Formation of Electron Strings in Narrow Band Polar Semiconductors

F. V. Kusmartsev
Department of Physics, Loughborough University, LE11 3TU, United Kingdom and Landau Institute, Moscow, Russia
(Received 18 September 1998)

We show that linear electron strings may arise in polar semiconductors. A single string consists of M spinless fermions trapped by an extended polarization well of a cigar shape. Inside the string the particles are free although they interact with each other via Coulomb forces. The strings arise as a result of an electronic phase separation associated with an instability of small adiabatic polarons. We have found the length of the string which depends on dielectric constants of semiconductors. The appearance of these electron strings may have an impact on the effect of stripe formation observed in a variety of high-T_c experiments.

PACS numbers: 71.38.+i, 61.82.Fk, 63.20.Kr, 71.10.−w

The creation of polarons in solids has proved to be an important phenomenon. The idea originated with Landau in the 1930s to explain a spectrum of F centers in alkali halides. The first consistent theory was built up by Pekar in the 1940s and is elegantly presented in his book [1]. There a charged particle trapped by the polarization of a polar solid was considered. An adiabatic approximation for the motion of the atomic lattice was assumed; i.e., the kinetic energy of the atoms was neglected. This approximation is based on the fact that the mass of electrons is much smaller than the atomic mass and therefore the electrons move much faster than the atoms. Such a charged particle together with the surrounding polarization cloud was called a polaron. The effective radius of this polarization cloud is the radius of the polaron. Pekar considered polarons with a radius which is much larger than the distance between atoms in the lattice.

The adiabatic ground state of a single electron interacting with dispersionless nonpolar optical phonons on one dimensional molecular chain has been studied by Rashba and Holstein [2,3]. While the Holstein model has been studied intensively [4,5], until now the traditional small polarons described in the framework of Pekar-Frölich (PF) Hamiltonian [1] have not been considered. The creation of electronic states with small radii involves a large amount of a polarization or a deformational energy and therefore, such states give rise to an electronic mechanism for the creation of defects in solids [6]. In this case the kinetic energy of the atomic lattice is much smaller than the polarization energy, and so it may be neglected. Therefore these states with small radii may be studied in the adiabatic limit.

We study here M spinless fermions ($M \gg 1$) interacting with "polar" (longitudinal optical) phonons in the framework of the PF polaron model [1]. The Hamiltonian of interest has both long-range electron-phonon (EP) interactions ($\sim 1/q$ in momentum space) and long-range electron-electron (EE) Coulomb interactions, whereas the Holstein-Hubbard (HH) model much studied recently (see, in particular, the recent papers [4]) has both short-range EP and short-range EE interactions. We have found that in polar solids with a narrow band there may arise an electronic phase separation introduced originally in the physics of magnetic semiconductors by Nagaev [7] but here purely due to the EP interaction. Note that due to the long-range character of EE interaction the results obtained in this model differ strongly from those associated with the HH and deformation potential models [8], where the EP interaction is very short ranged (typically on-site or nearest neighbors).

In essence we show that the overall attraction caused by the strong polaronic effects leads effectively to a "collapse" of fermions into the smallest possible region, resulting in a phase separation into a "dense" blob of trapped fermions and an undoped lattice. This "blob" cannot be on a single site because the holes are fermions; the blob assumes a stringlike shape because this minimizes the Coulomb energy for a connected blob. Full minimization of the Coulomb energy *alone* would, at least in the low density limit, lead to a widely separated Wigner crystal. But it was found [9] that at strong EP coupling a Wigner crystal of polarons is unstable. Such an instability may suggest *novel types of electron structures such as electron strings*, found in the present paper. A single string consists of M electrons self-trapped by a single potential well self-created by these electrons. Inside the string the electrons move as free particles. All other flat disk-shaped multielectron configurations have higher energies than the string configuration. In agreement with all existing results [2–5] we find that for strong coupling with polar phonons the ground state for a single particle in any spatial dimension is of the same type as the single site Rashba-Holstein polaron [2,3]. However, we found that if the string consists of M particles and the number M is larger than some critical value M_c ($M > M_c \sim 5132$ for the model parameters we have chosen), then the string configuration has a lower energy than M separated polarons suggesting the formation of electron strings is possible.

