. This nonmonotonic behavior is due to both Hartree and exchange-correlation potentials. For a very high density, Hartree fields will compensate confining potentials and the gap will increase again with density of excited carriers.

In summary, we find that the electron-hole liquid should be stable in type-II staggered superlattices. The band-gap normalization with electron-hole density is a sensitive function of conduction- and valence-band offsets. While in both 2D and 3D semiconductors the band gap shrinks with increasing electron-hole density, we find that in type-II superlattices it is possible to increase the gap with increasing electron-hole density. The "positive differential band-gap renormalization" opens up new possibilities for light emitting devices. The nature of electron-hole droplets, the exact form of the exchange-correlation function E_{XC}, and the effect of details of band structure on electron-hole liquids in superlattices remains a challenge.

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