Possibility of an excitonic ground state in quantum wells

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The possibility of the formation of an excitonic insulator in semiconductors with an indirect gap smaller than the exciton binding energy has been considered in the past. The advent of quantum-well structures makes it feasible to consider indirect gaps in real space that can be conveniently tuned through the semiconductor-semimetal transition. We discuss various quantum-well configurations where a spontaneous condensation of excitons into a collective ground state may be possible.

Recent advances in film-growth techniques have made it possible to fabricate multiple layers of very thin films with sharp interfaces. The thickness of individual layers can be made as small as 10–20 Å.[1] These novel configurations raise the possibility of new and unique phenomena having no counterparts in bulk crystals. In this Brief Report we discuss theoretically the possibility of an excitonic ground state that can arise when two spatially separated quantum wells (having *n*- and *p*-type conductivities) with negligible overlap in their electronic wave functions are fabricated sufficiently close to interact via the Coulomb potential. The overlap between the electronic wave functions in the two wells depends both on the height of the energy barrier (W_B) and on the width of the barrier (z_B) separating the two wells, while the strength of the Coulomb interaction between the wells depends only on z_B. Consequently, it is possible to make tunneling effects negligible without any significant reduction in the Coulomb interaction by choosing W_B large and z_B small.

The basic idea underlying the effect we are discussing is as follows. Consider an ordinary semiconductor whose ground state consists of an empty conduction band and a full valence band. It is well known that the lowest excited state is an exciton state comprising an electron-hole pair bound together by their Coulomb attraction.[2] The energy of the exciton state E_X is equal to the band gap energy E_G minus the binding energy E_B:

$$E_X = E_G - E_B \ . \tag{1}$$

The binding energy E_B of Wannier excitons in bulk semiconductors is well described by a hydrogen-atom model:

$$E_B = e^2/8\pi\bar{\epsilon}a_0 \ , \tag{2a}$$

where

$$a_0 = 4\pi\epsilon\hbar^2/\mu e^2 \ , \tag{2b}$$

$\bar{\epsilon}$ is the dielectric constant, and μ is the reduced mass of the electron-hole pair. Many researchers in the past have considered the possibility of a phase transition due to the spontaneous formation of exciton pairs if the binding energy E_B were to exceed the gap energy E_G, since E_X would then be negative.[3,4] This is unlikely in direct-gap materials for which the dielectric constant $\bar{\epsilon}$ increases with decreasing band gap leading to a smaller binding energy; consequently, the band-gap energy (E_G) always exceeds the binding energy (E_B).[5] However, in indirect-gap materials it is possible for E_B to exceed E_G, since the dielectric constant depends on the direct gap, which could be large even though the indirect gap E_G is small. Various materials in which an indirect gap can be tuned through zero with an applied pressure were proposed for the realization of this effect; however, to our knowledge, no experimental evidence has yet been reported.

The advent of quantum-well structures makes it feasible to consider *indirect gaps in real space rather than in k space,* so that an empty band in one quantum well nearby overlaps (in energy) a filled band in another quantum well spatially separated from the first. In fact, in type-II superlattices, using InAs and GaSb, first proposed by Esaki *et al.*,[6] this indirect gap in real space was tuned by size quantization effects to realize a semiconductor-semimetal transition; recent experiments have also revealed the possible existence of novel excitonic states at high magnetic fields.[7] However, the excitonic effects we are proposing are expected to be greatly enhanced if a wide-gap semiconductor like AlSb is interposed between the InAs and GaSb so as to minimize any overlap between the electron and hole wave functions; otherwise virtual transitions between the bands result in an increased dielectric constant $\bar{\epsilon}$ that lowers the exciton binding energy. If the spatial separation between the wells is less than or comparable to the Bohr radius of the exciton (typically ~150 Å), the reduction in the binding energy of the excitons caused by the separation is not very large; this is confirmed by recent experimental results showing that the binding energy of excitons in quantum wells is not affected significantly by large electric fields which pull apart the electron and hole wave functions by ~50–100 Å.[8] Consequently, it is possible for the binding energy E_B to exceed the energy gap E_G between the empty band in one quantum well and the filled band in the other. The energy of an exciton pair composed of spatially separated electrons and holes will then become negative, making it the ground state rather than an excited state. Since excitons behave more like bosons (rather than fermions), it is then possible for the system to generate exciton pairs spontaneously and condense into a collective ground state[4] with novel properties. The possibility of observing a new type of excitonic matter using high-power laser sources to generate large numbers of excitons has led to an exciting field of research in solid state physics.[9] By contrast, the effect we are proposing here would lead to the *spontaneous* production of large numbers of excitons without external sources, since the excitonic state would be the ground state rather than an excited state of the system.

