Asymptotic expansions in the path-integral approach to the bipolaron problem

M. A. Smondyrev,* J. T. Devreese,† and F. M. Peeters‡
Departement Natuurkunde, Universitaire Instelling Antwerpen, Universiteitsplein 1, B-2610 Wilrijk, Belgium
(Received 17 January 1995)

Large bipolarons are studied in two (2D) and three (3D) space dimensions. The bipolaron energy is expanded in inverse powers of the electron-phonon coupling constant α, which leads to $E_{\text{bip}} = -(2\alpha^2/3\pi)\, A(u) - B(u) + O(\alpha^{-2})$, where $u = U/\alpha$ and U is the dimensionless Coulomb-repulsion coupling constant. We derive closed analytical formulas for the coefficients $A(u)$ and $B(u)$ which allow us to find the bipolaron stability region both in the scope of a model formulated in terms of Feynman path integrals and from fitting known results. Analytical expressions for the leading terms of the bipolaron effective mass and mean-square separation between electrons are also presented.

I. INTRODUCTION

The aim of the present work is to establish analytical expansions for the characteristics of large bipolarons in inverse powers of the Fröhlich electron-phonon coupling constant α. We will base our analysis mostly on expressions derived in a paper by Verbist, Peeters, and Devreese[1] in the scope of the Feynman path-integral formulation of quantum mechanics and its application to the polaron theory. Conventional perturbation methods fail to describe bipolaron states because of the simple fact that bipolarons do not exist at small values of the coupling constant. Physically for bipolarons to exist it is necessary that the attraction due to phonon exchange overcomes the Coulomb repulsion. Therefore it is the strong-coupling mechanisms that underlie the bipolaron dynamics. The availability of analytical expressions in general clarifies the main features of any physical theory. In this study we derive analytical strong-coupling asymptotic expansions for the bipolaron self-energy, including the two leading terms, which allows a meaningful extrapolation to intermediate coupling.

For single polarons the basic ideas of the strong-coupling regime have been put forward in pioneering investigations by Landau, Pekar, Bogolubov, and Tyablikov.[2] Numerical calculations for three-dimensional (3D) polarons have been performed by Miyake[3] and for 2D polarons by Wu, Peeters, and Devreese.[4] Although there exists no obvious distinction between weak- and strong-coupling regimes for free polarons,[5,6] the study of the response function reveals the gradual appearance of internal excited states[7] for increasing coupling strength. Sharp resonances exist for large values of the electron-phonon coupling constant α, while at small values of α the response function is characterized by broad resonances due to scattering states. As it follows from the numerical results of Ref. 7 the region around $\alpha \approx 6$ can be considered as a diffuse boundary between the weak- and strong-coupling regimes.

As was mentioned, bipolarons only exist in the strong-coupling regime, that is, in the region $\alpha > 6$ (for the 3D case). Consequently, the strong-coupling limit becomes an essential feature of any theory of bipolarons. To the best of our knowledge, for bipolarons no systematic strong-coupling expansions with analytical coefficients have been published and most work relies on a numerical analysis. The study of bipolarons is of possible relevance in the understanding of high-T_c superconductivity (e.g., for high-T_c superconductors,[8,9] the recently discovered fullerites,[10,11] and the proposed Bose-Einstein condensation of large bipolarons[12,13]).

The present paper is organized as follows. In Sec. II we describe the bipolaron model formulated in Ref. 1. In Secs. III and IV the strong-coupling expansion for the bipolaron energy is found and two leading terms are calculated explicitly. In Sec. V the bipolaron stability region is studied. An extrapolation of results obtained is given to proceed beyond the model of the paper.[1] The effective bipolaron mass and its extension are estimated in Sec. VI. The study is made both for 3D bipolarons and for bipolarons in reduced space dimensions.

II. THE BIPOLARON MODEL

The Hamiltonian for three-dimensional (3D) Pekar-Fröhlich bipolarons is given by

$$H = \frac{\vec{p}_1^{\,2}}{2m} + \frac{\vec{p}_2^{\,2}}{2m} + \sum_{\vec{k}} \hbar\omega_{\vec{k}}\; a^\dagger_{\vec{k}} a_{\vec{k}} + \frac{e^2}{\epsilon_\infty \mid \vec{r}_1 - \vec{r}_2 \mid} + \sum_{\vec{k}} \left[a_{\vec{k}}\, V_{\vec{k}} \left(e^{i\vec{k}\cdot\vec{r}_1} + e^{i\vec{k}\cdot\vec{r}_2} \right) + \text{H.c.} \right] , \qquad (2.1)$$

where $\vec{r}_i(\vec{p}_i)$ are the positions (momenta) operators of the ith electron, m is the electron band mass, and $a^\dagger_{\vec{k}}$ $(a_{\vec{k}})$ are the creation (annihilation) operators of phonons with wave vector $\vec{k}$ and frequency $\omega_{\vec{k}}$. The quantities $V_{\vec{k}}$ are the Fourier transforms of the electron-phonon interaction. For LO phonons $\omega_{\vec{k}} = \omega_{\text{LO}}$ and $V_{\vec{k}}$ takes the form

$$V_{\vec{k}} = -i\hbar\omega_{\rm LO}\left(\frac{4\pi\alpha}{Vk^2}\sqrt{\frac{\hbar}{2m\omega_{\rm LO}}}\right)^{1/2}. \tag{2.2}$$

