Semiconductor Physics, Quantum Electronics & Optoelectronics, 2017. V. 20, N 3. P. 319-324.
doi: https://doi.org/10.15407/spqeo20.03.319

PACS 71.38, 71.38.-k, 74.20.-z, 74.72-h

Gross–Tulub polaron functional in the region of intermediate and strong coupling

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Abstract. Properties of the polaron functional obtained as a result of averaging the Fröhlich Hamiltonian on the translation-invariant function have been investigated. The polaron functional can be represented in two different forms. It has been shown that the functional of translationally invariant Gross–Tulub polaron cannot be applied in the strong coupling region, where the real part $\mathrm{Re}\,D(s)$ of the complex quantity $D\left(s^2\right)$ takes negative values. The function $D\left(s^2\right)$ coincides in its structure with the dynamic susceptibility of degenerate electron gas. The necessary condition for obtaining correct results is investigation of the region of admissible values of the Gross–Tulub functional depending on properties of the function $D\left(s^2\right)$, variational parameters, and the electron-phonon interaction parameter α (Fröhlich coupling constant). A simple and exact formula for the recoil energy of the translationally invariant polaron has been derived, which makes it possible to extend the range of admissible values of the parameters of the electron-phonon interaction to the region of extremely strong coupling ($\alpha > 10$), where $\mathrm{Re}\,D(s) \le 0$. Numerical investigation of different forms of polaron functionals obtained using the field theory methods has been carried out.

Keywords: polaron, Fröhlich Hamiltonian, electron-phonon interaction.

Manuscript received 05.07.17; revised version received 09.08.17; accepted for publication 06.09.17; published online 09.10.17.

The study of properties of translationally invariant polaron was initiated in the classical work by Lee, Low, and Pines [1]. Consistent application of two canonical transformations to the electron Hamiltonian in the phonon field made it possible to describe the polaron moving in a phonon field for intermediate values of the electron-phonon interaction constant.

The initial Hamiltonian has the form:

$$H = H_0 + H_{e-ph}, \qquad (1)$$

where H_0 includes electron kinetic energy operator and phonon field operator, and H_{e-ph} – Hamiltonian of the electron-phonon interaction.

The first canonical transformation proposed in [1] excluded the electron coordinate from the effective Hamiltonian, the second transformation described the displacement of phonon operators from the equilibrium position under action of electron-phonon interaction. Because the total momentum of the system $\mathbf{P}_{op}$ consisting of electron and phonons interacting with it $\mathbf{P}_{op} = \sum_k \hbar\mathbf{k} a_k^+ a_k + \mathbf{p}$ (where $\mathbf{k}$ is the phonon wave vector, a_k^+, a_k - phonon creation and annihilation operators, $\mathbf{p} = -i\hbar\nabla$ – electron momentum, m^* – effective electron mass band) commutes with the Hamiltonian (1), then it is possible to go over to representation, in which $\mathbf{P}_{op}$ is the "c-number" $\mathbf{P}$, and the

wave function (WF) of the original Hamiltonian will not depend on the electronic coordinates.

The first canonical transformation changes the WF Φ of the Hamiltonian (1) according to the rule: 1) $\Phi(r,Q) = S\Psi(Q) = \exp\left[i/\hbar\left(\mathbf{P} - \sum_k a_k^+ a_k \mathbf{k}\right)\mathbf{r}\right] \Psi(Q)$. The second is represented in the form: 2) $\Psi(Q) = U\Psi_0 = \exp\left[\sum_k \left(a_k^+ f_k - a_k f_k^+\right)\right] \Psi_0$, where f_k is the variational function. If the second unitary transformation is applied directly to the initial Hamiltonian (1), then after varying (1) on f_k and averaging over the phonon and electron variables, we obtain the Pecar strong coupling polaron functional [2]. For a multiplicative polaron wave function that allows separation of phonon and electronic coordinates, the variational function f_k is the Fourier component of the electron part in the polaron wave function. In this case, the second transformation is also called the strong-coupling transformation.