To describe the fermion strings in a Hartree-Fock (HF) approximation we have to first find an appropriate basis of single particle wave functions. Therefore, we start with the

 0031-9007/00/84(3)/530(4)$15.00

set of discrete Schrödinger equations describing a single fermion in a tight-binding model on a hypercubic lattice:

$$-t\hat{\Delta}\psi_{np} + e\varphi_n\psi_{np} = E\psi_{np}\,, \tag{1}$$

where t is a hopping integral, e is the fermion charge, ψ_{np} is the wave function of the pth self-trapped fermion on the nth site, φ_n is the total polarization potential created by M fermions trapped by this potential, and $\hat{\Delta}$ is a lattice version of the Laplacian operator which for the hypercubic lattice is defined as $\hat{\Delta}\psi_{n\,p} = -\sum_i(\psi_{n\,p} - \psi_{n+i\,p})$, where the sum is carried out over all the nearest-neighbor sites around the nth site. The overall potential φ_n is actually a superposition of individual potentials created by the individual fermions and is self-consistently determined with the aid of the discrete Poisson equation:

$$\frac{\epsilon^*}{a^2}\,\hat{\Delta}\varphi_n = +4\pi e\sum_{p=1}^{M}|\,\psi_{np}|^2, \tag{2}$$

where the index p indicates the summation carried over all trapped fermions, the parameter a is the distance between neighboring atoms in the lattice, and ϵ^* is the effective dielectric constant, which was originally obtained by Pekar [1] in the case of large polaron as $\epsilon^{*^{-1}} = \epsilon_\infty^{-1} - \epsilon_0^{-1}$.

Pekar [1] invented the functional J (adiabatic potential) describing the total energy of the fermions and the lattice degrees of freedom which is defined as

$$J = T + \sum_{np} e\varphi_n\,|\,\psi_{np}|^2 - \frac{\epsilon^*}{8\pi a^2}\sum_n \varphi_n\hat{\Delta}\varphi_n\,, \tag{3}$$

where $T = -t\sum_{np}\psi_{np}^*\hat{\Delta}\psi_{np}$ is an electron kinetic energy. Equations (1) and (2) may be obtained by a minimization of J with respect to the fermion wave function ψ_{np} and the total polarization potential φ_n, respectively, provided that the wave function satisfies the normalization condition $\sum_n|\,\psi_{np}|^2 = 1$. In the sums the index n indicates a summation over all lattice sites. The polarization potential can be easily obtained from Eq. (2) and substituted into Eq. (3) to get

$$J = T + \frac{2\pi e^2a^2}{\epsilon^*}\sum_{npmq}|\,\psi_{np}|^2\hat{\Delta}^{-1}|\,\psi_{mq}|^2. \tag{4}$$

The extrema, critical points of J are determined with the aid of the following systems of equations having nonlocal nonlinearity:

$$-t\hat{\Delta}\psi_{np} - ca^3\psi_{np}\sum_{mq}\hat{\Delta}^{-1}|\,\psi_{mq}|^2 = E\psi_{np}\,, \tag{5}$$

where the dimensionless coupling constant, c, is defined as $c = \frac{4\pi e^2}{\epsilon^* a}$. In the limit $(Mc/t)\to\infty$ Eq. (5) allows a complete set of the following exact string solutions, in which each of the M fermions is occupying N neighboring sites with equal probability; i.e., $|\,\psi_{np}|^2 = 1/N$ if the nth site is in the string and $\psi_{np}\equiv 0$ otherwise. In other words, this complete set of the single particle wave functions of the trapped fermions takes the form

$$\psi_{np}(k_x) = \begin{cases} \frac{1}{\sqrt{N}}\exp(ik_xn_x) & \text{if} \quad 1\le n_x\le N\,, \\ \equiv 0, & \text{otherwise}\,, \end{cases} \tag{6}$$

where k_x is the momentum of the pth fermion. We assume that the string is oriented in the **x** direction and located on the sites, from $n_x = 1$ to $n_x = N$. Inside the string each trapped fermion has free motion with the quasimomentum k oriented in the direction of the string.