The dimensionless Coulomb potential and electron-phonon coupling constants are defined in the standard way,

$$U = \frac{1}{\hbar\omega_{\rm LO}}\frac{e^2}{\epsilon_\infty}\sqrt{\frac{m\omega_{\rm LO}}{\hbar}},$$
$$\alpha = \frac{1}{\hbar\omega_{\rm LO}}\frac{e^2}{\sqrt{2}}\left(\frac{1}{\epsilon_\infty}-\frac{1}{\epsilon_0}\right)\sqrt{\frac{m\omega_{\rm LO}}{\hbar}}, \tag{2.3}$$

and depend on the static (ϵ_0) and high-frequency (ϵ_∞) dielectric constants. Introducing the ratio of the dielectric constants $\eta = \epsilon_\infty/\epsilon_0$ we obtain the following relation between the Coulomb and electron-phonon coupling constants:

$$U = \frac{\sqrt{2}\,\alpha}{1-\eta}, \tag{2.4}$$

from which it follows that only $U \geq \sqrt{2}\,\alpha$ are physically reliable.

The above problem was tackled by Verbist, Peeters, and Devreese in Ref. 1 (hereafter called I) using a path-integral representation for the partition function and applying Feynman's variational method to approximate the path integral. The trial action was chosen corresponding to a model where two fictitious particles of mass M are connected with each of the electrons by strings of strengths κ and κ' and another string of strength K connects the two electrons to simulate the direct Coulomb interaction. In addition, a vector $\vec{a}$ is used to describe fluctuations of electrons around a mean vector distance $\vec{a}$ from each other. Note that a related trial action was used in Ref. 14 to study a system without repulsion.

The Hamiltonian describing this trial system is

$$H_{\rm tr} = \sum_{j=1,2}\left[\frac{\vec{p}_j^{\,2}}{2m}+\frac{\vec{P}_j^{\,2}}{2M}+\frac{\kappa}{2}(\vec{r}_j-\vec{R}_j)^2\right] +\frac{\kappa'}{2}\left[(\vec{r}_1-\vec{R}_2-\vec{a})^2+(\vec{r}_2-\vec{R}_1+\vec{a})^2\right] -\frac{K}{2}(\vec{r}_1-\vec{r}_2-\vec{a})^2. \tag{2.5}$$

All mentioned oscillator strengths, the mass M, and the mean separation $\vec{a}$ are considered as variational parameters. It was found in I that in the bipolaron state the vector parameter $\vec{a}$ equals zero. The squares of the eigenfrequencies of the model system are

$$\Omega_1^2 = \frac{M+m}{Mm}(\kappa+\kappa'), \tag{2.6a}$$

$$\Omega_{2,3}^2 = \frac{M+m}{2Mm}(\kappa+\kappa')-\frac{K}{m} \pm\frac{1}{2}\sqrt{\left[\frac{M-m}{Mm}(\kappa+\kappa')-\frac{2K}{m}\right]^2+\frac{4(\kappa-\kappa')^2}{Mm}}, \tag{2.6b}$$

$$\nu^2 = \frac{\kappa+\kappa'}{M}. \tag{2.6c}$$

These variational parameters satisfy the following inequalities:

$$\Omega_1^2 \geq \Omega_2^2+\Omega_3^2, \quad \Omega_2 \geq \nu \geq \Omega_3 \geq 0. \tag{2.7}$$

In what follows we use dimensionless units $\hbar = m = \omega_{\rm LO} = 1$, so that the bipolaron energy is given in units of $\hbar\omega_{\rm LO}$ and the length in units of $\sqrt{\hbar/m\omega_{\rm LO}}$. In I an upper bound was obtained for the free energy. Taking the zero temperature limit of the expression given in I we obtain the upper estimate for the bipolaron energy,

$$E_{\rm bip} = \frac{3}{2}\sum_{j=1}^{3}\Omega_j - 3\nu - \frac{3}{4}\left(\Omega_1\frac{\Omega_1^2-\nu^2}{\Omega_1^2}+\Omega_2\frac{\Omega_2^2-\nu^2}{\Omega_2^2-\Omega_3^2} +\Omega_3\frac{\nu^2-\Omega_3^2}{\Omega_2^2-\Omega_3^2}\right) - \alpha\sqrt{\frac{2}{\pi}}\int_0^\infty dx \times e^{-x}\left(\frac{1}{\sqrt{D_{11}(x)}}+\frac{1}{\sqrt{D_{12}(x)}}\right)+\frac{U}{\sqrt{\pi D_{12}(0)}}, \tag{2.8}$$

where

$$D_{11(12)}(x) = \frac{\nu^2}{\Omega_1^2}\frac{x}{4}+\frac{\Omega_1^2-\nu^2}{\Omega_1^2}\frac{1-e^{-\Omega_1 x}}{4\Omega_1} +\frac{\Omega_2^2-\nu^2}{\Omega_2^2-\Omega_3^2}\frac{1\mp e^{-\Omega_2 x}}{4\Omega_2} +\frac{\nu^2-\Omega_3^2}{\Omega_2^2-\Omega_3^2}\frac{1\mp e^{-\Omega_3 x}}{4\Omega_3}. \tag{2.9}$$

It follows from Eq. (2.9) that

$$D_{12}(0) = \frac{\nu^2+\Omega_2\Omega_3}{2\Omega_2\Omega_3(\Omega_2+\Omega_3)}. \tag{2.10}$$

Equations (2.8)–(2.10) generalize Feynman results[15] to the case of two polarons.

