

On the Bipolaronic Mechanism of High-Temperature Superconductivity in "Ginzburg Sandwiches" FeSe-SrTiO_3 ; $\text{SrTiO}_3\text{-FeSe-SrTiO}_3$

Stepan Beril*, Alexander Starchuk

Department of Theoretical Physics, T. G. Shevchenko Pridnestrovian State University, Tiraspol, Moldova

Email address:

president@spsu.ru (Stepan Beril), star-alex@idknet.com (Alexander Starchuk)

*Corresponding author

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Abstract: The mechanism of experimentally observed high-temperature superconductivity (HTSC) in thin FeSe films on SrTiO_3 -type substrates has been theoretically investigated. Applying the theory of large-radius bipolaronic states developed based on the exact Hamiltonian of electron-phonon interaction for arbitrary multilayer structures, the bipolaronic mechanism of Cooper pairing of polarons in FeSe monolayers on SrTiO_3 substrates is investigated and in three-layer structures $\text{SrTiO}_3\text{-FeSe-SrTiO}_3$, which are typical "Ginzburg sandwiches". Approach proposed by Ginzburg to enhance the electron-phonon interaction and achieve HTSC by separating the regions where electrons are located (forming Cooper pairs or bipolarons) with the regions in which excitons are excited (or inertial polarization is induced), made it possible to implement the criteria for the formation of bipolaronic states in multilayer structures with high binding energy, due to the possibility of selecting optimal geometric and material parameters (layer thicknesses, dielectric permittivity, optical frequencies, effective masses). It is shown that the binding energy of bipolarons (E_{bp}) in these structures is in the range of values for which bipolarons remain stable quasiparticles and can exist at temperatures significantly higher than their Bose condensation temperature. The formation of bipolarons with high binding energy in the FeSe monolayer on the SrTiO_3 substrate provides the emergence of a bipolaronic HTS with a critical temperature (T_c) more than an order of magnitude higher than T_c for massive FeSe crystals. At the same time, the binding energy of the bipolaron in the FeSe layer with thickness d on the SrTiO_3 substrate increases exponentially with decreasing d ($E_{bp} \propto \exp(-d/R_S)$, R_S is the radius of the polaron) and reaches its maximum value in the limit of the multilayer film FeSe ($d \rightarrow 0$). The presented theory allows modeling a multilayer system and determining the range of values of the material and geometric parameters of layers forming a multilayer structure with a large number of FeSe layers in which T_c values in the room temperature range can be achieved.

Keywords: Bipolarons, High-Temperature Superconductivity, Electron-Phonon Interaction, Multilayer Structures

1. Introduction

The discovery of high-temperature superconductivity (HTSC) in layered iron chalcogenides stimulated the great interest of researchers, since the nature of HTSC in these materials, as well as a number of their physical properties, differ significantly from compounds Y-Ba-Cu-O [1], in which the HTSC was first opened with $T_c > 92\text{K}$ [2]. Detailed experimental and theoretical studies of systems:

multilayer film FeSe, applied to the substrate SrTiO_3 ; intercalated connections based on FeSe; multilayer systems based on FeSe, TiO_2 , SrO , SrTiO_3 they were aimed at developing new models of HTSC, since the existing theories proved to be inapplicable to explain the reasons for such a high growth T_c ($\sim 80 \div 100\text{K}$) [3, 4] in comparison with T_c for bulk crystals FeSe, the critical temperature of which did not exceed the values in the interval $T_c \sim (8 \div 10)\text{K}$. Research of HTSC in the system: multilayer film FeSe on a

substrate of a highly polar semiconductor SrTiO_3 on a substrate of a highly polar semiconductor T_c in a monolayer FeSe up to record temperatures $\sim 100\text{K}$.

In the review [5], a systematic review and analysis of the results of theoretical and experimental studies of HTSC in structures of the type $\text{FeSe}/\text{SrTiO}_3$ and in other compounds and possible mechanisms for increasing T_c in these systems, representing typical "Ginzburg sandwiches" [6, 7].

We will highlight the most relevant models from the work [5] that provide the possibility of implementing the exciton mechanism of the Ginzburg HTSC:

- 1) Magnification model T_c in a monolayer FeSe due to the interaction of charge carriers moving in the conduction band of the layer FeSe, with elementary excitations in the substrate SrTiO_3 , by the picture of the exciton mechanism of Ginzburg [6-8].
- 2) The exciton mechanism of Allender–Bray–Bardin [9], which, as shown in the review [5], was considered when explaining high T_c in FeSe monolayers on substrates SrTiO_3 (BaTiO_3), but it turned out to be not quite effective.
- 3) Phonon mechanism in which the interaction of electrons in a metal monolayer FeSe with optical phonons of the substrate SrTiO_3 is considered as a variant of the implementation of the exciton mechanism in the geometry of the "Ginzburg sandwich";
- 4) Nonadiabatic phonon superconductivity due to a significant excess of the energy of the optical phonon SrTiO_3 Fermi energy E_F [10]. In this case, the interaction of electrons with high-energy optical phonons is considered a possible mechanism for increasing T_c in the system $\text{FeSe}/\text{SrTiO}_3$.

Based on the analysis of these mechanisms, it was concluded that high growth T_c in a monolayer FeSe on the substrate SrTiO_3 in comparison with bulk FeSe is caused by an additional pairing mechanism arising from the interaction of electrons with optical phonons SrTiO_3 , implementing a kind of "pseudo-exciton" mating mechanism.

Thus, in all the selected models, the interaction with the optical phonons of the substrate to one degree or another plays a decisive role in the formation of high values T_c .

Experiments high-resolution electron energy loss spectroscopy (HREELS) have confirmed the presence of a strong electron-phonon interaction at the interfaces $\text{FeSe}/\text{SrTiO}_3$ [11]. Experimental evaluation of the interaction constant of an electron in a monolayer FeSe with a surface optical phonon $\omega_{\text{SLO}} = 92\text{meV}$ of the substrate SrTiO_3 confirmed the presence of a strong electron-phonon bond with $\alpha \sim 1$ (it should be noted that the electron in FeSe interacts with the field of surface optical phonons of the substrate SrTiO_3 , since bulk optical phonons of SrTiO_3 do not create fields outside the substrate due to the confinement effect).

Experimental studies of the role of electron-phonon interaction in the enhancement of superconductivity in films were carried out FeSe on the substrates SrTiO_3 , including in the structure: monolayer FeSe applied to undoped and doped substrates SrTiO_3 [12].

In HREELS experiments with two-dimensional energy and momentum mapping, surface phonon excitations were investigated as in a monolayer and cleaned the surface separately (001) SrTiO_3 .