The described wave functions associated with fermions self-trapped into the string correspond to the following eigenvalues, E, of Eq. (5):

$$E_{NM}(k) = 2dt - 2t\cos(ka) + \frac{2t\cos(ka)}{N} - cMI_N\,, \tag{7}$$

where d is the dimension of the hypercubic lattice containing the string and the constant I_N is a dimensionless integral. When the string is embedded into a 3D atomic lattice the integral, I_N, takes the form

$$I_N = \int_{-\pi}^{\pi}\int_{-\pi}^{\pi}\int_{-\pi}^{\pi}\frac{dx\,dy\,dz\,G(x,y,z)}{(2\pi)^3}\left(\frac{\sin(Nx)}{N\sin\frac{x}{2}}\right)^2, \tag{8}$$

where $G^{-1}(x,y,z) = 3 - \cos x - \cos y - \cos z$ is a 3D lattice Green's function. For $N = 1$ the integral I_N can be expressed through some elliptic integrals and is equal to $I_1 = 0.526\,416$. With the increase of N the value of I_N decreases as $I_N \approx \frac{A}{N^\alpha}$, where A and α are some near constant parameters. The dependence of A and α on N is extremely weak. So for $10\le N\le 2.0\times 10^3$ the value I_N can be well extrapolated with $A\approx 0.9743$ and $\alpha\approx 0.85$.

For periodic boundary conditions at the ends of the string the fermion momentum **k** is simply quantized: $k_{nx} = 2\pi n/(aN)$, where, for example, for even M the value $n = -M/2,\ldots,-1,0,1,\ldots,M/2-1$. With the use of Eq. (7) and the Pauli exclusion principle putting each fermion on a separate energy level we first calculate the total electronic energy of M fermions. Taking into account this electronic energy and the polarization energy of the crystal we obtain the expression for adiabatic potential $J_{N,M}$ in the form:

$$J_{N,M} = 6tM - \frac{2t(N-1)\sin(\pi M/N)}{N\sin(\pi/N)} - \frac{cM^2I_N}{2}\,, \tag{9}$$

where M is the number of trapped particles and the second term on the right-hand side of this equation is associated with the filling of the energy levels in the string by the fermions.

One sees from the dependence J on N and M that for a single trapped particle, i.e., $M = 1$, the lowest energy corresponds to a string with just one site, i.e., $N = 1$. This is an adiabatic small polaron in the PF model similar to the Holstein-Rashba polaron. Also, one sees that the value of the adiabatic potential $J_{N,M}$ decreases faster with M than it

increases with the length of the string N: $J_{N,M} \sim -\frac{AM^2}{N^\alpha}$. The strings with lowest energy have the same number of sites as the number of particles $M = N$. Also, the larger the value of N the lower the energy of the string. A string of infinite length, $N = \infty$, (a stripe) will have the lowest (ground state) energy. All of the above calculations are exact in the limit $Mc/t \to \infty$. However, in numerical investigations of small clusters (linear chains) consisting of from 2 to 11 atoms we have obtained results similar to the expression for the adiabatic potential, Eq. (9), which are valid for a wider range of values of the parameter c/t (see, for example, in Ref. [10]). This indicates that the described estimations (9) are valid for a wider range of the value of the parameter Mc/t than it was originally assumed.