III. STRONG-COUPLING EXPANSION

The physical condition (2.4) implies that the repulsion cannot be arbitrary small. In the classical limit this implies that a bipolaron state cannot exist. Indeed, at large distances the effective potential due to phonon exchange reduces the effective attractive potential to $-\alpha\sqrt{2}/r$ (in dimensionless units). Adding the direct Coulomb repulsion between the two electrons it gives us the net *repulsion* potential $e^2/\epsilon_0 r$. It is because of phonon exchange and correlation that a bipolaron state can exist quantum mechanically. Bipolaron formation becomes possible at rather large values of the electron-phonon coupling constant α which provides us with a motivation to apply the strong-coupling expansion in inverse powers of α.

When $\alpha \to \infty$ the frequencies Ω_1, Ω_2 become large while Ω_3 and ν remain finite. Thus, we search for a solution, in which we expand the variational parameters in a series of inverse powers of α^2,

$$\Omega_j = \alpha^2\omega_j + \sum_{n\geq 0} \frac{\omega_j^{(n)}}{\alpha^{2n}}, \quad j = 1,2, \tag{3.1a}$$

$$\Omega_3 = \omega_3 + \sum_{n\geq 1} \frac{\omega_3^{(n)}}{\alpha^{2n}}, \tag{3.1b}$$

$$\nu = w + \sum_{n\geq 1} \frac{w^{(n)}}{\alpha^{2n}}. \tag{3.1c}$$

To find a regular procedure of the strong-coupling expansion of the integral in Eq. (2.8) we define a function which differs from those of Eq. (2.9) by terms decreasing exponentially in x,

$$D_{\rm as}(x) = \frac{\nu^2}{\Omega_1^2}\frac{x}{4} + \frac{\Omega_1^2-\nu^2}{\Omega_1^2}\frac{1}{4\Omega_1} + \frac{\Omega_2^2-\nu^2}{\Omega_2^2-\Omega_3^2}\frac{1}{4\Omega_2} + \frac{\nu^2-\Omega_3^2}{\Omega_2^2-\Omega_3^2}\frac{1}{4\Omega_3}. \tag{3.2}$$

Adding and subtracting $2/\sqrt{D_{\rm as}(x)}$ into the integral of Eq. (2.8) we obtain

$$\begin{aligned} E_{\rm bip} = {} & \frac{3}{2}\sum_{j=1}^{3}\Omega_j - 3\nu - \frac{3}{4}\left(\Omega_1\frac{\Omega_1^2-\nu^2}{\Omega_1^2} + \Omega_2\frac{\Omega_2^2-\nu^2}{\Omega_2^2-\Omega_3^2} + \Omega_3\frac{\nu^2-\Omega_3^2}{\Omega_2^2-\Omega_3^2}\right) \\ & -\alpha^2\, 2\sqrt{\frac{2}{\pi}}\int_0^\infty dx\,\frac{e^{-x}}{\sqrt{\alpha^2 D_{\rm as}(x)}} + \alpha^2\left(\frac{U}{\alpha}\right)\frac{1}{\sqrt{\pi\alpha^2 D_{12}(0)}} \\ & -\sqrt{\frac{2}{\pi}}\int_0^\infty dx\, e^{-x/\alpha^2}\left[\frac{1}{\sqrt{\alpha^2 D_{11}(x/\alpha^2)}} + \frac{1}{\sqrt{\alpha^2 D_{12}(x/\alpha^2)}} - \frac{2}{\sqrt{\alpha^2 D_{\rm as}(x/\alpha^2)}}\right], \end{aligned} \tag{3.3}$$

where the scaling $x \to x/\alpha^2$ is performed in the last line of Eq. (3.3). The idea of representing $E_{\rm bip}$ in the form (3.3) is the same one used in Ref. 6 to derive the strong-coupling expansion for a single polaron.

Inserting expansions (3.1) into Eq. (3.3) and collecting terms of the same order in α we arrive at the strong-coupling expansion for the bipolaron energy,

$$E_{\rm bip} = -\frac{2}{3\pi}\alpha^2 A(u) - B(u) + O(\alpha^{-2}), \quad u = \frac{U}{\alpha}, \tag{3.4}$$

which has to be compared to the strong-coupling expansion of twice the single polaron energy,

$$2E_{\rm pol} = -\frac{2}{3\pi}\alpha^2 - \left(\frac{3}{2} + 6\ln 2\right) + O(\alpha^{-2}). \tag{3.5}$$

Because the critical value of the coupling constant for the bipolaron stability is about $\alpha_c \approx 7$ (see I), terms of order $O(1/\alpha^2)$ may be skipped. Moreover, because of the minimization of the leading term of the expansion (3.4) over ω_1, ω_2 constant terms in the expansions (3.1a) will not contribute to the coefficient $B(u)$ in Eq. (3.4). Thus, in order to derive $E_{\rm bip}$ to first two orders, we insert the parameters $\Omega_j = \alpha^2\omega_j$ $(j = 1,2)$, $\Omega_3 = \omega_3$, $\nu = w$ into Eq. (3.3), and take the limit of large α which results into

$$\alpha^2 D_{11(12)}(x/\alpha^2) \to \frac{1-e^{-\omega_1 x}}{4\omega_1} + \frac{1\mp e^{-\omega_2 x}}{4\omega_2}, \tag{3.6a}$$

$$\alpha^2 D_{\rm as}(x/\alpha^2) \to \frac{\omega_1+\omega_2}{4\omega_1\omega_2}, \tag{3.6b}$$

and consequently the term in the last line of Eq. (3.3) contributes only to the coefficient $B(u)$. The following expansions are also valid:

$$\alpha^2 D_{12}(0) = \frac{1}{2\omega_2}\left(1 + \frac{1}{\alpha^2}\frac{w^2-\omega_3^2}{\omega_2\omega_3} + \cdots\right), \tag{3.7a}$$

$$\alpha^2 D_{\rm as}(x) = \frac{\omega_1+\omega_2}{4\omega_1\omega_2}\left[1 + \frac{1}{\alpha^2}\frac{\omega_1\omega_2}{\omega_1+\omega_2}\left(\frac{w^2-\omega_3^2}{\omega_2^2\omega_3} + x\frac{w^2}{\omega_1^2}\right) + \cdots\right]. \tag{3.7b}$$