The electronic structure of the monolayer was determined by angle-resolved photoemission spectroscopy (ARPES). It is established that the electrons of the layer FeSe interact with surface optical vibrations SrTiO_3 , which leads to the formation of dynamic interphase polarons. In the microscopic model [13] it is shown that the interaction "polaron-polaron" in the layer FeSe can cause the additional attraction between electrons, which, in turn, leads to increased superconductivity (growth T_c).

Thus, it is of interest to carry out a consistent theoretical description of polaronic and bipolaronic states in a monolayer film FeSe in the system $\text{FeSe}/\text{SrTiO}_3$, based on the first principles and on its basis to estimate the critical temperature of Bose-condensation of bipolarons. Since the polaronic and bipolaronic states in a multilayer film structure, they are formed as a result of the interaction of electrons from the conduction band FeSe with surface optical phonons SrTiO_3 , the resulting bipolaronic states should be considered as surface ones (it was mentioned earlier that bulk optical vibrations do not create electric fields outside the surface SrTiO_3 (in a monolayer FeSe).

It should be noted that the approach proposed by Ginzburg [6–8] to enhance the electron-phonon interaction and achieve HTSC by separating the regions where electrons are located (forming Cooper pairs or bipolarons) with the regions in which excitons are excited (or inertial polarization is induced), made it possible to implement the criteria for the formation of bipolaronic states in multilayer structures with high binding energy, due to the possibility of selecting optimal geometric and material parameters (layer thicknesses, dielectric permittivity, optical frequencies, effective masses) [13].

The formation of bipolarons in ionic crystals was first investigated by Pekar [14] in the framework of the continuum theory of polarons. Ginzburg [15] considered the Bose condensate of bipolarons in the study of superfluidity. The superfluidity of a charged Bose gas and the bipolaronic mechanism of superconductivity were investigated [16, 17]. Even though the mechanisms of formation of bipolarons and Cooper pairs differ in the methods of description, in essence, the basis of both effects is the electron-phonon interaction.

The criteria for the formation of bipolarons in homogeneous polar crystals (media) were investigated [18, 20]. In particular, the following criteria for the formation of large-radius bipolaronic states in polar crystals were obtained [18]:

$$\varepsilon_0 / \varepsilon_\infty > 20, \alpha \geq 10, \quad (1)$$

which have been improved by improving the variational

method [20]:

$$\varepsilon_0 / \varepsilon_\infty > 10, \alpha_0 \geq 7.3, \quad (2)$$

where: ε_0 , ε_∞ – static and high-frequency dielectric permittivity, respectively;

$$\alpha_0 = \frac{e^2}{\hbar} \left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0} \right) \left(\frac{m^*}{2\hbar\omega_{LO}} \right)^{\frac{1}{2}}$$

– the Pekar–Fröhlich constant of the electron-phonon interaction.

Note that criteria (1) and conditions (2) are difficult to implement for most homogeneous polar semiconductor crystals, so the formation of highly stable bipolaronic states with high binding energies is problematic. In this paper, based on the theory of bipolaronic states of large radius developed by us using the exact Hamiltonian of electron-phonon interaction for arbitrary multilayer structures, the processes of interaction of polarons and the formation of bipolaronic states in "Ginzburg sandwiches" are investigated: monolayer FeSe on the substrate SrTiO₃ and in a monolayer FeSe in a three-layer structure SrTiO₃/FeSe/SrTiO₃. In the systems under consideration, the critical temperature of condensation of bipolarons and the formation of a superfluid bipolaronic condensate have been estimated.

2. Basic Provisions of the Theory of Large-Radius Bipolarons in a Three-Layer Structure with a Quantum Well

Considered a three-layer structure $\langle 10 | 2 | 30 \rangle$ consisting of two semi-infinite polar crystals $\langle 10 \rangle$, $\langle 20 \rangle$ and non-polar semiconductor quantum layer $\langle 2 \rangle$ thickness d ($d \sim \lambda$, λ – the de Broglie wavelength of an electron in a layer $\langle 2 \rangle$) located between them and having a dielectric constant ε_2 .

The high-frequency and static dielectric permittivity of polar crystals are respectively equal: $\varepsilon_{i\infty} \equiv \varepsilon_i$, ε_{i0} ($i=1,3$).

2.1. The Hamiltonian of the Electron-Phonon Interaction

The theory is based on the exact Hamiltonian of the electron-phonon interaction for arbitrary multilayer systems [21–22].

About this system with two electrons in the layer (2) bordering on a semi-infinite polar crystal $\langle 10 \rangle$, $\langle 30 \rangle$, the interaction Hamiltonian of the electrons in the layer $\langle 2 \rangle$ with surface optical phonons of a polar crystal $\langle 10 \rangle$ and $\langle 30 \rangle$ has the form:

$$\begin{aligned} \hat{H}_{e_n-ph}^s = \sum_{\substack{\eta, s=1,2, \\ n=1,2}} C(\eta) e^{i\eta\rho_{e_n}} \left\{ \left[B_1 K_{13,22} + B_2 K_{13,24} F_{21} \right] \cosh \eta z_{e_n} + \left[B_1 K_{23,22} + B_2 K_{23,34} F_{21} \right] \sinh \eta z_{e_n} \right\} (\hat{b}_{1,-\eta}^+ + \hat{b}_{1,\eta}) \\ + \left\{ \left[B_3 K_{13,12} + B_4 K_{13,24} F_{21} \right] \cosh \eta z_{e_n} + \left[B_3 K_{23,22} F_{12} + B_4 K_{23,34} \right] \sinh \eta z_{e_n} \right\} (\hat{b}_{2,-\eta}^+ + \hat{b}_{2,\eta}) \end{aligned} \quad (3)$$

where

$$C(\eta) = \frac{e\sqrt{\hbar}}{4\sqrt{\varepsilon_0 \eta L_x L_y \sinh\left(\frac{\eta d}{2}\right)}}; \quad (4)$$

$$B_{1,2} = \frac{\omega_{1,3} \sqrt{2(\varepsilon_{1,30} - \varepsilon_{1,3})}}{\sqrt{\Omega_1(1 + F_{21}^2)}}; B_{3,4} = \frac{\omega_{1,3} \sqrt{2(\varepsilon_{1,30} - \varepsilon_{1,3})}}{\sqrt{\Omega_2(1 + F_{12}^2)}}; \quad (5)$$

$$K_{13,22} = -\frac{\tanh \frac{\eta d}{2}}{\tilde{B}} \left(\varepsilon_3 \coth \frac{\eta d}{2} + \varepsilon_3 \right); \quad (6)$$