In the papers of Gross and Tulub [3-5], three canonical transformations are successively applied to the Hamiltonian (1). The first two are the transformations of Lee, Low, and Pines, the third transformation leads to the diagonal form expression:

$$H = \sum_k \omega_k a_k^+ a_k + \frac{1}{2m^*}\sum_k \mathbf{k}\mathbf{k}' f_k f_{k'}\left(a_k^+ + a_k\right)\left(a_{k'}^+ + a_{k'}\right). \tag{2}$$

Here and henceforth, following [3-5], it is assumed that $\mathbf{P} = 0$, $\mathbf{P}$ is the eigenvalue of the operator $\mathbf{P}_{op}$; $\omega_k = \omega_k^0 + \frac{k^2}{2m}$, ω_k^0 - the energy of longitudinal optical phonons.

According to [3-6], after averaging over phonon variables, for $\mathbf{P} = 0$ the polaron energy can be represented in the form:

$$E_p = \Delta E + 2g\sum_k V_k f_k + \sum_k \omega_k^0 f_k^2, \tag{3}$$

where $V_k = k^{-1}\hbar\omega_k^0\sqrt{4\pi\alpha l_0}$,

$\alpha = g^2 = \frac{1}{2}\left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0}\right)\frac{e^2}{\hbar\omega_k^0 l_0}$, $l_0 = \sqrt{\frac{\hbar}{2m\omega_k^0}}$.

The first term in (3) describing the shift of frequencies of the zero-point oscillations in the system under action of the electron-phonon interaction has the form [3, 4]:

$$\Delta E = -\frac{3}{8\pi i}\oint_C \frac{ds}{\sqrt{s}} \ln D(s). \tag{4}$$

The integration contour is shown in Fig. 1.

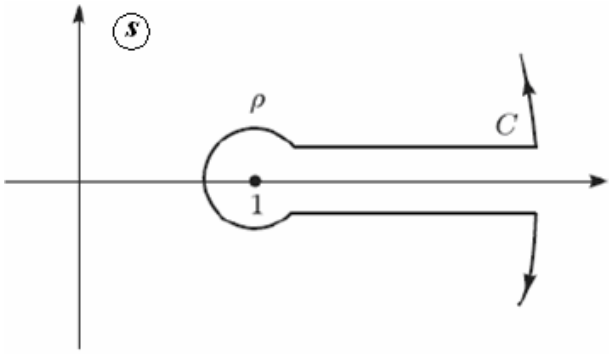

Fig. 1. The contour of integration in Eq. (4).

The function $D(s)$ coincides in its structure with the dynamic susceptibility of degenerate electron gas:

$$D(s) = 1 + Q(s) = 1 + \frac{1}{3\pi^2}\int_0^\infty \frac{p^4 f_p^2 \omega_p}{\omega_p^2 - s} dp. \tag{5}$$

In the complex plane,

$$\operatorname{Im} D\left(\omega_k^2\right) = \frac{k^3 f_k^2}{6\pi}, \tag{6}$$

$$D(s) = \operatorname{Re} D(s) + i \operatorname{Im} D(s) = \left|D(s)\right| \exp\left[i\varphi(s)\right], \tag{7}$$

$$\left|D(s)\right| = \sqrt{\operatorname{Re} D(s)^2 + \operatorname{Im} D(s)^2}$$

for

$$\operatorname{Re} D(s) \geq 0,$$

$$\varphi(s) = \arg D(s) = arctg\left(\operatorname{Im} D(s)/\operatorname{Re} D(s)\right) = \\ = \arcsin\left(\operatorname{Im} D(s)/\left|D(s)\right|\right). \tag{8}$$

After substituting (6) and (7) into (4), one can obtain the expression for the recoil energy written in the Gross–Tulub form [3, 4]:

$$\Delta E = \frac{3}{4\pi}\int_1^\infty \frac{ds}{\sqrt{s}} \operatorname{arctg}\left[\operatorname{Im} D(s)/\operatorname{Re} D(s)\right] = \\ = \frac{3}{2\pi}\int_0^\infty dk \cdot k \cdot \operatorname{arctg}\left[\operatorname{Im} D\left(\omega_k^2\right)/\operatorname{Re} D\left(\omega_k^2\right)\right]. \tag{9}$$