IV. COEFFICIENTS OF THE STRONG-COUPLING EXPANSION

Using the previous expansions we arrive at the expression for the coefficient $A(u)$,

$$A(u) = \left(-\frac{3\pi}{2}\right)\left[\frac{3}{4}(\omega_1+\omega_2) - 4\sqrt{\frac{2}{\pi}}\sqrt{\frac{\omega_1\omega_2}{\omega_1+\omega_2}} + u\sqrt{\frac{2}{\pi}}\sqrt{\omega_2}\right]. \tag{4.1}$$

The expression in square brackets has to be minimized over $\omega_1 \geq \omega_2$. When $u > 4$ the minimum is reached at the end point $\omega_2 \to 0$ and the corresponding value $E_{\rm bip} = 0$. When $u \leq 4$ the differentiation of $A(u)$ over frequencies leads to the pair of equations

$$\frac{3}{4} - 2\sqrt{\frac{2}{\pi}}\frac{1}{\sqrt{\omega_1}}\left(\frac{\omega_2}{\omega_1+\omega_2}\right)^{3/2} = 0, \tag{4.2a}$$

$$\frac{3}{4} - 2\sqrt{\frac{2}{\pi}}\frac{1}{\sqrt{\omega_2}}\left(\frac{\omega_1}{\omega_1+\omega_2}\right)^{3/2} + \frac{1}{2}\sqrt{\frac{2}{\pi}}\frac{u}{\sqrt{\omega_2}} = 0. \tag{4.2b}$$

Introducing the notation

$$\zeta = \sqrt{\frac{\omega_1}{\omega_1+\omega_2}}, \tag{4.3}$$

we obtain from Eq. (4.2a) expressions for the frequencies:

$$\omega_1 = \frac{128}{9\pi}(1-\zeta^2)^3, \tag{4.4a}$$

$$\omega_2 = \frac{128}{9\pi}\frac{(1-\zeta^2)^4}{\zeta^2}. \tag{4.4b}$$

The dependence of ω_1, ω_2 on the scaled repulsion u is shown in Fig. 1. Both ω_1 and ω_2 approach zero at the limiting point $u = 4$ and furthermore $\omega_2/\omega_1 \to 0$ at this point.

Equation (4.2b) then becomes a quadratic equation for the function $\zeta(u)$ with the solution

$$\zeta(u) = \frac{u}{16} + \frac{1}{2}\sqrt{2 + \frac{u^2}{64}}. \tag{4.5}$$

Inserting known frequencies into Eq. (4.1) we find the leading term of the strong-coupling expansion,

$$A(u) = 4 - 2\sqrt{2}u\left(1 + \frac{u^2}{128}\right)^{3/2} + \frac{5}{8}u^2 - \frac{u^4}{512}. \tag{4.6}$$

The dependence $A(u)$ is shown in Fig. 2. As was mentioned above, the curve (4.6) is replaced by $A(u) \equiv 0$ when $u > 4$. A bipolaron stability region is defined in this approximation for $u = U/\alpha$ such that $A(u) \geq 1$. This occurs when the repulsion does not exceed a maximal value: $U/\alpha \leq u_{\rm max}$, which from Eq. (4.6) is given by

$$u_{\rm max} = \left[\frac{1}{3}\left(49 + \sqrt[3]{5232\sqrt{327} - 91\,151} - \sqrt[3]{5232\sqrt{327} + 91\,151}\right)\right]^{1/2} = 1.534\,770\,321. \tag{4.7}$$

Equivalently, we find the critical value for the ratio of dielectric constants $\eta_c = 0.078\,550\,358$ [$u_{\rm max} = \sqrt{2}/(1-\eta_c)$, cf. Eq. (2.4)]. It is not really a surprise that we obtained the same numerical value (0.079) while averaging the Hamiltonian over a product of Gaussian wave functions, one for the center of mass and the other for the relative coordinates.[16] The maximal value $u_{\rm max}$ is shown in Fig. 2 by the dashed vertical line while the solid vertical line represents the minimal physical value $u_{\rm min} = \sqrt{2}$. The bipolaron stability region in this approximation corresponds to the space between these two lines.

In the absence of the Coulomb repulsion the bipolaron energy is eight times larger than the single polaron energy in the strong-coupling limit: $A(0) = 4$ [cf. Eq. (3.4)]. The physical meaning of this behavior of the coefficient

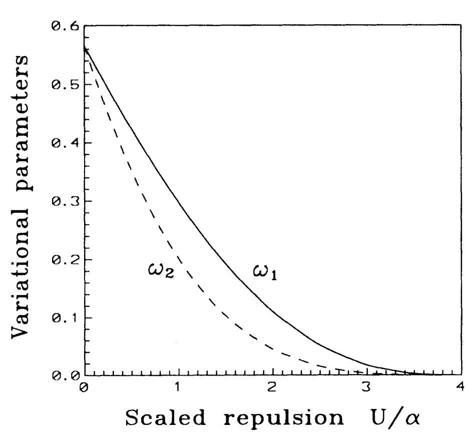

FIG. 1. Two variational frequencies ω_1, ω_2 contributing to the leading order of the strong-coupling expansion vs the scaled repulsion $u = U/\alpha$.

