Existence of Exciton Crystals in Quantum Wires

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An exciton crystal (EC) is predicted in a quasi-one-dimensional (1D) semiconductor quantum wire for the densities ($1 < r_s \leq 5.5$) due to the repulsive interaction between singlet excitons (x's). Low-lying excitations, stability, and melting of the EC are determined within the Heitler-London approximation. The expected short condensation times (ps) will allow an EC formation in, e.g., GaAs quantum-well wires with relatively short x lifetimes. For $T \leq 10$ K the EC is a pure quantum crystal which favors collective, coherent emission in the form of superradiance.

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Collective electronic phases are of fundamental interest in solid-state physics. Recently, the concept of a Wigner electron crystal (WEC) [1–4] has been verified in a number of experiments in 2D electron systems [5,6]. However, a "true" 2D-WEC at zero magnetic field requires a very dilute electron system and thus is difficult to realize. The WEC can be the ground state of the system of free electrons moving in an ion lattice which is usually approximated by a jellium model. Then the ground energy is determined by the dimensionless interparticle spacing r_s, which is the ratio of the particle distance and the *electron Bohr radius* calculated with an effective electron mass and a background dielectric constant. Only in the low-density limit ($r_s \geq 10$) a simple unscreened interaction between nearest-neighbor electrons can be considered. If this assumption holds, WEC condensation can occur.

In a neutral electron-hole (e-h) system the ground state is rather different. Here the e-h pairing to x states minimizes the ground-state energy and introduces the *exciton Bohr radius* a_x as a natural length scale. Higher order e-h correlations are connected with x-x interactions. Because the x is a neutral complex, the x screening is suppressed and becomes only important in the high-density limit ($r_s \leq 2$) [7,8]. In this limit, the x overlap is large, whereas in the low-density limit the x system is an ideal Bose gas becoming weakly nonideal at intermediate densities ($2 \leq r_s \leq 10$) [9]. Thus, the ground state at these densities is a Bose-Einstein condensate of the x's. This state has been considered as an excitonic insulator phase in connection with the metal-insulator transition in semimetals [10], as well as in ordinary semiconductors [9]. The x representation for the description of the ground state of the degenerate e-h system is valid below the Mott transition to an e-h plasma which occurs at relatively high densities ($r_s \simeq 1.7$ for the 3D case [7], e.g.).

In this Letter we propose a crystal-like excitonic phase as the ground state of a 1D x system at intermediate x densities. For this density range, the generalized Hartree-Fock method [9,10] is often used. This treatment implies a *homogeneous* x distribution in real space. Instead of this, we consider a spatially ordered structure of x's. The one dimensionality of the x system plays an important role in our treatment.

We consider a quasi-1D system of Wannier x's and assume that all x's have the same singlet spin structure with, e.g., $e\uparrow$ and $h\downarrow$. This situation can be realized, e.g., in GaAs/$Ga_{1-x}Al_xAs$ quantum-well wires (QWW) by exciting the x's with a circularly polarized light pulse. Because of the Pauli principle, the interaction potential between these singlet x's is repulsive. This repulsion drives the x system into a state with equal separation. A simple analog is a set of identical point masses coupled by identical springs. The whole chain is under an external pressure. This situation has been realized in a dilute system of the laser-cooled Mg^+ ions stored in a closed 1D ring trap [11]. For a 1D x system the question arises whether the large zero-point motion connected with the light x mass will destroy such an x condensate with 1D diagonal order.

