Autoionization bands of electron–exciton complexes in periodic structures of reduced spatial dimension

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The problem of interaction in the periodic structures of reduced spatial dimension of the Feshbach-type, one-center resonance states is raised and solved. It is shown that as a result of a strong exchange interaction these structures form autoionization bands of two-particles bound states (electron–exciton complexes) whose imaginary part of the dispersion law is a nonanalytic function of energy and of quasi momentum. The predicted states could have been observed in the experiments on reflection of monoenergetic electrons from the surface of a solid (graphite or metal) covered by a submonolayer film of inert gas.

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1. Recently, the properties of electron–quasi particle complexes (bound pairs) whose composition includes a nonconserving particle–optical phonon, exciton, magnon, etc. (see, for example, Refs. 1–3) have been studied intensively in the theory of elementary excitations of solids.

However, heretofore the properties of only the bulk crystal structures have been investigated, and energies below the boundary of the vacuum continuum have been examined. The goal of this communication is to evaluate qualitatively the new features of the electron–exciton complexes (EEC) in the periodic structures of reduced spatial dimension–two-dimensional, one-dimensional, and semi-limited crystals. As is well known, the present-day investigation of the electronic spectra of such systems has been stimulated by the search for methods of producing high-temperature superconductivity and by numerous problems associated with the rapidly developing physics of surface phenomena.[4] We can expect that at positive energies (the boundary of the vacuum continuum is assumed to be the origin) the dispersion law for electron–exciton bound pairs is a complex nonanalytic function of energy and of quasi momentum because of the possibility of autoionization in the periodic structures that have a channel for a free departure of an electron to infinity.[5]

2. Let us examine the properties of EEC in terms of a simple, exactly solvable model which nonetheless preserves all qualitative features of the general case. We determine the spectrum of the eigenvalues of the Hamiltonian:

$$\hat{H}(\mathbf{r}) = -\frac{\Delta}{2}\hat{1} + \hat{H}_0 + \sum_{s=1}^{N} \hat{u}_s(r_s), \quad \mathbf{r}_s = \mathbf{r} - \mathbf{R}_s \tag{1}$$

$$(e = h = m = 1).$$

Here r is the coordinate of the electron, $\mathbf{R}_s$ are the radius vectors of the interaction

centers (atomic cores), $\hat{H}_0$ is the matrix Hamiltonian of the system of cores whose dimension is equal to $N+1$, and $\hat{1}$ is the unit matrix. Each atomic center is assumed to have two levels, and the possibility of simultaneous excitation of two centers in the investigated energy interval is disregarded. The $\hat{H}_0$ matrix has the form:

$$(\hat{H}_0)_{nn'} = (1-\delta_{n0})(1-\delta_{n'0})[\omega\delta_{nn'} + (1-\delta_{nn'})V(|n'-n'|)] \qquad (2)$$

[at $n \neq 0$ the index for the channel of electron motion coincides with the index of the center where (in zeroth approximation with respect to V) the excitation is localized. The channel with the index $|0\rangle$ corresponds to the motion of an electron in the field of the unexcited cores]. The matrices of the one-center interactions of the electron $\hat{u}_s$ have the form:

$$(\hat{u}_s)_{nn'} = u_0(r_s)\delta_{n0}\delta_{n'0} + u_1(r_s)\delta_{ns}\delta_{n's} + u_{01}(r_s)(\delta_{n0}\delta_{n's} + \delta_{ns}\delta_{n'0}). \qquad (3)$$

at $n, n' = 0, s$. When $n, n' \neq 0$, s $(\hat{u}_s)_{nn'} = u_0(r_s)\delta_{nn'}$. We use the method of zero-radius potentials,[6] which allows generalization to the case of the interacting scattering centers.[7] In the exciton representation (which diagonalizes the $\hat{H}_0$ matrix) the system of exact algebraic equations, which determine the spectrum of the resonance states of the Hamiltonian (1), has the form[1)]

$$\begin{aligned}
&(ik+\kappa_0)r_s(0) - 2\pi\sum_{s'\neq s} G_0^{(+)}(E, \mathbf{R}_s, \mathbf{R}_{s'})r_{s'}(0) \\
&\quad + \frac{\kappa_{01}}{\sqrt{N}}\sum_{p=1}^{N} e^{i\,2\pi/N(s-1)(p-1)} r_s(p) = 0, \\
&(ik(p)+\kappa_0)r_s(p) - 2\pi\sum_{s'\neq s} G_0^{(+)}(E(p), \mathbf{R}_s, \mathbf{R}_{s'})r_{s'}(p) \\
&\quad + \frac{\kappa_{10}}{\sqrt{N}} e^{-i\,2\pi/N\,(s-1)(p-1)} r_s(0) \\
&\quad + \frac{\kappa_1-\kappa_0}{N}\sum_{p'=1}^{N} e^{i\,2\pi/N\,(s-1)(p'-p)} r_s(p') = 0 \\
&(E(p) = k^2(p)/2 = E - \epsilon(p), \quad \mathbf{R}_s = (as, 0, 0), \quad N >> 1).
\end{aligned} \qquad (4)$$

In Eq. (4) $k = \sqrt{2E}$, E is the energy, κ_0, κ_1, and κ_{01} are the parameters of the one-center (two-channel) scattering of the electron by an isolated atomic center,[6] $\epsilon(p)$ is the spectrum of excitonic excitations of the Hamiltonian of the cores, $G_0^{(+)}(E,\mathbf{R}_s,\mathbf{R}_{s'})$ is the Green's function of the free motion of an electron with the energy E, and $\epsilon(p) \sim \omega + \beta\cos pa$.[2)]