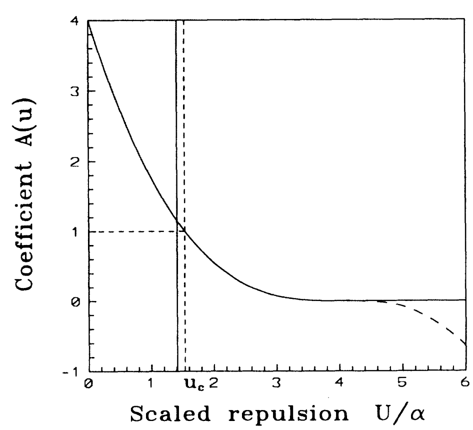

FIG. 2. The dependence of the coefficient $A(u)$ in the leading term of the strong-coupling expansion for the bipolaron energy on the Coulomb repulsion coupling constant $u = U/\alpha$. The maximal value $u_{\rm max} = 1.534\,77$ and the minimal value $u_{\rm min} = \sqrt{2}$ are shown by dashed and solid vertical lines, respectively. The space between these lines is the bipolaron stability region.

$A(u)$ follows from the definition (2.3) of the Fröhlich coupling constant α. In the strong-coupling limit when the Coulomb repulsion is switched off the bipolaron state is equivalent to a single polaron created by a particle with the doubled electric charge $2e$ and the reduced mass $m/2$. Then it follows from Eq. (2.3) that for this "effective polaron" the coupling constant α obtains a factor of $2\sqrt{2}$ in comparison with a "normal polaron." Because the energy of the latter behaves like α^2 in the strong-coupling limit, the bipolaron energy is indeed eight times larger than the single polaron energy. This conclusion does not depend on approximations being made. It follows also from lower and upper estimates for the bipolaron energy which coincide in the strong-coupling limit in the absence of the Coulomb repulsion as was shown in Ref. 17.

To proceed further we collect now terms of order α^0. As was mentioned, the term in the last line of Eq. (3.3) depends only on the variational parameters ω_1 and ω_2 which are determined now. The contribution of the first two lines of Eq. (3.3) to the coefficient $B(u)$ is

$$3w - \frac{3}{4}\,\omega_3 - \frac{3}{4}\,w^2\,\frac{1+\omega_3}{\omega_3}, \tag{4.8}$$

which reaches the maximal value $\frac{3}{4}$ at $w = \omega_3 = 1$. In order to obtain the coefficient $B(u)$ we have to find the limit of the integral in the last line of Eq. (3.3). Then, introducing a new variable $z = \exp(-\omega_2 x)$ in the integral and performing partially the integration we arrive at the representation

$$\begin{aligned} B(u) = {} & \frac{3}{4} + \frac{3}{2}(\xi+1)\ln\frac{4\sqrt{\xi+1}}{\sqrt{\xi+1}+\sqrt{2\xi}} \\ & + \frac{3}{4}\xi(\xi^2-1)\int_0^1 dz\left\{\frac{1}{s_-[\sqrt{\xi+1}+s_-]}\right. \\ & \left. - \frac{1}{s_+[\sqrt{\xi+1}+s_+]}\right\} \end{aligned} \tag{4.9}$$

with $s_\mp = \sqrt{1 - z^\xi + \xi(1 \mp z)}$, where $\xi = \omega_1/\omega_2 = \zeta^2/(1-\zeta^2) = 64/[(\sqrt{128+u^2}-u)^2 - 64]$, as it follows from Eq. (4.4). Note that the function $\zeta(u)$ is given by Eq. (4.5). The dependence of the coefficient $B(u)$ on the scaled repulsion $u = U/\alpha$ is shown in Fig. 3.

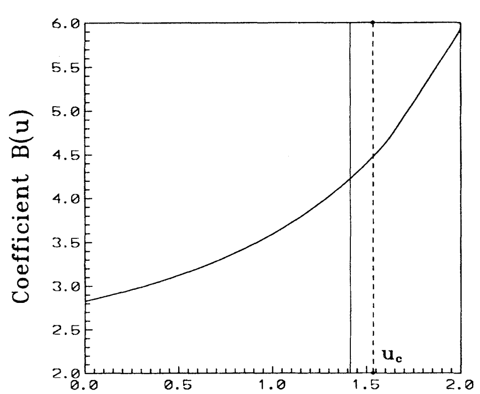

FIG. 3. The coefficient $B(u)$ in the first correction term of the strong-coupling expansion for the bipolaron energy vs the Coulomb repulsion coupling constant $u = U/\alpha$. The maximum value $u_{\max} = 1.534\,77$ and the minimum value $u_{\min} = \sqrt{2}$ are shown by dashed and solid vertical lines, respectively.