$$K_{23,22} = \frac{\varepsilon_2 \tanh \frac{\eta d}{2} + \varepsilon_3}{\tilde{B}}; \quad (7)$$

$$K_{13,24} = \frac{\tanh \frac{\eta d}{2}}{\tilde{B}} \left(\varepsilon_2 \coth \frac{\eta d}{2} + \varepsilon_1 \right); \quad (8)$$

$$K_{23,24} = -\frac{\varepsilon_2 \tanh \frac{\eta d}{2} + \varepsilon_1}{\tilde{B}}; \quad (9)$$

$$F_{12} = -\frac{\omega_1 \omega_3 \varepsilon_2 \sqrt{\varepsilon_{10} - \varepsilon_1} \cdot \sqrt{\varepsilon_{30} - \varepsilon_3}}{\sinh \eta d [\tilde{B} \Omega_2^2 - \omega_1^2 [\varepsilon_2^2 + \hat{\varepsilon}_2 (\varepsilon_3 + \varepsilon_{10}) + \varepsilon_{10} \varepsilon_{30}]]}; \quad (10)$$

$$F_{21} = -\frac{\omega_1 \omega_3 \varepsilon_2 \sqrt{\varepsilon_{10} - \varepsilon_1} \cdot \sqrt{\varepsilon_{30} - \varepsilon_3}}{\sinh \eta d [\tilde{B} \Omega_1^2 - \omega_3^2 [\varepsilon_2^2 + \hat{\varepsilon}_2 (\varepsilon_1 + \varepsilon_{30}) + \varepsilon_{10} \varepsilon_{30}]]}; \quad (11)$$

$$\Omega_{1,2}^2 = \frac{1}{2} p_1 \pm \sqrt{\frac{1}{4} p_1^2 - p_2}; \quad (12)$$

$$p_1 = \frac{1}{\tilde{B}} \{ \omega_1^2 [\varepsilon_2^2 + \hat{\varepsilon}_2 (\varepsilon_3 + \varepsilon_{10}) + \varepsilon_{10} \varepsilon_{30}] + \omega_3^2 [\varepsilon_2^2 + \hat{\varepsilon}_2 (\varepsilon_1 + \varepsilon_{30}) + \varepsilon_{30} \varepsilon_{10}] \}; \quad (13)$$

$$p_2 = \frac{\omega_1^2 \omega_3^2}{\tilde{B}} [\varepsilon_2^2 + \hat{\varepsilon}_2 (\varepsilon_{10} + \varepsilon_{30}) + \varepsilon_{30} \varepsilon_{10}]; \quad (14)$$

$$\tilde{B} = \varepsilon_2^2 + \hat{\varepsilon}_2 (\varepsilon_1 + \varepsilon_3) + \varepsilon_1 \varepsilon_3; \hat{\varepsilon}_2 = \varepsilon_2 \coth 2\eta d. \quad (15)$$

The complete Hamiltonian of the system under consideration with two electrons in the layer $\langle 2 \rangle$ has the form:

$$\hat{H} = \hat{K}_{e_1} + \hat{K}_{e_2} + V_{e_1-e_2} + V_{SA}(z_1, z_2) + \hat{H}_{ph}^S + \hat{H}_{e-ph}^S + V_B. \quad (16)$$

The following notation is used in formula (14): K_{e_n} ($n=1, 2$) – operators of kinetic energies of electrons:

$$\hat{K}_{e_n} = \frac{\hat{p}_{\parallel e_n}^2}{2m_{e_{\parallel}}^*} + \frac{\hat{p}_{\perp e_n}^2}{2m_{e_{\perp}}^*}, n=1, 2 \quad (17)$$

(designations « \parallel » and « \perp » refer respectively to the directions parallel and perpendicular to the z axis; the latter is perpendicular to the boundaries of the layer $\langle 2 \rangle$, and the origin is in its center). The potential energy of the electrons, due to both the direct electron-electron interaction and the interaction of each of the electrons with fast polarization of the layer and the crystals bordering it, has the form [23]:

$$V_{e_1-e_2} \equiv V_{e_1-e_2}(\rho, z_{e_1}, z_{e_2}) = \frac{e^2}{4\pi\varepsilon_0\varepsilon_2} \int_0^\infty d\eta \mathcal{Y}_0(\eta\rho) \{ e^{-\eta|z_{e_1}-z_{e_2}|} + \frac{2}{e^{2\eta d} - \delta_1\delta_3} [\delta_1\delta_3 \text{ch}(\eta(z_{e_1} - z_{e_2})) + e^{\eta d} \times (f_1 \cosh(\eta(z_{e_1} + z_{e_2})) + f_2 \sinh(\eta(z_{e_1} + z_{e_2})))], \quad (18)$$

where the following designations are introduced:

$$\delta_{1,3} = \frac{\varepsilon_2 - \varepsilon_{1,3}}{\varepsilon_2 + \varepsilon_{1,3}}, \quad (19)$$

$$f_1 = \frac{\varepsilon_2^2 - \varepsilon_1\varepsilon_3}{(\varepsilon_2 + \varepsilon_1)(\varepsilon_2 + \varepsilon_3)}, \quad f_2 = \frac{(\varepsilon_1 - \varepsilon_3)\varepsilon_2}{(\varepsilon_2 + \varepsilon_1)(\varepsilon_2 + \varepsilon_3)}; \quad (20)$$

$J_0(x)$ – zero-order Bessel function;

$$\hat{H}_{ph}^S = \sum_{\eta,s=1,2} \hbar\Omega_s \hat{b}_{\eta,s}^+ \hat{b}_{\eta,s} \quad (21)$$

– energy operator of surface optical phonons; $\hat{H}_{e_n-ph}^S$ ($n=1,2$) is the interaction Hamiltonian of the electrons in the layer $\langle 2 \rangle$ with surface optical phonons of a polar crystal $\langle 10 \rangle$ and $\langle 30 \rangle$, which has the form (3) – (15).