A 1D hydrogen atom with a singular 1D Coulomb potential, as well as its regularization, has been treated in Ref. [12]. This model has been adopted for 1D e-h systems in QWW [13] with free motion in the z direction and an infinite circular confinement potential with a radius R in the x-y plane. The effective Coulomb interaction between the carriers is obtained by averaging the 3D Coulomb potential with the transverse envelope wave function in the confinement potential of the wire. The resulting quasi-1D e-h Coulomb potential $U(z)$ can be approximated by the following regularized 1D Coulomb potential [13]:

$$U(z) = a_0/(|z| + \alpha R) \ , \tag{1}$$

with $\alpha \simeq 0.3$ for GaAs/$Ga_{1-x}Al_xAs$ QWW. Here, a_0 is the bulk x Bohr radius, while the energy is scaled to the bulk x Rydberg E_0. The corresponding x wave function is given in terms of the Whittaker function $W_{\kappa,1/2}(x)$: $\Psi_x(z) \propto W_{|\epsilon|^{-1/2},1/2}[|z|/a_{1D,x} + (1/2)e^{-|\epsilon|^{1/2}}]$, where $a_{1D,x} = (a_0/2)|\epsilon|^{-1/2}$ and $|\epsilon|E_0$ are the 1D x radius and binding energy of the ground state, respectively.

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There is a universal relation between R and ϵ, which for $R \to 0$ has the explicit asymptotic form [12,13]

$$\frac{2\alpha R\sqrt{|\epsilon|}}{a_0} = e^{-\sqrt{|\epsilon|}/2}. \tag{2}$$

Equation (2) allows us to use ϵ rather than R as the characteristic wire parameter.

The low-lying collective excitations of the exciton crystal (EC) are LA-phonon-like with the dispersion

$$\omega = \omega(k) = 2\sqrt{\frac{W''(a)\,E_0}{M}}\left|\sin\left(\frac{ka}{2}\right)\right|. \tag{3}$$

Here, W and W'' are the potential energy of the x-x interaction and its second spatial derivative, respectively, M is the total x mass, and $a = a_{x\text{-}x}$ is the lattice parameter of the EC, i.e., the distance between the centers of neighboring x's. According to the Mott criterion [2–4], a lattice exists at $T = 0$ K, if the potential energy of the lattice exceeds the corresponding zero-point energy ε_0. The zero-point motion includes the kinetic energy as well as some potential energy. For the EC this criterion takes the form

$$\varepsilon_0(r_s) = \frac{4\sqrt{2}}{\pi}\sqrt{\frac{\mu}{M}}\sqrt{|\epsilon|W''(r_s)} \le W(r_s), \tag{4}$$

where μ and M are the reduced and translational x mass, respectively. The dimensionless interparticle distance $r_s = a/a_{1\mathrm{D},x}$ is the normalized lattice parameter. The Mott criterion guarantees the stability of the EC phase against the x gas phase. Because the x wave function decays exponentially with the distance from the x lattice site, the inequality (4) defines the upper phase boundary r_{sc} of the EC. Apart from the energy $|\epsilon|$, the Mott criterion (4) contains the mass ratio μ/M. This ratio is the smallness parameter which ensures the high stability of usual atomic lattices. For further analysis, the heavy hole x of GaAs/$Ga_{1-x}Al_xAs$ QWW is considered, where $\mu/M = 0.11$.

In order to find $W(r_s)$, the Heitler-London method (see, e.g., [14]) is used. This method is conventionally applied for the chemical forces in the approximation of nearest-neighbor interactions, which is valid for the proposed EC in a wide range of intermediate x densities. The Heitler-London x-x interaction potential as a function of r_s is

$$W(r_s) = 2\sqrt{|\epsilon|}\frac{Q - A}{1 - S^2}, \tag{5}$$

where $S = S(r_s)$ is the overlap integral

$$S = \int dz_1\, \Psi_x(z_1)\, \Psi_x(a - z_1). \tag{6}$$

The Coulomb integral $Q = Q(r_s)$ is

$$Q = U(a) - 2\int dz_1\, \Psi_x^2(z_1)U(a - z_1) + \int dz_1\, dz_2\, \Psi_x^2(z_1)\, \Psi_x^2(z_2)\, U(a - z_1 + z_2), \tag{7}$$

while the exchange integral $A = A(r_s)$ is

$$A = S^2U(a) - 2S\int dz_1\, \Psi_x(z_1)\Psi_x(a - z_1)U(a - z_1) + \int dz_1\, dz_2\, \Psi_x(z_1)\Psi_x(a - z_1)U(a - z_1 + z_2) \times \Psi_x(z_2)\Psi_x(a + z_2). \tag{8}$$

Note that the distance between the heavy particles (here the holes) rather than the distance between the centers of mass has to be used as the adiabatically varying parameter. This approach is valid for $r_s \gg m_e/M = 0.12$, where m_e is the electron mass.