For numerical calculations, it is more convenient to express the argument of the function $D(s)$ through $\arcsin\left\{\operatorname{Im} D(s)/\left|D(s)\right|\right\}$ in relation with the fact that $\left|D(s)\right|$ does not vanish, when $\operatorname{Re} D(s) = 0$. We rewrite the expression (9) in its equivalent form:

$$\Delta E = \frac{3}{4\pi}\int_1^\infty \frac{ds}{\sqrt{s}} \arcsin\left\{\mathrm{Im}\, D(s)/\left|D(s)\right|\right\} =$$

$$= \frac{3}{2\pi}\int_0^\infty dk \cdot k \cdot \arcsin\left\{\mathrm{Im}\, D\left(\omega_k^2\right)/\left|D\left(\omega_k^2\right)\right|\right\}. \qquad (9a)$$

According to (8), the expressions (9) and (9a) were written in [3, 4] only for the first quarter of the complex plane, although this was not specified by the authors of the works. Therefore, the use of (9) for variational calculations requires verification of the fact that for parameters obtained using the variational method, for the entire range of argument s changing the inequality $\mathrm{Re}\, D(s) \geq 0$ is true.

In order to extend the allowable range of parameters to the upper half-plane, it is necessary to take into account that in the second quarter of the complex plane, when $\mathrm{Re}\, D(s) < 0$,

$$\varphi(s) = \arg D(s) = \pi + \mathrm{arctg}\left(\mathrm{Im}\, D(s)/\mathrm{Re}\, D(s)\right) =$$
$$= \pi - \arcsin\left(\mathrm{Im}\, D(s)/\left|D(s)\right|\right). \qquad (10)$$

The change from integration on s to integration with respect to the wave vector in (9) was carried out with allowance for the equality $s = \sqrt{1+k^2/2}$ ($\hbar = 1$, $\omega_k^0 = 1$, $m^* = 1$, and also for the convenience of the notation, we assume that the normalization volume of the crystal $V = 1$).

The integrand in the expression (9) must be analytic in the complex plane, with exception of a finite number of removable singularities. It means that the variational function f_k minimizing the expression (3) must be substituted into the expression (5) in order to check properties of $D(s)$ and the integrand in (7). This check is necessary for a self-consistent calculation, because it is necessary to exclude from the consideration the range of parameters in which the integrand loses the properties of the analytic function. When $\mathrm{Re}\, D(s) \to 0$, the integrand in (9) tends to the maximum permissible value because of the fact that $\mathrm{arctg}(x) \to \pi/2$ for $x \to \infty$. This property of the expression (9) in the work by Gross [3] is called as the striving of the integrand to "saturation". The author [3] did not assume the possibility of considering the range of parameters, for which $\mathrm{Re}\, D\left(\omega_k^2\right) \leq 0$. Gross chose $f_k^G = V_k/\left(1+k^2/b^2\right)$ as a trial function. As it has been shown in [3] for the weak and intermediate coupling regions $b = 1$, at the same time, we can assume that in the strong-coupling region the variation parameter b begins to depend on the magnitude of the coupling constant. Because the expression (9) has a saturation property, then it is impossible to go to the strong coupling limit when choosing a trial function in the form f_k^G.

In the work by Tulub [4], for the recoil terms the expression (9) is given without specifying the type of trial function. In [5] for the strong-coupling region, it was proposed to use as the trial function $f_k = -V_k \exp\left(-k^2/2a^2\right)$. In the work of Pekar [2], several test functions for the strong-coupling polaron were proposed. The quantity $f_k = -V_k \exp\left(-k^2/2a^2\right)$ corresponds to the Fourier component of the normalized electronic function chosen as a one-parameter Gaussian. In [5], the functional of the different type is derived, which is convenient for calculations of the strong-coupling polaron energy:

$$\Delta E = \frac{1}{4\pi^2}\int_0^\infty \frac{k^4 f_k^2 dk}{(1+Q(1))} +$$
$$+ \frac{1}{12\pi^4}\int_0^\infty\int_0^\infty \frac{k^4 f_k^2 p^4 f_p^2 \omega_p \left\{\omega_k\omega_p + \omega_k\left(\omega_k+\omega_p\right)+1\right\}}{\left(\omega_k+\omega_p\right)^2\left(\omega_p^2-1\right)\left|D\left(\omega_p^2\right)\right|^2} dpdk. \qquad (11)$$

The expression (11) was derived from other assumptions than those made in the derivation of the formula (9). As will be seen from further numerical calculations, the functional (11) has a more general character. In contrast to (9), the expression (11) allows a transition to the strong coupling region, since it has no "saturation" property, which was mentioned in the work by Gross. It, unlike (9), is not limited to the area for which $\mathrm{Re}\, D(s) > 0$.