The potential energy of interaction of each electron with the induced fast polarization of the layer and neighboring crystals (the so-called self-action energy) can be written as:

$$V_{SA}(z_{e_n}) = \frac{e^2}{4\pi\epsilon_0\epsilon_2} \int_0^\infty \frac{d\eta}{e^{2\eta d} - \delta_1\delta_3} \{ \delta_1\delta_3 + e^{2\eta d} (f_1 \text{ch}(2\eta z_{e_n}) + f_2 \text{sh}(2\eta z_{e_n})) \}. \quad (22)$$

The potential energy of electrons in a rectangular well with infinite barriers is equal to

$$V_B(z_{e_n})|_{n=1,2} = \begin{cases} 0, -\frac{d}{2} < z_{e_n} < \frac{d}{2}; \\ \infty, z_{e_n} \leq -\frac{d}{2}, z_{e_n} \geq \frac{d}{2}. \end{cases} \quad (23)$$

2.2. Effective Hamiltonian of the System

We assume that the thickness of the d layer $\langle 2 \rangle$ is so small that the energy of dimensional quantization is much greater than the energy of phonons and electron-phonon interaction. In this approximation, the motion of an electron along the z -axis can be considered as fast and the wave functions can be chosen in the form describing the ground dimensional quantized state:

$$\psi(z_{e_1}, z_{e_2}) = \left(\frac{2}{d} \right) \cos\left(\frac{\pi z_{e_1}}{d} \right) \cos\left(\frac{\pi z_{e_2}}{d} \right). \quad (24)$$

After averaging the Hamiltonian (16)–(23) on the wave function (24), we exclude the variables z_{e_1} , z_{e_2} and we get a quasi-two-dimensional problem with a Hamiltonian

$$\hat{H}_1(\rho) = \left\langle \psi(z_{e_1}, z_{e_2}) | \hat{H}(\rho, z_{e_1}, z_{e_2}) | \psi(z_{e_1}, z_{e_2}) \right\rangle. \quad (25)$$

Let's place the origin of the coordinate system XOY to the center of mass of the electronic system (the radius vectors of the electrons are respectively denoted ρ_{e_1} , ρ_{e_2}).

We perform over the Hamiltonian $\hat{H}_1(\rho)$ unitary transformation

$$\hat{H}_2(\rho) = \hat{U}_1^{-1} \hat{H}_1 \hat{U}_1, \quad (26)$$

where the conversion operator \hat{U}_1 has the form

$$\hat{U}_1 = \exp \left\{ \sum_{\eta,s=1,2} f(\eta, \rho_1, \rho_2) \hat{b}_{s,\eta}^+ \hat{b}_{s,\eta} \right\}; \quad (27)$$

here: $f(\eta, \rho_{e_1}, \rho_{e_2})$ – variational amplitudes of displacement of phonon mode operators.

When averaging the resulting Hamiltonian \hat{H}_2 by phonon vacuum $|\Phi_0\rangle$ we obtain an effective Hamiltonian that does not contain phonon variables:

$$\hat{H}_3(\rho_{e_1}, \rho_{e_2}) = \langle \Phi_0 | \hat{H}_2 | \Phi_0 \rangle. \quad (28)$$

Let's choose the variational amplitudes of the displacement in the formula (27) in a form similar to:

$$f(\eta, \rho_{e_n})|_{n=1,2} = \frac{C(\eta)F_n(\eta)}{\hbar\Omega_s} \left\{ \frac{e^{i\eta\rho_{e_1}} + e^{i\eta\rho_{e_2}}}{1 + \eta^2 R_s^2} + \frac{\lambda}{(1 + \beta^2 \eta^2 R_s^2)^2} \right\}, \quad (29)$$

where λ and β are variational parameters, [20, 23, 24]

$$R_s = \left(\frac{\hbar}{2m^* \Omega_s} \right)^{1/2}, \quad s = 1, 2.$$

When $\lambda = 0$ function $f(\eta, \rho_1, \rho_2)$ it takes the form of the Haken amplitude [25], taking into account the specific nature of the system under consideration. Factors $F_n(\eta)|_{n=1,2}$ have the form:

$$F_1(\eta) = \frac{\pi^2 \sinh\left(\frac{\eta d}{2}\right) [B_1 K_{13,22} + B_2 F_{21} K_{13,24}]}{\frac{\eta d}{4} \left(\frac{\eta^2 d^2}{16} + \pi^2 \right) \sqrt{\hbar\Omega_1}}; \quad (30)$$

$$F_2(\eta) = \frac{\pi^2 \sinh\left(\frac{\eta d}{2}\right) [B_3 K_{13,22} F_{12} + B_4 F_{13} K_{13,24}]}{\frac{\eta d}{4} \left(\frac{\eta^2 d^2}{16} + \pi^2 \right) \sqrt{\hbar\Omega_2}}; \quad (31)$$

The second term in formula (29) was introduced according to the theory of bulk bipolaronic and exciton states in polar crystals to more accurately describe the distribution of slow polarization induced by the field of a two-charge system [20, 23, 24]: two electrons in a bipolaron [20, 24] or an electron and a hole in an exciton [26, 27]. It should be noted that the variational parameter λ describes the magnitude of the

induced polarization charge arising in a two-electron system in addition to the polaron charge, which is localized near the center of mass, the variational parameter β takes into account the size of the region in which the induced polar charges are distributed. As a result, we obtain the effective Hamiltonian of the system:

$$\hat{H}_{eff} = E_0(d) - \sum_{n=1,2} \frac{\hbar^2}{2m_{e_n\perp}} \Delta_{e_n} + W_{eff} + W_p + E_p(d). \quad (32)$$

Term

$$E_0(d) = \frac{\pi^2 \hbar^2}{m_{e_n\parallel} d^2} + \langle V_{SA} \rangle \quad (33)$$

includes the energy of the main level of dimensional quantization and the energy of self-action:

$$\langle V_{SA} \rangle = \frac{e^2}{4\pi\epsilon_0 \epsilon_2 d} \int_0^\infty \frac{dx}{e^{2x} - \delta_1 \delta_3} \left\{ \delta_1 \delta_3 + f_1 \frac{\pi^2 e^x \sinh x}{x(x^2 + \pi^2)} \right\}.$$

The term represents the effective potential of electron-electron interaction and includes energy $V_{e_1-e_2}$ after averaging the Hamiltonian (16) on the wave function (24) and the electron-phonon interaction energy describing the interaction of each of the electrons with the induced polarization of the other:

$$W_{eff}(\rho, d) = V_{e_1, e_2}(\rho, d) + \frac{e^2}{16\pi\epsilon_0 d} \int_0^\infty J_0\left(\frac{x\rho}{d}\right) \times \{F_1^2(x) a_{11}(a_{11}-2) + F_2^2(x) a_{12}(a_{12}-2)\} dx, \quad (34)$$

where:

$$a_{1s}(x) = \left(1 + \frac{R_s^2 x^2}{d^2}\right)^{-1}, \quad s=1,2. \quad (35)$$

Term W_p in the right part (32)

$$W_p(\rho_{e_1}, \rho_{e_2}, d) = \sum_{\eta, n=1,2} |C(\eta)F_n(\eta)|^2 \times \{2(2a_{1s}-1)a_{2s} \cos(\eta \rho_{e_n})\}. \quad (36)$$

is determined by the second term in the formula (29):

$$a_{2s} = \lambda(1 + \beta^2 \eta^2 R_s^2)^{-2}. \quad (37)$$

It describes the potential energy of the interaction of an electron with a polarization charge induced by the joint action of both electrons, which is located in the region of the center of mass of a two-electron system. W_p makes an additional contribution to the attraction of electrons and the formation of a bipolaron.