All the integrals in Eqs. (6), (7), and (8) can be evaluated analytically for $|\epsilon| \gg 1$. In this strong-confinement limit the normalized x wave function is [12]

$$\Psi_x(z) = \frac{1}{\sqrt{2a_{1\mathrm{D},x}}}e^{-|z|/2a_{1\mathrm{D},x}}. \tag{9}$$

With Eq. (9) the x-x interaction potential $W(r_s)$ becomes

$$W(r_s) = \frac{\sqrt{|\epsilon|}}{2}\frac{e^{-r_s}B + C}{1 - e^{-r_s}\left(1 + \frac{r_s}{2}\right)^2}, \tag{10}$$

with

$$B = -\frac{4}{r_s} - 4 + r_s\left(4\sqrt{|\epsilon|} - 2\gamma - 1\right) + 4r_s e^{r_s}E_1(r_s) + 2r_s \ln r_s \tag{11}$$

and

$$C = \frac{4}{r_s} - \left(1 + \frac{3}{r_s}\right)r_s e^{r_s}E_1(r_s) + \left(1 - \frac{3}{r_s}\right)r_s e^{-r_s}E_i(r_s). \tag{12}$$

Here, $\gamma = 0.577$ is the Euler constant and $E_1(r_s)$ and $E_i(r_s)$ are the exponential integral functions. Equations (4) and (2) yield the limiting value of the upper boundary $r_{sc}(-\epsilon \to \infty, R \to 0) = 3.4$.

We also evaluated the integrals (6), (7), and (8) numerically with the exact x wave functions. In Fig. 1 the interaction potential and the zero-point energy of the EC are compared for the confinement parameter $|\epsilon| = 4.0$, corresponding to $E_{1\mathrm{D},x} = 16.8$ meV and $R \simeq 0.1a_0$. Such binding energies can be realized experimentally [15]. The resulting upper critical lattice parameter is $r_{s,\mathrm{uc}} \simeq 4.5$. This critical parameter reaches at small but finite wire radii $R \simeq 0.2a_0$ (corresponding to $|\epsilon| = 2.38$ or $E_{1\mathrm{D},x} = 10.0$ meV) a maximum of $r_{s,\mathrm{uc}} = 5.4$, because the x wave function decreases more slowly for smaller $|\epsilon|$ values. For x binding energies $E_{1\mathrm{D},x} \le 10.0$ meV an instability of the EC lattice occurs. This instability arises when W'' becomes negative, so that $\sqrt{W''}$ in (3) and (4) becomes imaginary. Imaginary values of $\omega(k)$ are a manifestation of a dimerization transition to a lattice with two x's per elementary cell. Note that our calculations also

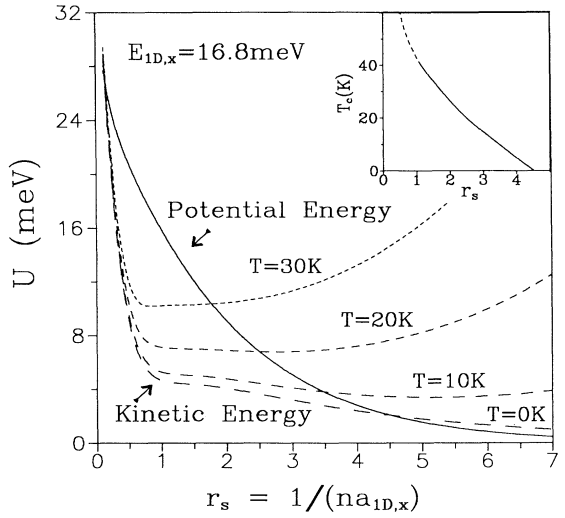

FIG. 1. x-x interaction potential (full line), kinetic energy at various temperatures (dashed lines), and zero-point energy ($T = 0$ K) versus lattice parameter r_s for the confinement parameter $|\epsilon| = 4.0$ ($E_{1D,x} = 16.8$ meV, $R = 0.1a_0$). Inset: crystallization temperature T_c versus r_s.