V. STABILITY REGIONS

A. The bipolaron model of I

To find the critical value of α at which bipolaron formation occurs, one has to compare the bipolaron energy with twice the polaron energy as given by Eqs. (3.4) and (3.5), respectively. This leads us to the relation

$$\alpha = \frac{1}{2}\sqrt{3\pi\frac{3 + 12\ln 2 - 2\,B(u)}{A(u) - 1}}. \tag{5.1}$$

From this equation we can find numerically the function $U_c(\alpha)$, at which point bipolaron formation is possible at a given value of α. This function is presented in Fig. 4. The plotted curve (solid line) exceeds the line $U = \sqrt{2}\alpha$ (short dashes) at $\alpha \geq \alpha_c = 6.753$. The value obtained is in agreement with the direct minimization procedure done in I where the value $\alpha_c = 6.8$ was reported. Besides, we obtained in the vicinity of the critical point ($u \approx \sqrt{2}$) $U_c \approx 1.68\alpha - 1.82$. The result $U_c \approx 1.63\alpha - 1.49$ was obtained in I (a misprint occurred there in which the second term had a positive sign).

To present the equation for the boundary of the bipolaron formation region in a more simple form let us

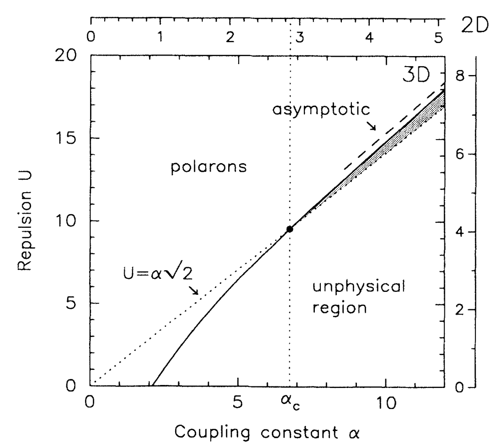

FIG. 4. The boundary of the bipolaron stability region. The solid curve corresponds to $U_c(\alpha)$, the dotted line $U = \sqrt{2}\alpha$ shows the boundary with the physical region, and the dashed line shows the asymptotic $U_c \approx 1.534\,77\alpha$ for large α. The shadowed sector between the solid and the dashed lines is the region where bipolarons exist. Different scales are shown to present results in 3D and 2D. The critical values are $\alpha_c = 6.75$ in 3D and 2.866 in 2D.

note that the physical region corresponds to an interval between $u_{\min} = \sqrt{2}$ and $u_{\max} = 1.534\,77$. This interval is narrow enough to use linear approximations for the functions $A(u), B(u)$. Our numerical calculations give $A(\sqrt{2}) = 1.148\,08$, $B(\sqrt{2}) = 4.226\,13$ and $A(u_{\max}) = 1$, $B(u_{\max}) = 4.476\,65$, from which we find the following linear fits to these coefficients: $A(u) \approx -1.2282u + 2.8851$ and $B(u) \approx 2.0771u + 1.2886$. If we insert these equations into Eq. (3.4) we find an expression for the bipolaron energy which is adequate in the physical region of coupling constants. Comparing with twice the polaron energy of Eq. (3.5) we arrive at the simple formula for the boundary of the bipolaron formation region,

$$U_c(\alpha) = 1.534\,77\,\alpha\,\frac{\alpha^2 - 10.925}{\alpha^2 - 7.969}. \quad (5.2)$$

We remind the reader once more that this expression can be used for $\alpha \geq \alpha_c = 6.753$, in which region it can be considered as almost exact (in the scope of the model under consideration).

From Eq. (5.2) we notice that the asymptotic behavior $U_c \approx 1.534\,77\alpha$ is improved by terms of order $1/\alpha$. Only the first two terms of this expansion are exact within this model,

$$U_c(\alpha) \approx 1.534\,77\alpha - \frac{4.536}{\alpha}. \quad (5.3)$$

At large α this coincides with Eq. (5.2). Near the critical point discrepancies occur, e.g., the critical value following from Eq. (5.3) is $\alpha_c = 6.13$ which is 9% smaller than the value obtained above. This can be improved by artificially changing the coefficient of the $1/\alpha$ term,

$$U_c(\alpha) \approx 1.534\,77\alpha - \frac{5.497}{\alpha}. \quad (5.4)$$

By construction this expression gives the same critical value $\alpha_c = 6.753$ and the boundary curve behaves as $U_c \approx 1.66\alpha - 1.62$ in the vicinity of the critical point. The slope parameter differs by less than 2%. For large α the discrepancy between Eqs. (5.3) and (5.4) equals approximately $0.96/\alpha$ and consequently the relative discrepancy is about $0.63/\alpha^2 \leq 0.63/\alpha_c^2 = 0.014$. Thus, one may use the formula (5.4) to approximate the boundary curve within the bipolaron model of I in the physical region $\alpha \geq \alpha_c$ with the accuracy of about 1–2 %.

B. Fitted results in 3D

From the above we find a possibility to go beyond the bipolaron model of I and to exploit results of other authors. Rewrite Eq. (5.4) as follows:

$$U_c(\alpha) \approx \frac{\sqrt{2}}{1-\eta_c}\left(\alpha - \eta_c\frac{\alpha_c^2}{\alpha}\right). \quad (5.5)$$

Results for the critical value of the dielectric constants were obtained by Mukhomorov[18] ($\eta_c = 0.107$), Suprun and Moizhes[19] and Vinetskii *et al.*[20] ($\eta_c = 0.138$), Adamowski[21] ($\eta_c = 0.14$), Sil *et al.*[22] ($\eta_c = 0.129$), and by our group[16] ($\eta_c = 0.131$). Thus, for reference we accept in what follows the value $\eta_c = 0.138$ which corresponds to $u_{\max} = 1.641$. As to the critical value of α, apart of the quoted result of I also the results $\alpha_c = 7.3$ of Ref. 21 and $\alpha_c = 6$ of Bassani *et al.*[23] are reported. Thus, one can take quite reliably the same value $\alpha_c = 6.753$ as was used before. This leads us to the approximate formula analogous to Eq. (5.4),

$$U_c(\alpha) \approx 1.641\alpha - \frac{10.342}{\alpha}. \quad (5.6)$$

The corresponding curve is plotted in Fig. 5 (solid line) together with the boundary obtained from Eqs. (5.1) and (5.2) (dashed line).

