Comment on ''Adiabatic theory for the bipolaron''

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(Received 28 June 1995)

Comments are given on the application of the Bogoliubov-Tyablikov approach to the bipolaron problem in a recent paper by Lakhno [Phys. Rev. B **51**, 3512 (1995)]. This author believes that his model (1) is the translation-invariant adiabatic theory of bipolarons and (2) gives asymptotically exact solutions in the adiabatic limit while the other approaches are considered as either phenomenological or variational in nature. Numerical results by Lakhno are in contradiction with all other papers published on the subject because his model leads to much lower energies. Thus, the author concludes that bipolarons ''are more stable than was considered before.'' We prove that both the analytical and the numerical results presented by Lakhno are wrong.

The conventional Hamiltonian to describe two nonrelativistic particles interacting with a scalar field

$$H=-\frac{\hbar^2}{4m}\nabla_R^2-\frac{\hbar^2}{m}\nabla_r^2+\sum_{\vec{k}}\hbar\omega_{\vec{k}}a_{\vec{k}}^{\dagger}a_{\vec{k}}+U(r)$$
$$+\sum_{\vec{k}}2\cos\frac{\vec{k}\cdot\vec{r}}{2}(a_{\vec{k}}V_{\vec{k}}e^{i\vec{k}\cdot(\vec{R}-\vec{r}_0)}+\text{H.c.}) \qquad (1)$$

is used as the starting point in Ref. 1. Here $\vec{R}$ and $\vec{r}$ are operators of the center of mass (c.m.) and the relative coordinates, respectively, m is the particle's (bare) mass, and $a_{\vec{k}}^{\dagger}$ ($a_{\vec{k}}$) are the creation (annihilation) operators of the field quantum with the wave vector $\vec{k}$ and the frequency $\omega_{\vec{k}}$. The coefficients $V_{\vec{k}}$ are the Fourier transform of the particle-field interaction and $U(r)$ stands for the direct interaction of the particles. Following the paper[1] of Lakhno we insert here an arbitrary vector $\vec{r}_0$, which plays no role and results simply in a constant phase of the field plane waves. It can be included into $V_{\vec{k}}$ and because all results depend only on absolute values $|V_{\vec{k}}|^2$ this vector $\vec{r}_0$ can be erased. Nevertheless we will keep it because of its dramatic role in the considerations presented in Ref. 1.

The idea of Bogoliubov[2] and of Tyablikov[3] developed initially for a single particle interacting with a quantum field is based on an elegant physical picture of the particle motion. The system is translation invariant, and the symmetry group is defined by

$$\vec{R}\to\vec{R}+\vec{R}_0,\quad a_{\vec{k}}\to a_{\vec{k}}e^{-i\vec{k}\cdot\vec{R}_0}. \qquad (2)$$

Correspondingly, the total momentum $\vec{P}=-i\hbar\nabla_R+\hbar\Sigma_{\vec{k}}\vec{k}a_{\vec{k}}^{\dagger}a_{\vec{k}}$ is conserved. Bogoliubov and Tyablikov suggested splitting up the particle coordinate operator into a translation-invariant part which is responsible for the momentum conservation and an internal quantum vibrational part. Applied to the two-particle problem this idea can be realized with the transformations

$$\vec{R}=\vec{q}+\vec{\lambda},\quad a_{\vec{k}}=b_{\vec{k}}e^{-i\vec{k}\cdot\vec{q}}, \qquad (3)$$

where $b_{\vec{k}}$ are new field operators. Then the group defined by Eq. (2) can be considered as corresponding to the transformation $\vec{q}\to\vec{q}+\vec{R}_0$ of the translation-invariant operator only, while the quantum vibrational coordinate operator $\vec{\lambda}$ and the field operators $b_{\vec{k}}$ are not influenced.

The decisive mistake was made by Lakhno just at the first step of his attempt to apply the Bogoliubov-Tyablikov concept to the two-particle problem. He performed the splitting of the c.m. radius-vector operator as follows: $\vec{R}=\vec{q}+\vec{r}_0$ [see Eq. (20) of Ref. 1]. As a result, the dependence on the vibrational part disappeared from the interaction term. Thus, Lakhno supposed the equivalence of the *constant c-number* vector $\vec{r}_0$ (which has no conjugate momentum and commutes with all operators in the Hamiltonian) to the *operator dynamical variable* $\vec{\lambda}$ which is not conserved. After the Hamiltonian (1) was broken so crudely one can no longer hope for an adequate description of the two-particle problem.