To estimate the contribution to the total energy from the Coulomb forces between fermions we employ the HF many-body wave function of M self-trapped particles $\Psi(1, 2, \ldots, M)$ having the form of a Slater determinant of single particle wave functions, Eq. (6).

$$\Psi(1, 2, \ldots, M) = \frac{1}{\sqrt{M!}} \det \|\psi_i(k_j)\| . \qquad (10)$$

Such a choice of wave function proved to be good when describing the Coulomb correlations in the Hubbard model in the limit of strong Hubbard repulsion $U/t \to \infty$ [11]. After having calculated the pair correlation function, we then use this function to find the dependence of the Coulomb energy on N and M given by

$$V_{\mathrm{HF}} = \epsilon_c \frac{2M^2}{N} \int_0^\pi \frac{dx}{x} \left(1 - \frac{\sin^2(Mx)}{M^2 \sin^2 x}\right), \qquad (11)$$

where $\epsilon_c = \frac{e^2}{\bar{\epsilon} a}$ and the parameter $\bar{\epsilon}$ is the effective dielectric constant of the crystal. Numerical estimations of the integral (11) show that the function V_{HF} behaves similarly to that obtained independently in the electrostatic approximation where we assume that the M point charges are equidistantly located in the string. For such an assumption the long-range part of the Coulomb interaction of M particles with charge e separated by a distance aN/M and self-trapped into a string of a length N is approximately equal to

$$E_C \approx \frac{\epsilon_c M^2 \log M}{2N} . \qquad (12)$$

For the next illustration we use E_C instead of V_{HF} in the expression for the adiabatic potential (9). Because of the negative contribution in V_{HF} arising from exchange forces the value $V_{\mathrm{HF}} \leq E_C$. Such a substitution is therefore justified if we are interested in estimating an upper bound of the string Coulomb energy. Also it gives the explicit analytic expression (12) which is convenient to analyze. As a result the total energy which consists of the adiabatic potential J_{NM} and the energy of the Coulomb repulsion E_C equals $E_S(N, M) = J_{NM} + E_C$, where J_{NM} is defined in Eq. (9). For a fixed M the total energy always has a minimum when N is equal to

$$N \simeq \left(\frac{\epsilon_c \log M}{\alpha A c}\right)^\beta \quad \text{where } \beta = 1/(1 - \alpha) . \qquad (13)$$

The total energy $E_S(N, M)$ decreases rapidly with M and increases with N. Since the double occupation of the sites for the spinless fermions discussed is prohibited, the number M cannot be larger than N. Then, the minimum of the total energy E_S corresponds to the string with $M = N$ and this energy is given by

$$(E_S)_{\min} = 4tM - \frac{(1 - \alpha) c A M^{2-\alpha}}{2} , \qquad (14)$$

where the number of trapped fermions is determined with the aid of the equation

$$M = \left(\frac{\epsilon_c}{\alpha A c} \log M\right)^\beta . \qquad (15)$$

On the other hand, the energy of M separated individual polarons is estimated as equal to or greater than

$$J = MJ_{11} \geq 4tM - \frac{cMI_1}{2} . \qquad (16)$$

The inequality in this equation is realized when the interpolaron Coulomb repulsion is taken into account. The comparison of Eqs. (14) and (16) shows that the total energy of the string $(E_S)_{\min} \sim M^{2-\alpha}$ decreases faster with M than the energy of the individual polarons $J \sim M$. Therefore there is the critical value $M_c = (\beta I_1/A)^\beta \approx 5132$ so that if $M > M_c$ the energy of the string is smaller than the energy of M separated polarons: $(E_S)_{\min} < MJ_{11}$ and the string configuration corresponds to the ground state.

It is very interesting to compare this result with other calculations. Aubry *et al.* [4] have found exact results in the "anti-integrable limit" ($t = 0$) of HH model. Depending on the ratio between the Hubbard and EP coupling constants U/c there arises three types of states: bipolaronic, polaronic, and a mixture of bipolaronic and polaronic structures, respectively. Because of the short-range character of the Coulomb-Hubbard EE repulsion and the absolute localization ($t = 0$), these structures are strongly degenerate, i.e., have the same energy. For example, at large value $U/c \gg 1$, when a double occupation of the lattice sites is prohibited, the energy of any such polaronic structure does not depend on the distance between polarons: they *can collapse into a single blob of any shape* or are separated into a Wigner crystal having the same total energy. We obtain exactly the same result with the use of our HF approach applied to HH model. In this case the many-body HF wave function of the spinless fermions has the same form as a Slater determinant built up from single particle wave functions obtained from the solutions of the discrete nonlinear Schrödinger equation in the limit $cM/t \to \infty$ [10]. In spite of the different shapes and sizes for the localized blobs the energy per particle remains the same and is equal to a polaron shift. This degeneracy is broken when $t \neq 0$. Then, due to antiferromagnetic superexchange between electron spins, with the coupling