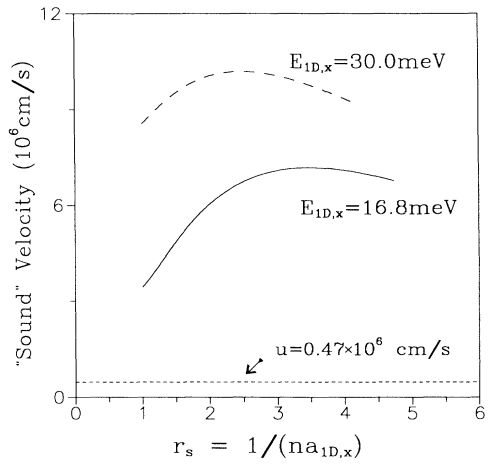

FIG. 2. "Sound" velocity v_s of the EC versus lattice parameter r_s for two wire parameters given in terms of $E_{1D,x}$. The short-dashed line shows the LA-phonon velocity u in bulk GaAs.

yield a lower critical lattice parameter $r_{s,\mathrm{lc}} \simeq 0.2$; however, at these high x densities our approach is no longer valid. In any case the lattice parameter corresponding to the Mott transition $r_{s,M} \simeq 1$ will be the lowest possible r_s value of the EC phase.

The thermal stability of the EC against melting requires that $\varepsilon_0 + \varepsilon_{\mathrm{th}} \leq W(r_s)$ where the thermal energy due to activation of the low-lying collective excitations Eq. (3) is given by

$$\varepsilon_{\mathrm{th}}(r_s) = \frac{\pi}{12\sqrt{2}}\sqrt{\frac{M}{\mu}}\frac{1}{\sqrt{|\epsilon|W''(r_s)}}\left(\frac{k_BT}{E_0}\right)^2. \quad (13)$$

With increasing temperature, the stability range of the EC in terms of r_s decreases as shown in the inset of Fig. 1. The melting temperature $T_c \leq 50$ K for $|\epsilon| = 4.0$, and increases even further for stronger confinement. In 3D or 2D a similar behavior cannot be realized, because there is no additional energy scale apart from the bulk Rydberg E_0. Moreover, the 1D thermal energy $\varepsilon_{\mathrm{th}} \propto T^2$ increases more slowly with T than in 3D, where $\varepsilon_{\mathrm{th}} \propto T^4$. The large value of the EC "sound" velocity v_s further improves the stability of the $1D$ lattice, because $\varepsilon_{\mathrm{th}} \propto 1/v_s$. This velocity $v_s = \frac{\partial\omega}{\partial k}_{k\to 0}$ is given by

$$v_s(r_s) = \sqrt{\frac{E_0}{M}}r_s\sqrt{W''(r_s)}. \quad (14)$$

The dependence $v_s = v_s(r_s)$ is shown in Fig. 2.

With a homogeneous resonant optical excitation one generates rather uniformly distributed Wannier x's with essentially no overlap due to Pauli blocking and with a typically interparticle distance of a few $a_{1D,x}$. If the free x motion is not suppressed by large interface fluctuations, the x's will start to oscillate back and forth between the nearest two neighboring x's, because they cannot pass each other in the QWW. After a few oscillation periods the x's will be localized around their quasiequilibrium sites of the EC, if the condensation conditions are fulfilled. One can estimate the crystallization time as $\tau \simeq 1/\omega(k) \simeq 1/v_sk \simeq L/2\pi v_s$, where L is the crystal coherence length. For $v_s = 5 \times 10^6$ cm/s, $L = 1$ μm, one gets $\tau \simeq 3$ ps. This time is smaller than the corresponding x radiative lifetime in $GaAs/Ga_{1-x}Al_xAs$ QWW's of $\simeq 300$ ps [16]. The described fast condensation kinetics is specific for quasi-1D systems, and has no analog in higher dimensions.