Because the dependence on the vibrational operator variable $\vec{\lambda}$ disappeared from Lakhno's transformed Hamiltonian, his scheme is equivalent to the ''approximation'' in which one just sets the c.m. coordinate operator $\vec{R}$ (which does not commute with the Hamiltonian) equal to a fixed vector $\vec{r}_0$. Doing so one would arrive, instead of (1), at the Hamiltonian

$$H_L=-\frac{\hbar^2}{4m}\nabla_R^2-\frac{\hbar^2}{m}\nabla_r^2+\sum_{\vec{k}}\hbar\omega_{\vec{k}}a_{\vec{k}}^{\dagger}a_{\vec{k}}+U(r)$$
$$+\sum_{\vec{k}}2\cos\frac{\vec{k}\cdot\vec{r}}{2}(a_{\vec{k}}V_{\vec{k}}+a_{\vec{k}}^{\dagger}V_{\vec{k}}^{*}). \qquad (4)$$

This is in essence a one-particle Hamiltonian which is evidently *not equivalent* to the initial Hamiltonian (1). Because the c.m. position operator is replaced by its classical value at the potential minimum, the corresponding energy levels of (4) will be much lower in comparison with those of (1). To simplify our analysis we consider in what follows bipolarons at rest. Dealing with the Hamiltonian (4) one should note that to the leading order of the adiabatic limit the field operators may be safely replaced by their classical parts so that one can consider $a_{\vec{k}}, a_{\vec{k}}^{\dagger}$ as commuting c numbers. Then one can

TABLE I. The ground-state energy of the (broken) bipolaron in units $\hbar\omega_{\rm LO}\alpha^2$ is given for different values of $\eta=\epsilon_\infty/\epsilon_0$. The second row presents the results E_L by Lakhno (Ref. 1). Upper bounds $E_{L,1}$ and $E_{L,2}$ to the same energies obtained with the trial wave functions (9) and (11), respectively, are shown in the 3rd and 4th rows.

η	0	0.053	0.094	0.132	0.166	0.199	0.228	0.256	0.282	0.305	0.317
E_L	-1.31	-1.22	-1.15	-1.08	-1.02	-0.95	-0.89	-0.83	-0.76	-0.72	-0.69
$E_{L,1}$	-1.36	-1.28	-1.22	-1.15	-1.09	-1.03	-0.97	-0.92	-0.86	-0.81	-0.78
$E_{L,2}$	-1.41	-1.33	-1.25	-1.19	-1.12	-1.05	-0.99	-0.93	-0.87	-0.82	-0.79

readily check that the Hamiltonian (4) *completely* coincides with Lakhno's H_0 [see Eq. (28) of Ref. 1] taken for the system at rest.

The wave function of the relative motion $\phi(\vec{r})$ obeys the Schrödinger equation $H_L\phi(\vec{r})=E_L\phi(\vec{r})$. Minimizing E_L with respect to (commuting) $a_{\vec{k}}, a_{\vec{k}}^\dagger$ one arrives at the Pekar type of energy functional

$$E_L=\frac{\hbar^2}{m}\int|\nabla_r\phi(\vec{r})|^2d\vec{r}+\int d\vec{r}U(r)|\phi(\vec{r})|^2 - \sum_{\vec{k}}\frac{|V_{\vec{k}}|^2}{\hbar\omega_{\vec{k}}}\left(\int 2\cos\frac{\vec{k}\cdot\vec{r}}{2}|\phi(\vec{r})|^2d\vec{r}\right)^2. \quad (5)$$

The same expression follows from Eqs. (35)–(38) of Ref. 1. Minimizing E_L further with respect to the wave function $\phi(\vec{r})$ one finds the (supposed) ground-state energy. Evidently, because the Hamiltonian H_L is not equivalent to the initial Hamiltonian (1) one does not arrive with the energy functional (5) at the correct solution to the initial two-particle problem.

One can look at Lakhno's mistake from another point of view. His first transformation is nothing more than a shift of the c.m. position operator by a constant vector $\vec{r}_0$. Thus his $\vec{q}$ coincides in essence with $\vec{R}$ and *not* with its translation-invariant part. Then the second transformation of field operators should lead to the replacement of $(-i\hbar\nabla_R)^2$ in the first kinetic term of Eq. (4) by $(-i\hbar\nabla_R-\hbar\Sigma_{\vec{k}}\vec{k}a_{\vec{k}}^\dagger a_{\vec{k}})^2$. With this correction the transformed Hamiltonian would be equivalent to the initial one. The conserved total momentum indeed becomes $-i\hbar\nabla_R$, but the price one pays is the appearance of the quartic term in the field operators. Such a transformation was introduced by Bogoliubov,[2] Tyablikov,[3] and Lee, Low, and Pines[4] for single polarons and indeed was used for bipolarons too (cf. Refs. 5 and 6). But it is certainly not the Bogoliubov-Tyablikov adiabatic theory. Because the quartic term is missed in Lakhno's scheme, the field contribution to the total conserved momentum is neglected. In other words, Lakhno supposed implicitly the equivalence of the *two particles* c.m. coordinate operator to the c.m. coordinate operator of their *bound state* which is not the same in the presence of the field.