The coordinate-independent part of the potential energy $E_p(d)$ in formula (32) has the form:

$$E_p(d) = \sum_{\substack{\eta, n=1,2, \\ s=1,2}} |C(\eta)F_n(\eta)|^2 \times (2a_{1s}^2 - 4a_{1s} + 2a_{1s}^2 R_s^2 \eta^2 + a_{2s}^2) \quad (38)$$

and takes into account the polar energy, as well as the interaction of each of the electrons with the polarization created by it and the energy of the elastically deformed lattice.

2.3. Investigation of the Effective Potential of Electron-Electron Interaction

It is of interest to investigate the profile of the effective potential of electron-electron interaction $W_{eff}(\rho, d)$, defined by formulas (30), (31), for two actual cases:

- a) in a monolayer FeSe, applied to a massive polar substrate SrTiO₃ (one interface FeSe/SrTiO₃), in which HTSC was observed experimentally;
- b) in a monolayer FeSe, separating the crystal layers SrTiO₃ (two interfaces FeSe/SrTiO₃).

Both cases under consideration are typical structures –

"Ginzburg sandwiches", in which two electrons from the conduction band of a monolayer film FeSe interact with the surface optical phonons of the substrate SrTiO₃ (case a)) and monolayer film FeSe, separating polar crystals SrTiO₃ (case b)).

As it was shown, due to boundary conditions, bulk longitudinal optical oscillations in SrTiO₃ do not create electric fields in neighboring media (confinement effect), in this case – in a monolayer FeSe, in which free electrons will interact only with surface optical phonons SrTiO₃ [21, 22, 24]. We will assume that the FeSe monolayer is a quasi-two-dimensional quantum well with infinite walls.

For case a), the effective potential of the electron-electron interaction can be obtained from formulas (34), (35) in the limit $d \rightarrow 0$:

$$W_{eff}^{2D}(\rho) = \frac{e^2}{2\pi\epsilon_0(\epsilon_1+1)\rho} + \frac{e^2}{2\pi\epsilon_0 R_s} \left\{ \frac{1}{\epsilon_1+1} - \frac{1}{\epsilon_{10}+1} \right\} \int_0^\infty J_0\left(\frac{\rho x}{R_s}\right) \left(\frac{x^2-1}{x^2+1} \right) dx, \quad (39)$$

where:

$$R_s = \left(\frac{\hbar}{2m_e^* \Omega_s} \right)^{\frac{1}{2}}; \quad \Omega_s = \omega_1 \sqrt{\frac{\epsilon_{10}+1}{\epsilon_1+1}}; \quad (40)$$

Ω_s – the frequency of the longitudinal surface optical phonon.

Carrying out the integration, we get:

$$W_{eff}^{2D}(\rho) = \frac{e^2}{2\pi\epsilon_0\rho} \left(\frac{2}{\epsilon_1+1} - \frac{1}{\epsilon_{10}+1} \right) - \frac{e^2}{2\epsilon_0 R_s} \left(\frac{1}{\epsilon_1+1} - \frac{1}{\epsilon_{10}+1} \right) \left\{ I_0\left(\frac{\rho}{R_s}\right) - L_0\left(\frac{\rho}{R_s}\right) \right\}. \quad (41)$$

Here $I_0(x)$, $L_0(x)$ – modified zero-order Bessel and Struve functions, respectively.

Then the effective Hamiltonian of the system has the form:

$$\hat{H}_{eff}^{2D} = -\frac{\hbar^2}{2m_{e_1}^*} \Delta_{\rho_1} - \frac{\hbar^2}{2m_{e_2}^*} \Delta_{\rho_2} + W_{eff}^{2D}(\rho). \quad (42)$$

Figure 1 shows graphs of the dependence $W_{eff}(\rho)$ for case a): vacuum-monolayer film FeSe–SrTiO₃ for three different values of the effective mass of the electron в FeSe.

For case b), the effective potential of the electron-electron interaction is obtained from formulas (34), (35), taking into account the symmetry of the structure SrTiO₃ – multilayer film FeSe – SrTiO₃:

$$W_{eff}(\rho) = \frac{e^2}{2\pi\epsilon_0\rho} \left(\frac{2}{\epsilon_1} - \frac{1}{\epsilon_{10}} \right) - \frac{e^2}{2\epsilon_0\bar{R}_S} \left(\frac{1}{\epsilon_1} - \frac{1}{\epsilon_{10}} \right) \left\{ I_0 \left(\frac{\rho}{\bar{R}_S} \right) - L_0 \left(\frac{\rho}{\bar{R}_S} \right) \right\}, \quad (43)$$

where

$$\bar{R}_S = \left(\frac{\hbar}{2m_e^* \bar{\Omega}_S} \right)^{1/2}; \quad \bar{\Omega}_S = \alpha_1 \sqrt{\frac{\epsilon_{10} + \epsilon_1}{2\epsilon_1}}.$$

Note that in the case of a symmetric structure SrTiO₃–FeSe–SrTiO₃ the electrons in the $\langle 2 \rangle$ layer will interact with only one surface optical mode of each of the crystals SrTiO₃ ($\Omega_{S_1} = \Omega_{S_2} \equiv \bar{\Omega}_S$; $\alpha_{S_1} = \alpha_{S_2}$).

The dependency graph $W_{eff}(\rho)$ for the case SrTiO₃–FeSe–SrTiO₃ and various values of the effective mass of the electron are shown in Figure 2.

In a study a modification of case b) is discussed when

there is more than one interface between SrTiO₃ and FeSe, and two interfaces (SrTiO₃/FeSe и FeSe/SrTiO₃) [28]. This creates an indirect phonon attraction of electrons twice as strong as in the case of a single interface, which, in turn, will lead to an increase in T_c about twice. Effective potential in this actual case (case b)) can be obtained from formula (43) by doubling the effective potential energy of the interaction of electrons.

Thus, for the case c) the effective potential:

$$W_{eff}^{2D}(\rho) = \frac{e^2}{\pi\epsilon_0\rho} \left(\frac{2}{\epsilon_1+1} - \frac{1}{\epsilon_{10}+1} \right) - \frac{e^2}{\epsilon_0\bar{R}_S} \left(\frac{1}{\epsilon_1+1} - \frac{1}{\epsilon_{10}+1} \right) \left\{ I_0 \left(\frac{\rho}{\bar{R}_S} \right) - L_0 \left(\frac{\rho}{\bar{R}_S} \right) \right\}. \quad (44)$$

The dependency graph $W_{eff}(\rho)$ for case b) and various values of the effective mass of the electron are shown in Figure 3.