We seek the solution of the system of equations (4) in the quasi-momentum representation:

$$\tau_s(0) = A(0)e^{iqas}, \qquad \tau_s(p) = A(p)\,e^{i(q-p)as}.$$

Finally, we obtain the following dispersion equation:

$$2\pi(\kappa_0 - \kappa_1 + \kappa_{01}^2 Z_0^{-1}(E, q))^{-1} = aD(E, q), \tag{5}$$

$$Z_0(E, q) = (ik + \kappa_0 - 4\pi \sum_{s=1}^{\infty} G_0^{(+)}(E, 0, as)\cos qas), \tag{6}$$

$$D(E, q) = \int_0^{2\pi/a} \frac{dp}{Z_1(E, p, q)}, \tag{7}$$

$$Z_1(E, p, q) = (ik(p) + \kappa_0 - 4\pi \sum_{s=1}^{\infty} G_0^{(+)}(E(p), 0, as)\cos(q-p)as),$$

which determines the total spectrum of the electronic and electron–excitonic states of the investigated system ($N \to \infty$).

In terms of the accepted interaction model the dispersion equation (5) is an exact equation. By varying in it the characteristic parameters, we can easily trace the limiting transition to different special cases examined earlier in Refs. 9–11 and also in the theory of bound subthreshold states of the electron with a nonconserved quasi particle[1] (the latter case corresponds to the conditions: $\kappa_1 = \kappa_0 \gg \beta a|e^{ik(p)a}|$). Using the dispersion equation (5), we examine the possibility for the existence in the periodic structures of the bound electron–exciton pairs whose excitation spectrum is set against the background of the continuum of the free electron states. Let $\kappa_{01} \ll \kappa_1$ and $\beta \neq 0$. In zeroth (with respect to the parameter κ_{01}/κ_1) approximation Eq. (5) determines the zone of the interacting but unbound electron–exciton states [the dispersion law for the latter is determined from the equation $Z_1(E, p, q) = 0$] and also the branch of the electron–exciton complex whose dispersion law can be determined from the equation

$$\frac{2\pi}{(\kappa_0 - \kappa_1)a} = D(E, q), \tag{8}$$

The real solutions of Eq. (8) $E^0(q)$ can be easily obtained analytically (in the approximation of strong coupling for the electronic and excitonic bands) and also numerically. On the basis of an estimating calculation, which was performed for a two-dimensional crystal lattice produced by Xe atoms at the surface of the graphite[3] [all characteristic parameters of the problem in this case are known: $\kappa_0 = -0.17$, $\kappa_1 = 0.2$, $\omega = 0.30$ (a.u.),[12] $\beta = 0.5$ eV, and $\alpha = 9$ (a.u.)[13]], we can conclude that in this case the branch of the EEC, which is situated ~ 0.5 eV from the boundary of the unbound electron–exciton states, is formed; here $|E_{\min} - E_{\max}| \sim 0.1 - 0.2$ eV.

The interaction of the channels ($\kappa_{01} \neq 0$) dampens the examined states, and we have

$$\mathrm{Im}E(q) = \frac{\kappa_{01}^2}{2\pi}\left(\frac{d}{dE}\,\frac{1}{D(E,q)}\right)^{-1}_{E=E^\circ(q)} \frac{\left(ka + \pi\left(1 - \left\{\frac{a(k+q)}{2\pi}\right\}\right)\left\{\frac{a(k-q)}{2\pi}\right\}\right)}{|Z_0(E^\circ(q), q)|^2} \tag{9}$$

$$(\mathbf{R}_s = (as, 0, 0)).$$

Here $\{x\}$ is a fractional part of x. Thus, the average lifetime of the complex (with respect to detachment of the electron from the structure) turns out to be a nonanalytic function of energy and of quasi momentum. At the points which are the roots of the equation

$$a(\sqrt{2E^\circ(q)} \pm q) = 2\pi m, \quad m = 0, +1, \ldots \tag{10}$$

In $E(q)$ changes abruptly by an amount of the order of its original value (for the one-dimensional structures). For the two-dimensional structures the singularities have a root-divergent nature. An estimate of Im $E(q)$ for xenon [the parameter κ_{01} was reconstructed from the known width of a one-center Feshbach resonance of a $(Xe^-)^{a.i.}$ ion $\Gamma_{Xe^-} \cong 3\times10^{-3} - 1\times10^{-2}$ eV[12]] gives a value of the order of 10^{-3} eV for $a(k+q)/2\pi \sim 1/2$.

3. From the physical point of view, the situation examined above corresponds to a strong exchange interaction of one-center Feshbach resonances.[4] Such resonances occur in almost all atomic particles,[14] which makes the prospect of observing experimentally the autoionization EEC bands rather encouraging. A branch of (electron–exciton) two-particle excitations in the two-dimensional crystals can appear in the most diverse physical experiments, for example, in the processes such as photodetachment of an electron from adsorbed atomic monolayers and also in the resonance reflection of electrons from the surface of a solid in the presence of an ordered monolayer of adsorbed atoms. In the last case one could measure the dependence (energy and angular) of the intensity ratio of the reflexes corresponding to the reflection of electrons from the monolayer and atoms of the substrate. The EEC should give a resonance dependence of the measurable values.

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[1)]For simplicity, we write Eq. (4) for a linear, array-type periodic structure. The fact that the solution can be generalized to an effectively more important case of two-dimensional crystals follows directly from the dispersion equation (5).

[2)]Analogous equations can also be derived within the framework of more realistic models such as the multichannel "muffin-tin" potentials.[8]

[3)]In going over to two-dimensional crystals the summation in the dispersion equation (5) is taken over the two discrete indices and the integration with respect to the two projections of the vector **p** is substituted for the integration with respect to $d\mathbf{p}$.

[4)]The autoionization bands produced as a result of a strong exchange interaction of potential ("shape") resonances are examined in a separate publication.[15]

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