Quantum polaron

A. O. Gogolin and A. S. Ioselevich
L. D. Landau Institute of Theoretical Physics, Academy of Sciences of the USSR, 142432, Moscow

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A polaron of a new type is proposed. In it, the localization of the electron is not accompanied by a deformation of the lattice, as in an ordinary polaron. The "clothing" on the polaron in this case arises from a local suppression of quantum fluctuations of the lattice. Such a polaron arises if the quadratic electron–phonon interaction is strong.

The polaron problem is an old one and is regarded as understood quite well (Ref. 1, for example). When the coupling is strong, the problem can be interpreted at a classical level: Nonzero mean displacements of ions from their equilibrium positions arise near an electron. The radius of the polaron may be either small (the electron is concentrated at one site) or large (the polaron is much larger than the lattice constant). By virtue of translational invariance, the polaron can of course move as a whole through the crystal, while having a large effective mass. This (ordinary) polaron arises as a result of an electron–phonon interaction which is linear in the lattice displacements.

However, an electron–phonon interaction is forbidden by the symmetry in the case of certain modes. For example, a linear interaction with any incompletely symmetric vibrations is forbidden. The interaction of an electron with such modes is quadratic, and in several cases it may be important. An example is the interaction of an electron with rotational modes in molecular crystals.[2] Another is the interaction with bending vibrations of planes in quasi-2D compounds, or in filaments in quasi-1D compounds. It is particularly important when the bending vibrations are coupled to a structural transition and are soft. This is precisely the situation which prevails in the high-T_c superconductor La_2CuO_4 (Ref. 3).

Could polarons form as a result of a quadratic electron–phonon interaction? To answer this question, we consider a simple model which describes an electron that is interacting locally with the system of dispersion-free phonons. The Hamiltonian of this model is

$$\mathcal{H} = \frac{t}{2}\sum_{<\vec{n},\vec{n}'>}(a^+_{\vec{n}'} - a^+_{\vec{n}})(a_{\vec{n}'} - a_{\vec{n}}) + \sum_{\vec{n}}\left\{-\frac{\hbar^2}{2M}\frac{\partial^2}{\partial Q^2_{\vec{n}}} + \frac{1}{2}[M\omega_0^2 + \gamma a^+_{\vec{n}}a_{\vec{n}}]Q^2_{\vec{n}} - \frac{1}{2}\hbar\omega_0\right\}. \quad (1)$$

Here the operators $a^+_{\vec{n}}$ and $a_{\vec{n}}$ create and annihilate an electron, t is the electron hopping integral, $Q_{\vec{n}}$ is the magnitude of the displacement at site $\vec{n}$, M is the mass, ω_0

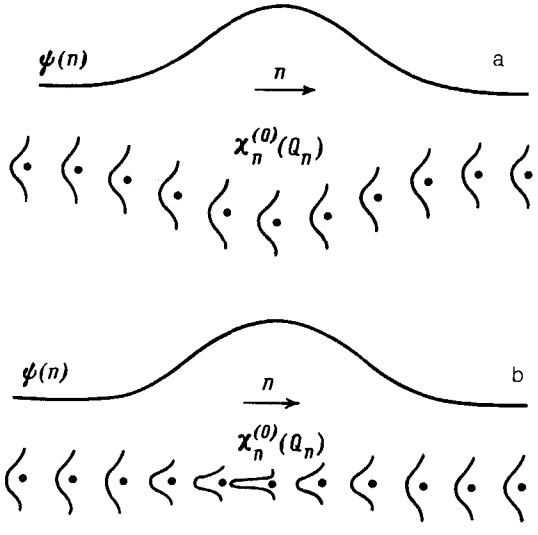

FIG. 1. a—Classical polaron, with displacements of the centers of oscillators; 1 b—quantum polaron, with zero-point vibrations suppressed.

is the frequency of the oscillator, and γ is the quadratic electron–phonon coupling constant. The energy is reckoned from the ground state of the noninteracting system.

If γ is small, the electron–phonon coupling can be dealt with by perturbation theory, and it does not lead to the formation of a polaron. If γ is instead large, then we cannot use a perturbation theory; i.e., the ground state of the system must undergo a pronounced restructuring. Just how this restructuring proceeds in the case of a large negative value of γ is understandable: The oscillator stiffnesses near an electron go negative (there is an instability). As a result, large spontaneous displacements $\langle Q \rangle$ develop. Such displacements are stabilized exclusively by nonlinear effects [which are not considered in Eq. (1)]. A state of this sort is qualitatively the same as an ordinary polaron which arises by virtue of a linear electron–phonon interaction.