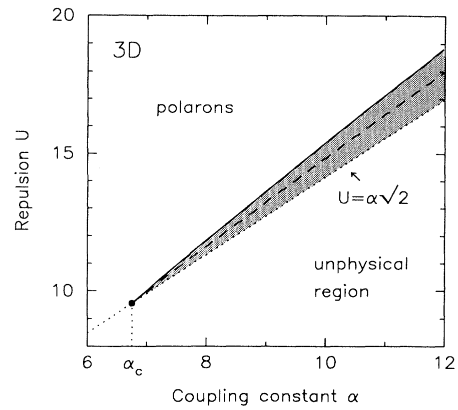

FIG. 5. The three-dimensional bipolaron stability regions calculated in the bipolaron model of I (dashed curve) and as the result of the fit (solid curve). The frontier of the physical region is also shown by the dotted line. Now the shadowed region of the bipolaron stability is much larger than the sector between the dashed and the dotted lines which was already presented in Fig. 4.

C. Results in 2D

To apply the present results to lower dimensions we make use of the fact that Feynman-type approximations satisfy a scaling law introduced by two of the present authors and Wu.[4,24,25] This law establishes links between polaron characteristics in spaces of different dimensions. In particular, (bi)polaron energy in 2D can be obtained as

$$E^{(2D)}(\alpha, u) = \frac{2}{3}E^{(3D)}\left(\frac{3\pi}{4}\alpha, u\right). \quad (5.7)$$

The equation for the critical values of α in two dimensions is readily obtained from Eq. (5.1),

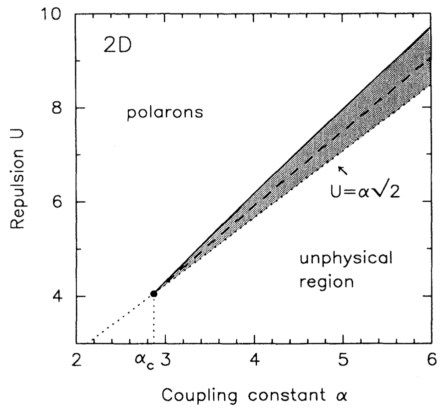

FIG. 6. The stability region for the two-dimensional bipolarons (notations are the same as in Fig. 5).

$$\alpha = \frac{2}{3\pi}\sqrt{3\pi\frac{3+12\ln 2-2\,B(u)}{A(u)-1}} \tag{5.8}$$

with the same functions $A(u)$ and $B(u)$. The boundary of the bipolaron formation region for the 2D case is presented in Fig. 4 with the scales given at the top and right side of the figure. The critical value of the electron-phonon coupling constant is now given by $\alpha_c^{(2D)} = 2.866$. In the vicinity of this point $U_c \approx 1.683\alpha - 0.77$ (cf. the result $U_c \approx 1.63\alpha - 0.63$ of I, where the same misprint in the sign occurred as in the 3D case). Note that the slope of the curves in 3D and 2D coincides because of the scaling law.

The same scaling law can be applied to arrive at formulas analogous to Eqs. (5.2) and (5.3). But the scaling is not valid outside the scope of the bipolaron model of I (or any other model based on a path-integral approximation with a quadratic trial action). Consequently, to derive an approximate formula in 2D of the type of Eq. (5.6) one can use again the known value for $\eta_c = 0.158$ ($u_{\max} = 1.680$) of Ref. 16 and the quoted value $\alpha_c = 2.866$ (2.9 in I and 2 in Ref. 23). The result obtained is then as follows:

$$U_c(\alpha) \approx 1.680\alpha - \frac{2.183}{\alpha}. \tag{5.9}$$

The corresponding curve is plotted in Fig. 6 (solid line) together with the curve of Eq. (5.8) (dashed line).

A detailed study of a bipolaron confined to a one-dimensional structure is given in Ref. 26. An analogous strong-coupling analysis for 1D bipolarons can be found in Ref. 27.

VI. EFFECTIVE MASS AND EXTENSION OF THE BIPOLARON

Another important bipolaron characteristic is its effective mass $m^*_{\rm bip}$, which influences the bipolaron mobility. It was defined in I as the total mass of the two electrons and the fictitious particles: $m^*_{\rm bip} = 2(m+M)$. In terms of oscillator frequencies (2.6) it can be written as

$$m^*_{\rm bip} = 2m\frac{\Omega_1^2}{\nu^2}. \tag{6.1}$$

Inserting here the series (3.1) we arrive at the strong-coupling expansion for the bipolaron effective mass:

$$m^*_{\rm bip} = 2m\alpha^4\left(\frac{\omega_1}{w}\right)^2\left[1+\frac{2}{\alpha^2}\left(\frac{\omega_1^{(0)}}{\omega_1}-\frac{w^{(1)}}{w}\right)+o(\alpha^{-2})\right]. \tag{6.2}$$