As can be seen from Figures 1–3, within $\rho \ll R_S$ и $\rho \gg R_S$ potential $W_{eff}(\rho)$ interactions between electrons have a repulsive character, but the range of values $\rho \sim nR_S$ ($n \sim 1 \div 5$) $W_{eff}^{2D}(\rho)$ changes the nature of behavior from repulsion to attraction.

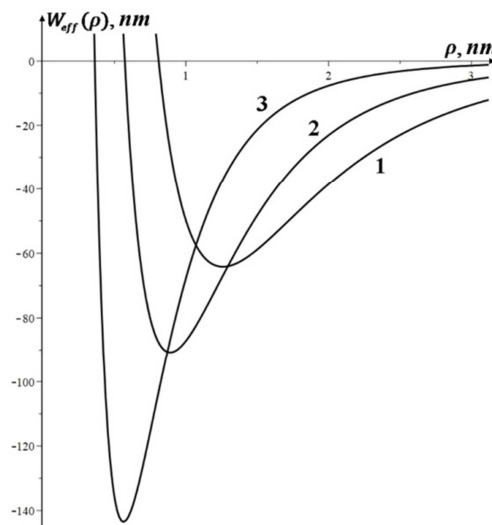


Figure 1. Graphs of the dependence of the effective potential energy for the case SrTiO₃ / FeSe / vacuum. Curve 1 corresponds to $m^* = m_0$, where m_0 – free electron mass, curve 2 – $m^* = 2m_0$, curve 3 – $m^* = 5m_0$.

Analysis of a similar expression for a massive crystal obtained based on the Haken method in the study [25]:

$$W_{eff}^{3D}(r) = \frac{e^2}{\epsilon r} - \frac{e^2}{r} \left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0} \right) \left\{ 1 - e^{-\frac{r}{R_p}} \right\}, \quad (45)$$

shows that under no parameter values ϵ_∞ , ϵ_0 potential W_{eff}^{3D} , as a function of r , it does not change the nature of its behavior and is repulsive.

In the general case, when the polar crystals are different, and the layer $\langle 2 \rangle$ between them has a finite thickness, the calculation of the potential profile $W_{eff}^{2D}(\rho)$ should be performed according to formulas (34)–(35).

The appearance of a tendency to weaken the repulsion of electrons in the FeSe monolayer and the appearance of attraction between them at distances of the order of their polaron radius ($\rho \sim R_S$) occurs due to interaction with the optical phonons of the substrate SrTiO₃ and it is an important condition for the formation of bipolarons.

In the effective Hamiltonian (28), the term $W_p(\rho_{e_1}, \rho_{e_2}, d)$ (formulas (36), (37) describe the potential energy of interaction of electrons in a monolayer of FeSe with an additional polarization charge induced by the joint action of both electrons and located in the region of the center of mass of a two-electron system.

We estimate the density of the total surface charge for case a), which can be calculated by the formula:

$$\begin{aligned} \sigma(\rho) &= 2\epsilon_0 E_z|_{z=0} = \\ -2\epsilon_0 \frac{\partial V_p}{\partial z} \Big|_{z=0} &= \frac{e^2(\epsilon_{10} - \epsilon_1)}{2\pi(\epsilon_1 + 1)(\epsilon_{10} + 1)} \int_0^\infty d\eta \cdot \eta e^{-2\eta z} \times \\ &\left\{ \frac{\lambda_1}{1 + \beta_1^2 R_S^2 \eta^2} [J_0(\eta|\rho_1 - \rho|) + J_0(\eta|\rho_2 - \rho|)] + \right. \\ &\left. \frac{\lambda_2 J_0(\eta\rho)}{1 + \beta_2^2 \eta^2 \rho^2} \right\}, \end{aligned} \quad (46)$$

where: V_p – polarization field potential, z_0 – the distance to the plane in which the electrons are located (monolayer FeSe); λ_1 , λ_2 , β_1 , β_2 – variational parameters.

Considering $\lambda_1 = \lambda_2 = \beta_1 = \beta_2 = 1$, $z_0 = 0$ and by integrating into (38 a), we obtain an explicit expression for $\sigma(\rho)$:

$$\sigma(\rho) = \frac{e^2(\epsilon_{10} - \epsilon_1)}{4\pi(\epsilon_1 + 1)(\epsilon_{10} + 1)R_S^2} \times$$

$$\left\{ 2 \left[K_0 \left(\frac{|\rho_1 - \rho|}{R_S} \right) - K_0 \left(\frac{|\rho_2 - \rho|}{R_S} \right) \right] + \frac{\rho}{R_S} K_1 \left(\frac{|\rho_1 - \rho|}{R_S} \right) \right\}; \quad (47)$$

here: $K_0(x)$, $K_1(x)$ – McDonald functions.

Integrating the expression (47) by ρ gives the value of the effective electric charge e^* induced by electrons:

$$e^* = \frac{e(\epsilon_{10} - \epsilon_1)}{(\epsilon_1 + 1)(\epsilon_{10} + 1)}. \quad (48)$$

As $\epsilon_{10} \gg \epsilon_1 > 1$, then from formula (37) for an effective charge we obtain: $e^* \approx e / (\epsilon_1 + 1)$.

3. The Binding Energy of the Bipolaron

The energy of the ground state of the bipolaron is found by the variational method.

We will choose the trial wave function of the electron pair in the form:

$$\psi(\rho) = 2(3\pi)^{-1/2} \gamma^2 \rho e^{-\gamma\rho}, \quad (49)$$

where γ – variation parameter.