If γ is instead large and positive, the oscillator stiffnesses can only increase, so there can be no instability, and there can be no $\langle Q \rangle$. There is accordingly no polaron effect (in the usual understanding of this term), and it might seem that the electron should be delocalized at any, arbitrarily large values $\gamma > 0$. That is not the case, however, since the corrections to the delocalized state calculated by perturbation theory are large. We will now show that the ground state corresponds to a self-localized electron again in this case. The localization in this case, however, results not from the appearance of average displacements $\langle Q \rangle$ but from a local suppression of quantum fluctuations of Q (Fig. 1). We refer to this formation as a "quantum polaron."

We first assume that the electron is localized at one site (i.e., we assume a small-radius polaron). The energy of such a state is

$$E_p = Dt + \frac{1}{2}\hbar\sqrt{\Omega^2 + \omega_0^2} - \frac{1}{2}\hbar\omega_0, \tag{2}$$

where $\Omega = \sqrt{\gamma/M}$, and we have restricted the discussion to the case of a D-dimensional cubic lattice. It is just as easy to calculate the energy of the free state, in which the electron is completely delocalized:

$$E_f = \frac{1}{2}\gamma < Q^2 >= \hbar\gamma/4M\omega_0 = \hbar\Omega^2/4\omega_0. \tag{3}$$

We see that under the condition $\hbar\Omega \gg (Dt, \hbar\omega_0)$ the polaron state ($E_p < E_f$) is the ground state. Why? The reason is that the electron stiffens the nearby oscillators. This stiffening results in a decrease in $\langle Q^2 \rangle$ and, according to (1), a decrease in the interaction energy.

It would be difficult to expect a small-radius polaron to arise, however, since the width of the electron band, Dt, would have to be small in comparison with the frequency $\hbar\Omega$, which is usually on the order of the Debye frequency.

A more realistic situation is

$$\hbar\omega_0 \ll \hbar\Omega \ll Dt. \tag{4}$$

Under these conditions a large-radius polaron can arise. In the adiabatic approximation, the wave function of the system factorizes:

$$\Phi(\vec{n}, \{Q_{n'}\}) = \psi(\vec{n})\Pi_{n'}\chi_{n'}^{(0)}(Q_{n'}), \tag{5}$$

where $\chi_n^{(0)}(Q_n)$ is the wave function of the ground state of oscillator n, with the renormalized frequency

$$\omega_n = \sqrt{\omega_0^2 + |\psi_n|^2\Omega^2}. \tag{6}$$

The electron wave function ψ must be determined by minimizing the adiabatic energy functional

$$J\{\psi\} = \int d^D\vec{x}\left\{\frac{t}{2}|\nabla\psi|^2 + \frac{1}{2}\hbar[\omega(\vec{x}) - \omega_0]\right\}. \tag{7}$$

Expression (7) is written in the continuum approximation, which is justified by inequality (4). The lattice constant is $a_0 = 1$, so $\vec{x}$ and $\psi(\vec{x})$ are dimensionless. The quantum-mechanical nature of the effect can be seen explicitly in (7): The second term, with Planck's constant $\hbar$, is the energy of zero-point vibrations, modulated by the presence of an electron.

Let us consider the strong-coupling limit. In other words, we ignore ω_0 in comparison with $|\psi|\Omega$ in (6) and (7). We then find

$$J\{\psi\} = \int d^D\vec{x}\left\{\frac{t}{2}|\nabla\psi|^2 + \frac{\hbar\Omega}{2}|\psi|\right\}. \tag{8}$$

We can easily find an estimate of the minimum of functional (8) and of the radius of the corresponding localized state, a, from the virial theorem. The kinetic energy is $\sim t/a^2$, and the potential energy $\sim\hbar\Omega a^{D/2}$, so we find

$$a \sim \left(\frac{t}{\hbar\Omega}\right)^{\frac{2}{D+4}} \gg 1, \quad J \sim \hbar\Omega \left(\frac{t}{\hbar\Omega}\right)^{\frac{D}{D+4}}. \tag{9}$$

Since the characteristic values here are $|\psi| \sim a^{-D/2}$, the strong-coupling condition $\omega_0 \ll |\psi|\Omega$ holds if

$$\frac{\Omega}{\omega_0}\left(\frac{\hbar\Omega}{t}\right)^{\frac{D}{D+4}} \gg 1. \tag{10}$$

The adiabatic condition—the condition that the characteristic electron time $\tau_e \sim \hbar/J$ be much shorter than the characteristic phonon time, $\tau_{ph} \sim \hbar/|\psi|\Omega'$,—holds. It is the same as the condition $\tau \gg \hbar\Omega$.

Interestingly, a quantum polaron differs from an ordinary polaron in that it can have a large radius in any dimensionality, despite the local nature of the electron-phonon coupling.