Applying the Bogoliubov-Tyablikov ideas in a correct way one *never* arrives at a one-particle Hamiltonian. Instead, one obtains — to leading order — the equations derived using another approach by the present authors and coworkers in Ref. 7. In the application to bipolarons this leads to the same energy functional which appeared in the pioneering paper by Pekar and Tomasevich[8] and which was used by many authors afterwards (for the references see the review paper[6]).

The application to the bipolaron problem is specified by relations $\omega_{\vec{k}}=\omega_{\rm LO}$ and

$$V_{\vec{k}}=-i\hbar\omega_{\rm LO}\left(\frac{4\pi\alpha}{Vk^2}\sqrt{\frac{\hbar}{2m\omega_{\rm LO}}}\right)^{1/2}, \quad U(r)=\frac{e^2}{\epsilon_\infty r}. \quad (6)$$

The Fröhlich electron-phonon coupling constant is defined in the standard way

$$\alpha=\frac{1}{\hbar\omega_{\rm LO}}\frac{e^2}{\sqrt{2}}\left(\frac{1}{\epsilon_\infty}-\frac{1}{\epsilon_0}\right)\sqrt{\frac{m\omega_{\rm LO}}{\hbar}}, \quad (7)$$

and depends on the static (ϵ_0) and high-frequency (ϵ_∞) dielectric constants. In what follows we use the ratio of the dielectric constants $\eta=\epsilon_\infty/\epsilon_0$ so that the Coulomb repulsion can be written as

$$U(r)=\hbar\omega_{\rm LO}\frac{\sqrt{2}\alpha}{1-\eta}\frac{\sqrt{\hbar/m\omega_{\rm LO}}}{r}. \quad (8)$$

To check the numerical calculations by Lakhno we use one of the trial wave functions of the relative motion suggested in Ref. 5:

$$\phi(r)=\sqrt{\frac{b^5}{3\pi}}re^{-br}, \quad (9)$$

where b is a variational parameter. This wave function has a node for $r=0$. This prevents the two particles from gluing together — a feature consistent with the Coulomb repulsion. Performing all integrations and minimizing the energy functional (5) with respect to b we arrive at the following upper bound for the (supposed) bipolaron ground-state energy

$$E_{L,1}=-\hbar\omega_{\rm LO}\alpha^2\frac{3}{8}\left(\frac{93}{32}-\frac{1}{1-\eta}\right)^2, \quad (10)$$

which is valid under the constraint that the expression in parentheses is positive (that is, $\eta\leqslant 1-32/93\approx 0.6559$). In Table I we present results obtained in Ref. 1 and energy values which follow from Eq. (10). We use also the trial wave function with two variational parameters

$$\phi(r)=N(1+dbr)e^{-br}, \quad N=\sqrt{\frac{b^3}{\pi(3d^2+3d+1)}}, \quad (11)$$

which gives a good approximation (to within 0.45%) to the exact numerical calculations of the single-polaron ground-state energy in the strong-coupling limit.[9] The results obtained ($E_{L,2}$) are shown also in Table I. Let us stress once more that both the energies obtained by Lakhno and our variational estimates for the same model are much lower (by a factor of about 5 at $\eta=0$) than the actual bipolaron energies. Surprisingly our variational *upper* bound although close to the values given by Lakhno is *lower* than his energy. Energies obtained with the trial wave function (11) differ from the results of Ref. 1 by 7.6% at $\eta=0$ and by 14.5% at $\eta=0.317$. Because Lakhno supposes to have obtained exact numerical solutions to his equations (although they are not relevant to bipolarons) we must conclude that something is wrong also with his numerical work.

For the bipolaron to be stable its energy should be lower than twice the single-polaron energy. Because the latter is known[9] to be $E_{\rm pol}=-\hbar\omega_{\rm LO}\alpha^2(0.108513)$ for large α, we obtain from Eq. (10)

$$\frac{3}{8}\left(\frac{93}{32}-\frac{1}{1-\eta}\right)^2 \geqslant 0.217024 \Rightarrow \eta \leqslant 0.5339. \qquad (12)$$

Thus, $\eta_c \approx 0.53$ in this scheme of Lakhno which is significantly larger than the critical values $\eta_c \approx 0.13$–0.14 obtained by other authors. This should not be a surprise anymore. But Lakhno declared his result to be $\eta_c=0.31$. The discrepancy with (12) is too large and has to be addressed. The explanation is that Lakhno took the wrong expression $E_{\rm pol}=-\hbar\omega_{\rm LO}\alpha^2(0.3255)$ for the single-polaron energy [see Eq. (60) of Ref. 1], which is certainly a mistake. If he would have avoided it he could have reported an even larger bipolaron stability region with $\eta_c=0.53$ (which would, of course, be wrong also).