The expression (9), in view of its simplicity in comparison with the expression (11), has a more convenient form for carrying out numerical calculations by using various test functions. In real crystals, the value of the Fröhlich constant of electron-phonon interaction α is bounded from the side of large values and can hardly exceed quantities 8 to 10. This circumstance was noted in [5, 6], where it was indicated that the strong-coupling region for polaron practically reduces to zero.

The quantity $D(s)$ obtained in [5] for $f_k = -V_k \exp\left(-k^2/2a^2\right)$ has the form:

$$\mathrm{Re}\, D\left(\omega_k^2\right) = 1 + \lambda v(y), \qquad (13)$$

$$v(y) = 1 - ye^{-y^2}\int_0^y e^{t^2} dt - \xi e^{\xi^2}\int_\xi^\infty e^{-t^2} dt, \qquad (14)$$

$$\lambda = \frac{4g^2 a}{3\sqrt{2\pi}},\ y = \frac{k}{a},\ \xi = \sqrt{y^2 + \frac{4}{a^2}}.$$

The strong-coupling polaron functional given in [5] was obtained by substituting the $D(s)$ defined by the formula (13) in the expression (11). In ref. [5, 6], an approximate expression of the quantity ΔE is given, which is obtained under assumption that the quantities of order $1/a^2$ can be omitted. One can calculate the polaron energy in the intermediate-coupling region after integrating in (11) without any approximations. Let's choose the trial function in a two-parameter form $f_k' = -NV_k \exp\left(-k^2/2a^2\right)$ [7], where N and a are the variational parameters. After calculating the integrals included in the expression (11), we obtain:

$$\Delta E(a,N,\alpha) = \Delta E_0\left(a,N^2\alpha\right) + \Delta E_1\left(a,N^2\alpha\right), \tag{15}$$

$$\Delta E_0 = \frac{3a^2}{16}\frac{1}{1+\frac{1}{\lambda}+\frac{\sqrt{\pi}}{a}\exp\left(\frac{4}{a^2}\right)\left(erf\left(\frac{2}{|a|}\right)-1\right)}, \tag{16}$$

$$\lambda = \lambda\left(a,\alpha N^2\right) = 4\frac{\left(\alpha N^2\right)a}{3\sqrt{2\pi}} \quad \text{для} \quad a,\alpha >> 1\ ,$$

$$\Delta E_0(a,N,\alpha) \approx \frac{3a^2}{16}, \tag{17}$$

$$\Delta E_1\left(a,N,\alpha\right) = \frac{3a^2}{16}q\left(a,N^2\alpha\right), \tag{18}$$

$$q\left(a,N^2\alpha\right) = \int_0^\infty \left(\frac{2\exp\left(-y^2\right)}{\sqrt{\pi}}\cdot\frac{y^2+2/a^2}{y^2+4/a^2}\times\right.$$

$$\left.\times\frac{1-\Omega(y)}{\left(\frac{1}{\lambda\left(a,N^2\alpha\right)}+v(y)\right)^2+\frac{\pi y^2\exp\left(-2y^2\right)}{4}}\right)dy, \tag{19}$$

$$\Omega(y) = 2y^2\left[\left(1+2\xi(y)^2\right)\xi(y)e^{\xi(y)^2}\int_{\xi(y)}^\infty e^{-t^2}dt - \xi(y)^2\right]. \tag{20}$$

In the expression (16), there is a plus sign before the third term in the denominator instead of the minus given in [8], wherein an error was made when typing the formulas.