Because of the simple form of functional (8), we can find its extrema exactly. In the 1D case, for example, we find

$$\psi(x) = \begin{cases} \frac{2}{\sqrt{3}a}\cos^2\left(\frac{\pi x}{2a}\right), & |x| < a, \\ 0, & |x| > a, \end{cases} \tag{11}$$

where $a = (2\pi^2 t/\sqrt{S\hbar\Omega})^{2/5}$ and $J = (5\hbar\Omega/4\sqrt{S})\left[2\pi^2 t/\sqrt{3\hbar\Omega}\right]^{1/5}$. The extremum of our original functional (7) differs from (11) in the presence of an exponential tail; this tail is joined in a narrow region near $x = a$. We will not reproduce here the lengthy expressions for the extrema in the cases $D = 2$ and 3. They can be constructed from the free solution of the Schrödinger equation, as in the $D = 1$ case (Fig. 2).

The quantum-polaron problem has two dimensionless parameters, $\mu = \Omega t$ and $\alpha = \Omega/\omega_0$. At $\alpha < 1$ the electron is always (regardless of μ) delocalized; the corrections to this state are small. With $\alpha > 1$ and $\mu > 1$, a small-radius polaron forms.

With $\alpha > 1$ but $\mu < 1$ [i.e., under inequality (4)], either a large-radius polaron or a delocalized state forms (Fig. 3).

Let us take a brief look at the details associated with the dimensionality D. At $D = 1$, "weak" polarons, in which the frequency changes are small, arise along with the "strong" large-radius polarons described above, over a broad region of parameter values. These weak polarons are described by the ordinary nonlinear Schrödinger

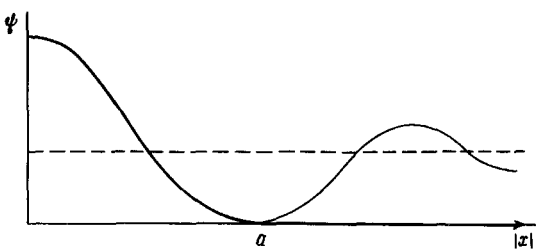

FIG. 2. Construction of an extremum of functional (8). The solution of the free Schrödinger equation has been shifted to bring its first minimum to the abscissa.

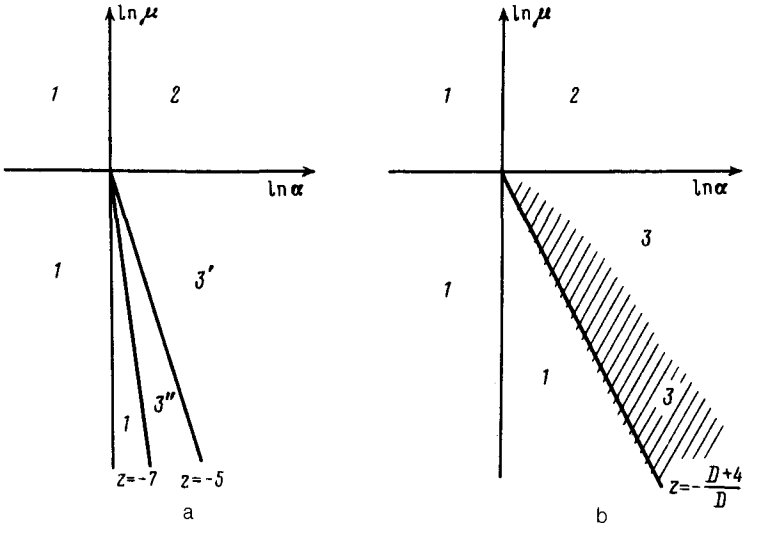

FIG. 3. Regions of the parameters μ and α corresponding to various types of ground states. 1—The electron is delocalized, and perturbation theory can be used; 2—small-radius polaron; 3—large-radius polaron (the parameter z is the slope of the line). a: $D = 1$. 3′, 3″) "Strong" and "weak" large-radius polarons. b: $D = 2,3$. The hatched region is the region in which a free state and a polaron state coexist (this region exists in the case $D = 3$).

equation which arises in an expansion of (6) and (7) to up to fourth order in ψ. In the $D = 2$ case the situation is qualitatively the same as at $D = 1$, but the region of weak polarons is very narrow. At $D = 3$ there can be no weak polarons. As α is raised, a self-localized state arises immediately as a strong large-radius polaron. This state is initially metastable; the free state subsequently becomes metastable. In a certain region of parameter values, the free and localized states are separated by a barrier and can coexist, as in the case of ordinary polarons.[4]

One can show that the mass of a quantum polaron, like that of an ordinary polaron, is large in comparison with the mass of a bare electron. It is not a trivial matter to calculate the mass of a quantum polaron. A corresponding calculation will be published separately.

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[1]J. Apple, *Polarons* [Russian translation], Nauka, Moscow, 1975.
[2]H. Gutfreund and M. Weger, Phys. Rev. B **16**, 1753 (1977).
[3]P. Böni *et al.*, Phys. Rev. B **38**, 185 (1988).
[4]É. I. Rashba, Opt. Spektrosk. **2**, 78, **88** (1957).

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