The EC condensation should be observable in luminescence along the wire axis. The radiative x decay is determined by the x distribution. In an EC $n_k = 2\pi(1/2 + 1/\{\exp[\hbar\omega(k)/k_BT] - 1\})$, where the first term is due to the zero-point motion, and in a gas $n_k = A\exp(-\hbar^2k^2)/2Mk_BT$, A is a normalization constant. The probability to find an x with the momentum k_{ph} of the resonant photon is in an EC $\propto n_{k_{\mathrm{ph}}} \simeq 2\pi[1/2 + k_BT/\hbar v_sk_{\mathrm{ph}}]$. At $T \leq 10$ K this probability is about 2 orders smaller than in the corresponding gas phase. Thus, the onset of EC condensation should result in a drop of the incoherent luminescence. On the other hand, at low temperatures where only the relatively large zero-point motion remains, the EC is a pure quantum crystal which is well suited for the emission of a coherent, short, superradiant light pulse. The most direct proof of the existence of an EC would be (near UV) light diffraction by the x lattice.

The specific feature of the EC compared to a WEC is the negative binding energy. Thus, the EC in a QWW

of finite length $z = L$ needs confinement potentials at $z = 0$ and $z = L$. In a QWW the surface potentials are several eV and suffice to stabilize up to 10^3 x's in the EC. Another possibility would be the use of a QWW ring. In any case the EC as a 1D crystal can only exist with finite L in order to avoid the destruction by long-wavelength phonons. The WEC concept implies that an electron in the lattice retains its identity because of the negligible exchange effects [3]. Contrary to that situation, the e-e and h-h exchange effects play an essential role for the EC. The hybridization of the neighboring x states gives rise to the appearance of the corresponding e and h bands. For high x densities, where the increase of the x energy $\Delta E = W + \varepsilon_0 + \varepsilon_{\text{th}}$ is about $E_{1D,x} = -\epsilon E_0$, the Heitler-London approximation becomes invalid and one has to use the linear combination of atomic orbitals method. For the proposed excitation with circularly polarized light, x's have identical spin structures. Then the e and the h bands are filled completely with particles of the given spin orientation. Therefore, the EC is an excitonic insulator. Within the e and h bands, a separate x cannot be identified, but e-h correlations in the form of the Coulomb-enhancement effect give rise to the existence of x fragments. These x fragments have to reveal a collective crystal-like correlation, i.e., the EC. This situation resembles that of a biexciton which can be considered a bound state of two x's; i.e., there is a strong e-h pairing to the x state inside the biexciton.

For an EC one can actually realize Mott's well-known gedanken experiment [4] to vary the lattice parameter a continuously, simply by varying the 1D x concentration. The Mott transition to the metallic state of the e-h plasma with decreasing a is still an open question for 1D systems. Most likely, this transition is continuous because 1D x correlations exist even for arbitrarily small e-h attractions [17]. This likely suppression of a discontinuous Mott transition is a further advantage of the 1D e-h system with respect to the EC formation.

So far, we have proposed all x's have the same interior spin structure, as it is valid during one spin relaxation time after the excitation with a circularly polarized light pulse. In complete equilibrium, the probabilities for e or h spin up or spin down are equal (as it would be for excitation with linearly polarized light). Singlet x's with opposite spins of the e (h) attract each other and form biexcitons, which would result in a doubling of the lattice parameter. In this case an elementary cell of the exciton molecule crystal (EMC) consists of two e's and two h's, both of them having the opposite spins. The e-e and h-h exchange interactions allow the formation of such an EMC from arbitrary initial e and h spin distributions along the z axis. A still further increase of the elementary cell is impossible because biexcitons always repel each other.

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