The variational function of the ground state energy has the form:

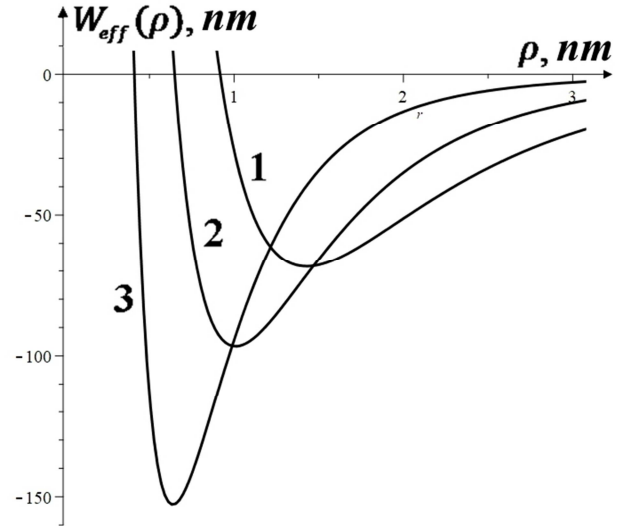


Figure 2. Graphs of the dependence of the effective potential energy for the case SrTiO₃ / FeSe / SrTiO₃. Curve 1 corresponds to $m^* = m_0$, where m_0 – free electron mass, curve 2 – $m^* = 2m_0$, curve 3 – $m^* = 5m_0$.

$$E(\gamma, \beta, \lambda) = \left\langle \psi(\rho) \left| \hat{H}_{eff}(\rho, \rho_1, \rho_2, d) \right| \psi(\rho) \right\rangle. \quad (50)$$

Substituting (41)–(43), (49) into (50) and performing

integration, we obtain an explicit expression for the variational functional of the bipolaronic state energy in the cases under consideration:

a) For contact: vacuum / FeSe / SrTiO₃

$$E(\gamma) = \frac{\hbar^2 \gamma^2}{6m^*} + \frac{e^2 \gamma}{3\pi\epsilon_0} \left[\frac{1}{\epsilon_1 + 1} - \left(\frac{1}{\epsilon_1 + 1} - \frac{1}{\epsilon_{10} + 1} \right) \times \right. \\ \left. (4\gamma^2 R_S^2 - 1)^{-7/2} (384\gamma^6 R_S^6 + 144\gamma^4 R_S^4) \arctan \sqrt{4\gamma^2 R_S^2 - 1} - \right. \\ \left. (64\gamma^6 R_S^6 + 240\gamma^4 R_S^4 - 8\gamma^2 R_S^2 + 1) \sqrt{4\gamma^2 R_S^2 - 1} \right]. \quad (51)$$

b) For a symmetric structure: SrTiO₃ / FeSe / SrTiO₃

$$E(\gamma) = \frac{\hbar^2 \gamma^2}{6m^*} + \frac{e^2 \gamma}{3\pi\epsilon_0} \left[\frac{1}{\epsilon_1} - \left(\frac{1}{\epsilon_1} - \frac{1}{\epsilon_{10}} \right) (4\gamma^2 \bar{R}_S^2 - 1)^{-7/2} \times \right. \\ \left. (384\gamma^6 \bar{R}_S^6 + 144\gamma^4 \bar{R}_S^4) \arctan \sqrt{4\gamma^2 \bar{R}_S^2 - 1} - \right. \\ \left. (64\gamma^6 \bar{R}_S^6 + 240\gamma^4 \bar{R}_S^4 - 8\gamma^2 \bar{R}_S^2 + 1) \sqrt{4\gamma^2 \bar{R}_S^2 - 1} \right]. \quad (52)$$

c) For the structure: SrTiO₃ / FeSe / SrTiO₃ with two interfaces [28]

$$E(\gamma) = \frac{\hbar^2 \gamma^2}{6m^*} + \\ \frac{2e^2 \gamma}{3\pi\epsilon_0} \left[\frac{1}{\epsilon_1 + 1} - \left(\frac{1}{\epsilon_1 + 1} - \frac{1}{\epsilon_{10} + 1} \right) (4\gamma^2 R_S^2 - 1)^{-7/2} \times \right. \\ \left. (384\gamma^6 R_S^6 + 144\gamma^4 R_S^4) \arctan \sqrt{4\gamma^2 R_S^2 - 1} - \right. \\ \left. (64\gamma^6 R_S^6 + 240\gamma^4 R_S^4 - 8\gamma^2 R_S^2 + 1) \sqrt{4\gamma^2 R_S^2 - 1} \right]. \quad (53)$$

The binding energy of a bipolaron is determined by the expression:

$$W_b = E_b - 2E_p, \quad (54)$$

where E_b – the minimum value of the functional $E(\gamma, \beta, \lambda)$ (i. e. the energy of the bipolar system); E_p – the energy of a single polaron.

By $d \rightarrow 0$ we get the expression:

$$E_p = -\frac{\pi}{2} \sum_{s=1,2} \alpha_s \hbar \Omega_s. \quad (55)$$

Based on the variational calculation of the binding energy W_b of the bipolaron in the FeSe layer on the substrate SrTiO₃ according to the formula (51), it is obtained:

$$m_e^* = m_0; E_b = -10,8 \text{ meV}.$$

$$m_e^* = 2m_0; E_b = -17,6 \text{ meV}.$$

$$m_e^* = 5m_0; E_b = -31,3 \text{ meV}.$$

In a three-layer structure SrTiO₃ – FeSe – SrTiO₃ the calculation is made according to the formula (52):

$$m_e^* = m_0; E_b = -13,2 \text{ meV}.$$

$$m_e^* = 2m_0; E_b = -20,6 \text{ meV}.$$

$$m_e^* = 5m_0; E_b = -35,7 \text{ meV}.$$

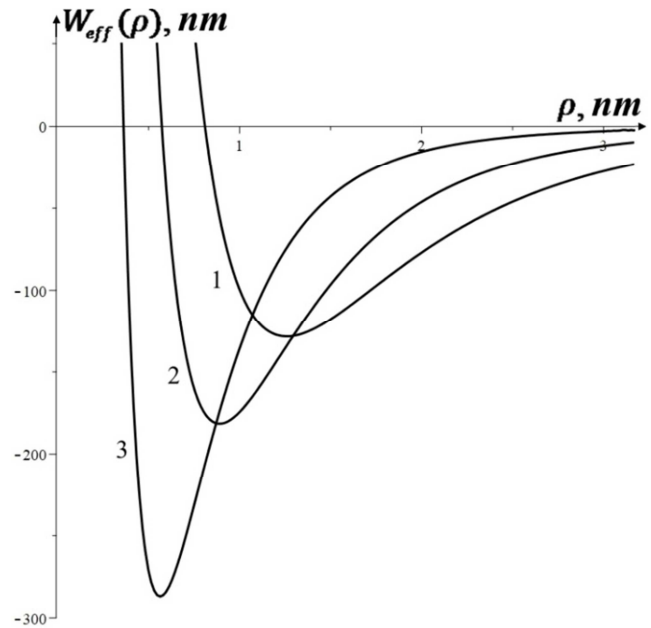


Figure 3. Graphs of the dependence of the effective potential energy with two separate interfaces SrTiO₃ / FeSe and FeSe / SrTiO₃. Curve 1 corresponds to $m^* = m_0$, where m_0 – free electron mass, curve 2 – $m^* = 2m_0$, curve 3 – $m^* = 5m_0$.