Expressions (16) and (17) for $\mathrm{Re}\,D(s) > 0$ can be expanded in a series in $1/\lambda << 1$. Leaving the terms corresponding to the first term of the expansion, we obtain the following approximate functionals:

$$J_{ps} \approx \frac{3}{16}a^2\left[1+\tilde{q}\left(\frac{1}{\lambda_1}\right)\right] - \frac{\alpha\, a}{\sqrt{\pi}}\left(2-\frac{1}{\sqrt{2}}\right), \tag{21}$$

where

$$\lambda_1 = 4\frac{\alpha\, a}{3\sqrt{2\pi}},$$

$$\tilde{q}\left(\frac{1}{\lambda_1}\right) = \frac{2}{\sqrt{\pi}}\int_0^\infty \frac{\exp\left(-y^2\right)\left(1-\tilde{\Omega}(y)\right)}{\left(\frac{1}{\lambda_1}+v(y)\right)^2+\pi y^2\exp\left(-2y^2\right)/4}dy,$$

$$\tilde{\Omega}(y) = 2y^2\left[\left(1+2y^2\right)y\exp\left(y^2\right)\int_y^\infty \exp\left(-t^2\right)dt - y^2\right].$$

$$J'_{ps} \approx \frac{3}{16}a^2\left[1+\tilde{q}\left(\frac{1}{\lambda}\right)\right] - \frac{\alpha\, Na}{\sqrt{\pi}}\left(2-\frac{N}{\sqrt{2}}\right). \tag{22}$$

The functional J_{ps} is obtained using the one-parameter function $f_k = -V_k\exp\left(-k^2/2a^2\right)$, the functional J'_{ps} corresponds to the two-parameter function $f_k' = -NV_k\exp\left(-k^2/2a^2\right)$. The polaron energies that correspond to the minima of the functionals J_{ps} and J'_{ps} are denoted by E_{ps} and E'_{ps}. Let's pay attention to the fact that minimization of approximate functionals (21) and (22) that were obtained by expansion of functional (15) in λ^{-1} does not guarantee finding the polaron energy upper bound.

Since the expressions (9) and (15) are obtained from the same expression (4), the numerical results obtained using the formulas (9) and (15) when minimizing the functional (3) must coincide or have the region in which they coincide. After the numerical comparison, it is also necessary to determine the allowable range of parameters for which the identity requirement of the expression (4) with the expressions (9) and (15) is satisfied. Tulub [5, 6] draws attention to the fact that the value of the electron-phonon interaction parameter is bounded from above $\alpha \sim 8\ldots10$, since 1) the requirements for feasibility of continuum approximation are violated, 2) the restriction of the phonon spectrum to the limiting wave number also leads to approximately the same restriction on the electron-phonon interaction constant, and 3) the constraint on the coupling constant follows from the "resonant" scattering of phonons. All three reasons are considered in detail in [5, 6]. The study of the range of variational parameters admissible values for a given α leads to mathematical limitation on the electron-phonon interaction constant. As it shown by the numerical verification carried out by us, the expressions

(9) and (15) are identical only in the region for which $\mathrm{Re}\,D(s) > 0$. When the expression (3) is minimized with the recoil terms (9) for $\alpha > 10.5$, a negative value region $\mathrm{Re}\,D(s)$ appears, which leads to violation of the allowable range of (9), and equivalence of the expressions (9) and (15) is lost. Thus, for the selected type of trial function f_k', the range of admissible values of the variational parameters limits applicability of the formula (9) to the region $\alpha \le 10.5$. Application of the expression (9) for the values $\alpha > 10.5$ leads to the understated values of the polaron energy. So, if we try to apply the formula (9) for the strong coupling region, then the sign of ΔE changes and the polaron energy "fails", because the kinetic energy, the role of which in the effective polaron functional (3) in the strong-coupling region is played by the expression (9), becomes negative. This circumstance allows us to immediately notice the error of the results. At the same time, if we express the argument $D(s)$ through $\arcsin\left[\mathrm{Im}\,D(s)/|D(s)|\right]$, ignoring the verification of the admissible values range would lead to quite plausible results: approximately from $\alpha = 12$, there is a significant decrease in energy with output under $\alpha \approx 15$ of polaron energy to the quadratic dependence on α: $E_p = -0.135\alpha^2$ for $1/\lambda << 1$. This dependence passes below the same given in [7], obtained using the formula (22) in the rang of $1/\lambda << 1$.