Both "type-III" superlattices (GaSb-AlSb-InAs) and doping (*n*-*i*-*p*-*i*) superlattices (with heavy doping to reduce the spacing between n and p layers) could be suitable for observing these effects. However, a major problem could be the presence of impurities and defects that trap the carriers. To alleviate this problem a new structure is proposed using modulation-doped GaAs-AlGaAs p-n junctions. The improved technology of the AlGaAs system could lead to better interfaces with reduced defects; also, the modulation doping separates the electrons and holes physically from the dopants which are introduced into the wide-gap material. An attractive feature of the structure proposed here is that electrons and holes are spatially separated by an insulator. It is thus possible to change their relative energies with an applied potential, raising the possibility of a novel voltage-controlled phase transition; it also provides a very convenient control parameter in an experiment.

Another intriguing possibility raised by the spatial separation of the electrons and holes has been considered by several authors[10,11] in the past. It has been proposed that the antiparallel currents which flow when we apply opposite electric fields to the two quantum wells may have many of the characteristics of a superflow. The advent of quantum-well structures makes it possible to fabricate material configurations that may lead to a practical realization of this effect. Techniques for making individual contacts to different layers in a superlattice have been discussed in the context of n-i-p-i superlattices.[12]

Spatially Separated Excitons. Consider a configuration in which the conduction band and the valence band belong to spatially separated quantum wells as shown in Fig. 1, so that the conduction band (band A) at $z=0$ is separated by E_G from the valence band (band B) at $z=d$. The Fermi level runs between bands A and B; the other bands (such as bands A' and B') are far removed from the Fermi level and can be neglected. In this case we can expect electrons in band A and holes in band B to form excitons for which the binding energy will be reduced relative to the usual situation where electron and hole wave functions overlap spatially. This reduction occurs because the Coulomb interaction between an electron and hole separated by a distance ρ in the plane of the layer is reduced by the factor $\rho/(\rho^2+d^2)^{1/2}$, where d is the separation between the quantum wells. However, the reduction in the binding energy is not very large as long as d is less than or comparable to the Bohr radius for the exciton (a_0). At the same time, the energy barrier between the quantum wells should be large enough to ensure negligible overlap between the electronic wave functions in bands A and B. Otherwise the electron-hole interaction will be reduced because of an increase in the dielectric constant $\overline{\epsilon}$ due to virtual transitions between bands A and B. With a barrier width (z_B) of 50 Å, a barrier height (W_B) of 0.6 eV is sufficient to ensure that the overlap of wave functions between bands A and B is negligible, since in this case the wave function decays by a factor of ~ 100 from one well to another. Under these conditions, with an appropriate choice of materials it should be possible to make E_G less than the exciton binding energy, so that the exciton energy E_X is negative leading to the spontaneous formation of excitons.

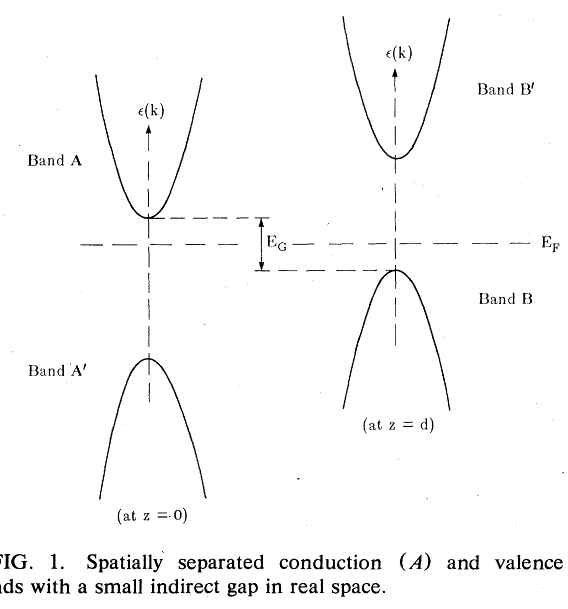

FIG. 1. Spatially separated conduction (A) and valence (B) bands with a small indirect gap in real space.

Using parameters typical of a semiconductor like GaAs ($\overline{\epsilon}=12.5\epsilon_0$, $\mu=0.04\ m_0$, where ϵ_0 is the permittivity of vacuum and m_0 is the free-electron mass), we can estimate the exciton binding energy and the Bohr radius for a bulk semiconductor from Eq. (2),

$$a_0=168\ \text{Å},\quad E_B=3.4\ \text{meV}\ .$$

To estimate the binding energy of a spatially separated exciton, we note that with zero spatial separation ($d=0$) the binding energy of an exciton in a quantum well is enhanced over the bulk value [Eq. (1)] by up to a factor of 4,[13–17] depending on the thickness of the well. The binding energy in a 50-Å-thick GaAs quantum well with 0.6 eV barriers is estimated to be ~ 10 meV in Ref. 16. As discussed earlier, the binding energy of a spatially separated exciton will be reduced somewhat compared to this value. Our own calculations, as well as the results in Ref. 13, show that the binding energy is reduced by a factor of ~ 4 if the spatial separation d is equal to the (bulk) Bohr radius a_0, and by a factor of ~ 8 if $d=2a_0$. Using parameters for a typical semiconductor like GaAs, we can expect a binding energy ~ 1.2 meV even if the electron and hole are separated by 300 Å, neglecting any reduction due to screening.[18] It should be possible to observe a transition to an excitonic state in a configuration with a small indirect gap in real space, even if the spatial separation is ~ 300 Å; of course, the effect is expected to be stronger for smaller separations.