The leading term follows from Eqs. (4.4a) and (4.8):

$$m^*_{\rm bip} = 2m\frac{16\alpha^4}{81\pi^2}M(u) + O(\alpha^2),$$

$$M(u) = 16\left(1-\frac{u^2}{64}-\frac{u}{8}\sqrt{2+\frac{u^2}{64}}\right)^6, \tag{6.3}$$

which compares to the strong-coupling expansion of the single polaron effective mass,

$$m^*_{\rm pol} = m\frac{16\alpha^4}{81\pi^2} + O(\alpha^2), \tag{6.4}$$

taken at the same order of approximation. Thus, $m^*_{\rm bip}/2m^*_{\rm pol} = M(u) + O(1/\alpha^2)$. At the critical point $u = u_{\min} = \sqrt{2}$ we obtain $M(u_{\min}) \approx 2.1703$ while at maximal repulsion $u = u_{\max} = 1.534\,77$ we have $M(u_{\max}) \approx 1.7177$. Within the model of I the bipolaron effective mass is approximately twice as large as the total mass of two free polarons. It decreases with repulsion and exhibits a discontinuity when a bipolaron state decays into two polaron states. The same conclusion is expected in 2D. The single polaron effective mass can be obtained by scaling from 3D as was derived in Ref. 28: $m^{*(2D)}_{\rm bip} \approx \alpha^4\pi^2/16$. As shown in I the same scaling $m^{(2D)}(\alpha) \approx m^{(3D)}(3\pi\alpha/4)$ is valid for the bipolaron mass so their ratio will be the same as in 3D.

In the absence of the Coulomb repulsion the bipolaron effective mass is 32 times larger than the single polaron effective mass: $M(0) = 16$. The explanation follows from arguments given in Sec. IV while discussing the same limit for the bipolaron energy where it was found that $\alpha \to 2\sqrt{2}\alpha$ when one goes from the polaron to the relevant bipolaron quantity. Therefore the "normal polaron" effective mass $m^*_{\rm pol} \sim m\alpha^4$ should be scaled to obtain the "effective polaron" (bipolaron) mass in the strong-coupling limit as follows: $m^*_{\rm bip} \sim (m/2)(2\sqrt{2}\alpha)^4 = 32m\alpha^4$.

Because the leading term of the strong-coupling expansion for the bipolaron effective mass varies not so crucially, we may use again a linear fit to simplify the formulas,

$$m_{\rm bip}^{*(3D)} \approx m\alpha^4(0.2994 - 0.1503u),$$
$$m_{\rm bip}^{*(2D)} \approx m\alpha^4(9.2283 - 4.6321u). \quad (6.5)$$

Note that to calculate the first correction to the bipolaron effective mass (6.5) one needs variational parameters $\omega_1^{(0)}$ and w_1 which were not determined while considering the first two terms of the strong-coupling expansion for the bipolaron energy.

Another characteristic of interest is the mean-square separation r_{12} between the electrons which was shown in I to be given by $r_{12} = \sqrt{2dD_{12}(0)}$, where d is the number of spatial dimensions. Using Eq. (2.10) and inserting there the series (3.1) we arrive at the leading term of the strong-coupling expansion for the inverse mean-square separation

$$\frac{1}{r_{12}} = \alpha\sqrt{\frac{\omega_2}{3}} + O(\alpha^{-1}) = \frac{2\alpha}{9}\sqrt{\frac{3}{\pi}}\frac{1}{R(u)} + O(\alpha^{-1}),$$
$$\frac{1}{R(u)} = \left(2 + \frac{u^2}{16}\right)\sqrt{1 + \frac{u^2}{128}} - \frac{u}{8\sqrt{2}}\left(10 - \frac{u^2}{16}\right), \quad (6.6)$$

which compares to the leading term of the strong-coupling expansion of the single polaron radius $1/r_{\rm pol} \approx (2\alpha/9)\sqrt{3/\pi}$. Thus, the ratio of the bipolaron mean-square separation to the polaron radius is given by $r_{12}/r_{\rm pol} = R(u) + O(\alpha^{-2})$. At the critical point $R(u_{\rm min}) \approx 1.102$, while at the maximal repulsion it reaches the value $R(u_{\rm max}) \approx 1.204$. In 2D this ratio remains the same as in 3D.

VII. CONCLUSIONS

We analyzed the strong-coupling limit for the bipolaron model introduced by Verbist, Peeters, and Devreese in paper I, which is based on Feynman path integrals. Leading terms of the strong-coupling asymptotic expansions are calculated for the bipolaron ground-state energy, effective mass, and mean-square separation. We presented also simple fitting formulas approximating the boundary between the bipolaron and the polaron regions.

ACKNOWLEDGMENTS

We are indebted to E. L. Bratkovskaya and P. Vansant for assistance in numerical calculations and to G. Verbist for discussions. M.A.S. is grateful to the University of Antwerp (UIA) for the support and kind hospitality during his visit to Belgium. One of us (F.M.P.) is supported by the Belgian National Science Foundation. This work is partly performed in the framework of "Diensten voor de Programmatie van het Wetenschapsbeleid" (Belgium) under Contract No. IT/SC/24, the FKFO Project No. 2.0093.91, and the Human Capital and Mobility Program C.E.C. No. CHRX-CT93-0124.

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* Permanent address: Bogoliubov Theoretical Laboratory, Joint Institute for Nuclear Research, 141980 Dubna, Russia; electronic address: smond@thsun1.jinr.dubna.su

† Also at Universiteit Antwerpen (RUCA), Groenenborgerlaan 171, 2020 Antwerpen, Belgium and Technische Universiteit Eindhoven, P.O. Box 513, 5600 MB Eindhoven, The Netherlands; electronic address: devreese@nats.uia.ac.be

‡ Electronic address: peeters@nats.uia.ac.be

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