$J \sim t^2/U$, spin resonance bipolarons corresponding to the lowest energy state are created [4]. We were not able to get the spin resonance bipolarons since the HF method is not applicable for a description of the spin-correlation effects. However, with the aid of another many-body wave function with Jastrow factors which take spin-spin correlations into account properly this result was indeed reproduced [10]. This shows that the spin-spin interactions in the HH model gives rise to a bipolaron ground state and the very extensive results on the HH model [4] are consistent with calculations obtained with the aid of our approach.

The results obtained within the HH model are truly different in a fundamental way from those we have obtained within the PF model. The reason is that the long-range EE attraction mediated by phonons in the PF model breaks down the degeneracy in the "anti-integrable limit" between polarons and strings. We have found that for the larger length of the localized string, the media around the string is polarized more efficiently and, therefore, the energy per particle is lower (the polaron shift increases, proportional to the coupling constant). This is in contrast with the properties of polaronic structures (including strings) in "the anti-integrable limit" of the HH model where the energy per particle does not depend on the string length or any shape of the blob (due to the degeneracy discussed above) [4]. This polaron shift in the PF model does not depend on the spin-spin exchange constant which was most important for a bipolaron formation in HH model [4]. The long-range Coulomb repulsion between the spinless fermions partially compensates this energy decrease and breaks down the infinite strings into finite ones with a length defined by Eq. (13).

Although in the adiabatic limit for PF and HH model one recovers similar (although different) nonlinear equations, the results obtained are fundamentally different: in HH models there are bipolarons and *no strings* in the ground state [4], while in the PF model the ground state corresponds to a string configuration. Most importantly, the described phase separation may be studied with the aid of the Lang-Firsov transformation [12] that allows not only analytic study of the strong coupling arguments but also to extend the applicability of our results far beyond the adiabatic limit. With the use of this unitary transformation the EP interaction can be reduced to an effective EE attraction, which has long-ranged and short-ranged character for PF and HH models, respectively. After the next averaging over the phonons one obtains two models with different types of effective EE interaction. Then, it is clear that the PF model has a phase separated ground state associated with the long-range attraction between the heavy ($t \to 0$) polarons. Such an attraction between polarons in the HH model is absent and, therefore, in the HH model there is no phase separation and the ground state is associated with bipolarons [4].

Thus, we arrive at the conclusion that in polar narrow gap semiconductors small adiabatic polarons are unstable; this instability induces the formation of strings which are linear multiparticle objects. These strings are created by a polarization potential and have a length equal to the number of self-trapped fermions, which is determined by the dielectric constants of the semiconductor. The string may not have only a linear form but may also be bent, be curved, or even be a closed loop. Such curved configurations will probably correspond to low energy excitations of the string. The concept of the string is much more general. The strings may be both insulator and metal. Such a variety in the different types of strings may give rise to different novel effects which could arise in materials with narrow bands: both semiconductors and metals. In fact, for narrow band metals the criterion for the string formation is significantly improved. Here, we have to compare the total energy of the string (as described above) with the Fermi energy (which is much larger than the energy of separated polarons). But in metals the Coulomb repulsion is significantly screened. It seems that for such metals there may arise a coexistence of strings and free fermions, the balance of which is dictated by a competition of Fermi energy and Coulomb forces. These complicated issues will be discussed in our future publications.

I am very grateful to E. I. Rashba, P. Littlewood, and H. S. Dhillon for fruitful discussions.

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