A possible configuration for realizing the proposed effect is based on type III superlattices first proposed by Esaki *et al.*[6] GaSb, AlSb, and InAs are closely lattice matched and are suitable for heteroepitaxy. AlSb is a wide-gap compound that serves as an energy barrier for both electrons and holes. GaSb and InAs have an unusual band lineup, wherein the valence-band edge in GaSb lies above the conduction-band edge in InAs. Consequently, E_G is negative (~ -0.15 eV). Moreover, E_G can be tuned through zero to positive values by using size quantization effects; that is, if the GaSb and InAs layers are of quantum-well dimensions, then the effective E_V in GaSb is lowered while the effective E_C in InAs is raised. In fact, this effect has been used to realize a semiconductor-semimetal transition in GaSb-InAs heterostructures. Recent experiments also suggest the possibility that a novel excitonic state is induced at high magnetic fields.[7] However, the excitonic effects which we are proposing are expected to be significantly enhanced by an intervening AlSb barrier; otherwise, the overlap of electron and hole wave functions affects the dielectric constant, tending to reduce the exciton binding energy. Another possibility is the use of doping (n-i-p-i) superlattices which have attracted considerable attention[19] recently as a configuration leading to a tunable indirect gap in real space. To observe the effect we are proposing, it is important to reduce the spatial separation between the electrons and holes to less than ~300 Å, so that the binding energy is not reduced significantly; this requires heavy doping in the n and p regions. Our calculations, as well as the results in Ref. 20, indicate that a doping level of $5\times10^{18}/\mathrm{cm}^3$ and an n-p separation of 300 Å leads to a "semimetal" configuration with a negative indirect gap in real space.

Modulation-doped p-n junctions. One of the major difficulties with both of the above structures (especially the latter) is that impurities and defects tend to capture carriers, preventing the formation of an excitonic state. The use of modulation-doped GaAs-AlGaAs p-n junctions should help overcome this problem. In these structures, the dopants can be physically separated from the carriers; also, the materials and the interfaces are relatively defect free and of high quality because of the advanced technology. Figure 2(a) shows a p-n junction with a quantum well on either side of the junction. At equilibrium there is a built-in potential at the junction comparable to the band gap of GaAs, so that the conduction band of one well overlaps the valence band of the other [Fig. 2(b)]. Note that the doping is confined to the AlGaAs layers, so that the GaAs quantum wells are free from impurities. Another attractive feature is that the electron and hole quantum wells are separated by low-conductivity material, so that the externally applied potential can be used to tune the overlap between the bands; this provides a very convenient control parameter for an experiment, with the possibilty of a voltage-controlled phase transition. The separation between the wells can be made smaller if a reverse bias is used to provide the additional band bending; however, if the separation is too small, problems can arise due to electrical breakdown.

In this Brief Report we have examined theoretically the possibility of a novel collective ground state due to the spontaneous condensation of excitons. Such a state can arise when the conduction band in one quantum well overlaps or nearly overlaps the valence band in another quantum well. The spatial separation between the two quantum wells should be less than or comparable to the Bohr radius of the exciton, so that the Coulomb interaction between them is not reduced significantly. At the same time, the wells should be separated by a wide-gap material to minimize the overlap between their wave functions; any overlap of wave functions tends to increase the dielectric constant and reduce the exciton binding energy. Possible configurations for the realization of this effect are suggested.

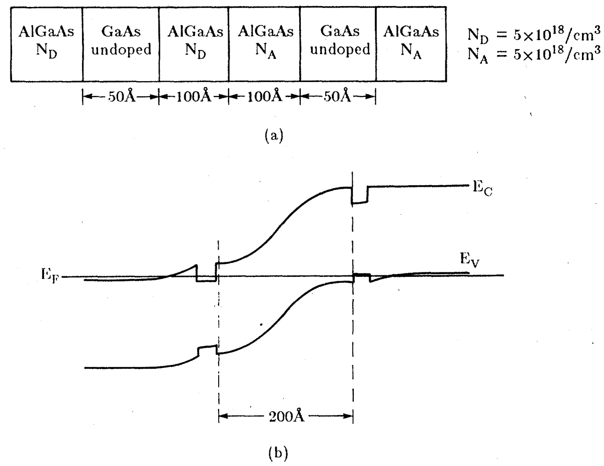

FIG. 2. A two-quantum-well p-n junction leading to a small indirect gap in real space that can be tuned with an applied potential: (a), configuration; (b), band diagram at equilibrium.

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