In a three-layer structure SrTiO₃ – FeSe – SrTiO₃ with two interfaces [28], the calculation is performed according to the formula (53):

$$m_e^* = m_0; E_b = -27,3 \text{ meV}.$$

$$m_e^* = 2m_0; E_b = -41,2 \text{ meV}.$$

$$m_e^* = 5m_0; E_b = -68,8 \text{ meV}.$$

From the above results for structures: monolayer FeSe film on a substrate SrTiO₃ and SrTiO₃ / FeSe / SrTiO₃ structure it follows that the binding energy of the bipolaron is in the range (150÷900) K. The interaction of electrons in the FeSe conduction band with their induced polarizations and an additional positive polarization charge in the substrate

SrTiO₃, located in the center of mass of a two-electron system (formula (48)), plays an important role in the polarization potential well in the formation of bipolarons in a monolayer film FeSe.

The pairing of the formed polarons is facilitated by the weakening of repulsion between them, due to the interaction of electrons with an additional polarizing charge, which turns into attraction, as is the case in structures FeSe/SrTiO₃ and SrTiO₃/FeSe/SrTiO₃. As follows from expressions (34), (35), the binding energy of the bipolaron increases with decreasing thickness of the FeSe film and reaches a maximum value in the limit $d \rightarrow 0$ (monolayer FeSe film on a substrate SrTiO₃), in this case, the constant of the electron-phonon interaction α_S the structures under study FeSe/SrTiO₃; SrTiO₃/FeSe/SrTiO₃ lies in the area of intermediate and strong coupling: $\alpha \sim (1 \div 5)$.

Note that when considering the Coulomb interaction of electrons with the surface optical phonons of the substrate, the screening effects that lower the binding energy of the bipolaron was not taken into account. At the same time, taking into account the exchange effects during the formation of bipolarons [18, 29], increases their binding energy [18, 29].

4. Assessment of Critical Temperature in Structures FeSe/SrTiO₃; SrTiO₃/FeSe/SrTiO₃

Let us estimate T_c based on the bipolar mechanism of superconductivity in a monolayer FeSe, deposited on a polar semiconductor substrate SrTiO₃.

A bipolaronic model of superconductivity due to the superfluidity of a charged Bose gas was considered [16]. Using the formula for the Bose condensation temperature under the bipolaronic superconductivity mechanism [30], we estimate T_c for case a): multi-layer film FeSe on a polar substrate SrTiO₃:

$$T_c \approx 3,31 \frac{\hbar^2 n_S}{m_{bp} k_0}, \quad (56)$$

where: n_S – concentration of bipolarons; $m_{bp} = 2m_p$ – effective mass of the bipolaron, m_p – the effective mass of the polaron.

For numerical estimates, we use the following parameter values: $m_p = (1 \div 5)m_0$, m_0 – free electron mass; $n_S \approx (10^{13} \div 10^{14}) \text{ cm}^{-2}$ [10].

As a result, for the critical temperature of the transition to the superconducting state, we obtain the interval: $T_c \approx (100 \div 300) \text{ K}$.

Considering that the binding energy of bipolarons lies in the interval: $E_{bp} \approx (150 \div 900) \text{ K}$, it can be assumed that the bipolaron remains a stable quasi-particle during the formation

of a superconducting Bose condensate.

Since the binding energy of a bipolaron in the case of a three-layer structure SrTiO₃/FeSe/SrTiO₃ (in case c)) increases in comparison with case a): FeSe/SrTiO₃ approximately by $(15 \div 20)\%$ and more than twice (in the case of b)), the above estimates T_c it is qualitatively preserved.

Evaluation of the maximum value T_c in the model with the Einstein spectrum of optical phonons for the structures under consideration, in the case of a strong electron-phonon coupling according to [31], the formula gives in the limit:

$$k_0 T_c^{\max} \approx 0,13 \hbar \Omega. \quad (57)$$

Considering that $\hbar \Omega = \hbar \Omega_S$ – the energy of the surface optical phonon SrTiO₃, we get the critical temperature:

$$T_c^{\max} \approx 140 \text{ K},$$

what correlates with the assessment T_c , performed according to the formula (57).

Note that an increase in the thickness of the FeSe film leads to an exponential decrease in the binding energy of the bipolaron in comparison with the FeSe monolayer ($E_b \sim \exp(-d/R_S)$) and accordingly to an exponential decrease T_c ($T_c \sim \exp(-d/R_S)$), where $R_S = (\hbar / (2m_0 \omega_{SLO}))^{1/2}$ – the radius of the surface polaron. The binding energy and the critical temperature T_c reach a maximum in the limit of the monolayer film FeSe ($d \rightarrow 0$).

5. Conclusion

The binding energy of bipolarons is in the range of values $E_{bp} \approx (150 \div 900) \text{ K}$, so bipolarons in the studied "Ginzburg sandwiches": FeSe/SrTiO₃ and SrTiO₃/FeSe/SrTiO₃ are stable quasiparticles and can exist in the structures under consideration at temperatures that can significantly exceed their Bose condensation temperature.

Calculations show that the binding energy of the bipolaron in the FeSe layer on the substrate SrTiO₃ strongly depends on the thickness of the layer ($\sim \exp(-d/R_S)$) and reaches a maximum in the limit of the monolayer film ($d \rightarrow 0$).

High values T_c the structures studied are determined by the high binding energy of bipolarons due to the presence of a highly polar semiconductor substrate SrTiO₃, which unequivocally confirms Ginzburg's prediction [6] about the important role of contact media in achieving high T_c .

The interaction of electrons in a monolayer FeSe film with optical phonons of the substrate SrTiO₃ for $m_e^* = 5m_0$; $E_b = -35,7 \text{ meV}$ was experimentally investigated [12-13]. It is established that the electric field associated with high-energy Fuchs-Kliwer surface phonons SrTiO₃ [32],

penetrates the monolayer FeSe and leads to the formation of polarons in it. It is theoretically shown that the dynamic polaron-polaron interaction in the FeSe layer leads to an increase in superconductivity.

The presented theory makes it possible to simulate the system and determine the range of values of the material and geometric parameters of the layers forming multilayer structures which can be achieved T_c in the area of room temperatures.

These can be multilayer structures of the type of composite superlattices, the layers of which are, along with the FeSe layers, also layers SrO, TiO₂, BaO, and others [28].

In conclusion, we note that in multilayer periodic structures, which are composite superlattices with polar semiconductor layers (type BaO, SrTiO₃ and others) and metal layers (type FeSe, CuO₂ and others), an important role in the formation of bipolarons and growth T_c spatially extended surface phonons can play – new elementary excitations that were predicted theoretically [33, 34] and discovered experimentally [35-37].

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