Fig. 2a shows the polaron energy E_p obtained using the functional (3). Minimization was carried out with account of the expression (10), which makes it possible to extend the allowable range of function $D(s)$ to the entire upper half-plane. For the regions with $\mathrm{Re}\,D(s) > 0$, the argument was found using Eq. (8), and if $\mathrm{Re}\,D(s) \le 0$ by Eq. (10):

$$\Delta E = \frac{3}{2\pi}\int_0^\infty dk \cdot k \cdot \arg D\left(\omega_k^2\right);$$

$$\arg D(s) = \begin{cases} \arcsin\left[\mathrm{Im}\,D(s)/|D(s)|\right], & \text{if } \mathrm{Re}D(s) \ge 0 \\ \pi - \arcsin\left[\mathrm{Im}\,D(s)/|D(s)|\right], & \text{if } \mathrm{Re}D(s) < 0 \end{cases}. \quad (23)$$

Calculations were carried out including α = 26. For the accuracy of up to 6 significant digits, the results are the same as those obtained by us for calculations using Eq. (15).

The questions devoted to investigation of the asymptotic behavior of the polaron energy obtained using the expression (15) in the strong coupling limit deserve special consideration. According to Klimin and Devriz calculations [9], in the limit α→∞ the polaron energy determined using the expression (15) goes to the asymptotics $E_p = -0.31683\alpha^{4/3}$, and not to the quadratic dependence for the strong coupling limit obtained by Tulub in [5]. At the same time, the authors of [9] refer to the work of Porsch and Röseler [10], which contains, as they believe, confirmation of their viewpoint. However, the authors of [10] do not criticize the work by Tulub, which they develop, reproducing the previously the results given in [5]. Moreover, in the abstract of ref. [10] it is indicated that the polaron energy in the strong-coupling region is described by the well-known quadratic dependence on α. Tulub's commentary, related to the remarks made in [4, 5], is given in [6].

The asymptotic behavior of the expressions (15) and (23) in the region of extremely large α deserves self-examination and will be carried out by the author of this article in a separate work.

The author is grateful to A.V. Tulub for numerous fruitful scientific advice and discussion of the results of work.

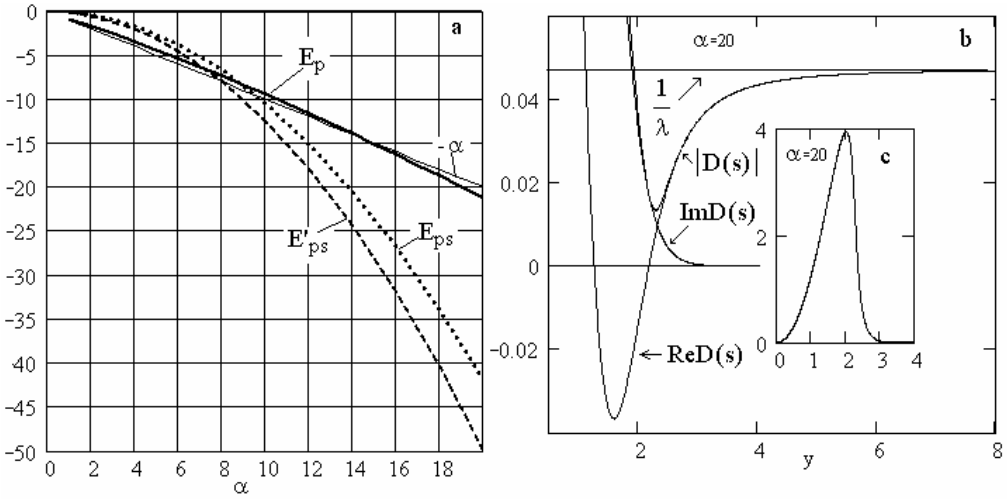

Fig. 2. (a) Polaron energy E_p dependence on α; the energies E_{ps} and E'_{ps} correspond to the minima of the functionals J_{ps} of Eq. (21) and J'_{ps} of Eq. (22). (b) Dependence of the function $D\left(\omega_k^2\right)$ on the dimensionless parameter $y = k/a$. (c) Dependence of the integrand (23) on y, the factor $3a^2/2\pi$ is omitted.

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