With the trial wave function (11) the energy reaches its minimum at finite values of d if $\eta<13/29\approx 0.448276$. At larger η the minimum is reached at $d\to\infty$ which gives the same results as the wave function (9). Thus, at small repulsion $|\phi(0)|^2$ is finite while at larger repulsion it decreases and reaches zero at $\eta\approx 0.448$. Then the energy is given by Eq. (10) and finally the ''bipolaron'' decays into two polarons at $\eta\approx 0.53$. In Lakhno's work a similar peculiarity can be observed somewhere in the range $\eta=0.58$–0.65 (see Fig. 1 of Ref. 1). That is, his ''bipolaron'' decays for lower values of η than for what the constituent particles would separate.

It is evident that to compare the bipolaron energy and the single-polaron energy they should be calculated within the same approximation. In the strong-coupling limit the leading terms of both energies are of order $O(\alpha^2)$. As a consequence, the critical value α_c for the bipolaron stability cannot be determined from the standard equation $E_{\rm bip}\leqslant 2E_{\rm pol}$ when only the leading terms are taken into account. In this way it is only possible to determine η_c and this has been done by Lakhno too. As for α_c one has to calculate corrections $O(\alpha^0)$ both for bipolarons and polarons [see, e.g., Ref. 10]. But Lakhno escaped this difficulty only by comparing the bipolaron energy in the *strong*-coupling limit with twice the single-polaron energy taken in the leading order of the *weak*-coupling limit ($-2\hbar\omega_{\rm LO}\alpha$). This is inconsistent. The critical value α_c corresponds to the smallest value $\eta=0$ (that is, to the smallest repulsion between polarons). Taking the variational estimate to the (supposed) bipolaron energy $E_{L,2}(\eta=0)=-1.41\hbar\omega_{\rm LO}\alpha^2$ (see Table I) this inconsistent way even gives $\alpha_c=2/1.41=1.42$ (Lakhno reported $\alpha_c=1.54$). The conventional values obtained in the literature are in the range $\alpha_c=6.8$–7.2. Lakhno presented some arguments why bipolarons could be in the strong-coupling regime while polarons are still in the weak-coupling regime. Even if these arguments were valid, then he could not derive η_c by comparing strong-coupling energies of a bipolaron and of two polarons. When a bipolaron decays into two polarons the latter could be either in a weak- or in a strong-coupling regime but not in both. Thus, Lakhno's two different ways to find the critical values η_c and α_c for the bipolaron stability region are mutually inconsistent.

It should be noted that the Bogoliubov-Tyablikov approach was applied to the two-particle problem already in Ref. 11. The authors used the correct method but they split up both the c.m. and the relative-coordinates operators. This made the scheme much more complicated. No numerical results were reported. We see no reason to split up the relative coordinate operator, although it would be interesting to check if this approach could give something new.

To summarize, the analysis of Ref. 1 leads us to the following conclusions:

(i) The underlying mathematics of Ref. 1 does not correspond to the ideas of Bogoliubov[2] and of Tyablikov.[3] The theoretical scheme developed by Lakhno contains a crucial mistake, so he considered a system which is not equivalent to the initial one.

(ii) The numerical calculations performed in Ref. 1 are not correct, not even in the scope of the considered model.

(iii) The determination of the critical parameters is not consistent and contains a mistake.

Thus, the results of this paper as a whole are completely wrong. We would like to stress once more that applying the Bogoliubov-Tyablikov method in a correct way one arrives to the leading order of approximation at the same results as in the more conventional approaches.

We should note also that Lakhno published other papers using the same approach to the two-particle problem in the adiabatic limit. Among them there are papers on bipolarons[12,13] and deuterons.[14] Because our main comment concerns the essense of the method it does not depend on the specific interaction between the two particles. Thus, all cited results are not reliable as well.

The authors are grateful to V. M. Fomin, H. Leschke, E. P. Pokatilov, and F. M. Peeters for discussions. One of us (M.A.S.) discussed with V. D. Lakhno problems of the adiabatic theory and the results obtained by him and would like to thank him although an agreement was not found. M.A.S. is grateful to the University of Antwerpen (UIA) for the support and kind hospitality during his visit to Belgium. This work is partly performed in the framework of the Human Capital and Mobility Program ''Polarons, bi-polarons and excitons. Properties and occurrences in new materials'' (C.E.C. No. CHRX-CT93-0124).

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