Microscopic theory for phase-sensitive experiments to determine the symmetry of the order parameter in Fe-based superconductors

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Abstract – We present a microscopic theory for the dc Josephson current, based on the construction of a coherent temperature Green’s function in the tight-binding approximation, in junctions with multiband superconductors. This theory is applied to junctions with a multiband Fe-based superconductor (FeBS) described by $s_{\pm}$-wave and $s_{++}$-wave order parameter symmetries, which is probably realized in FeBS. For the first time, phase dependences of the Josephson current have been calculated for different directions of an interface with respect to crystallographic axes of FeBS. The present approach is also suitable for a consistent description of the Josephson transport in structures with topological superconductors.

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Introduction. – For new unconventional superconductors one of the first goals is to determine the symmetry of the order parameter. It is known that the crucial experiments to determine the order parameter in unusual superconductors rely on some form of phase-coherent tunneling experiments. For the theory a complicating factor is that many of the new unconventional superconductors, such as Sr$_2$RuO$_4$, FeBS, doped superconducting insulators Cu$_x$Bi$_2$Se$_3$, are multiorbital metals. Therefore, a quantitatively correct microscopic theory that describes the coherent tunneling in junctions containing these unconventional superconductors should take into account both interband and intervalley scattering at the boundaries. Such a microscopic theory to describe the current of single-particle excitations in junctions of a normal metal with a multiband superconductor has been proposed recently by Burmistrova et al.\textsuperscript{[1,2]} This theory resolved the long-standing problem of the microscopic description of the interband and intervalley scattering at an interface with multiband superconductors, which was previously described by introducing a phenomenological parameter $\alpha_0$\textsuperscript{[3]}. A parameter $\alpha_0$ describes in a phenomenological way the relative contribution from different bands to the wave function of a two-band superconductor. The aim of this letter is the construction of a consistent microscopic theory of the Josephson tunneling in junctions with multiband superconductors, with an application to FeBS superconductors with the most popular types of conjectured symmetries of the order parameter. Although various attempts to construct a theory have been recently made, quite universally only a limited number of relevant characteristics of the materials have been taken into account. The Ginzburg-Landau approach used in [4] is valid only for temperatures close to critical. The tunnel Hamiltonian approach used in [5] is valid only for small transparencies of the junction, although in real structures, often employing an Andreev point contact, the transparency can have an arbitrary value. The theory in [6] considers Josephson transport in structures with diffuse FeBS, although in unusual superconductors the diffusive scattering usually masks the symmetry of the order parameter\textsuperscript{[7]}. An important contribution to the Josephson current from continuous states is not

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taken into account in the theory of [8]. The theory [9] has considered the Josephson junctions with FeBS both in diffuse and clean cases, but this consideration is based on phenomenological boundary conditions [10,11] which do not take into account interband scattering on the boundary microscopically. The theory [12] has considered the Josephson junctions between a conventional s-wave superconductor and FeBS with \( s_{++} \) and \( s_{\pm} \) pairing symmetry and proposed the experiment for determining the symmetry of an order parameter in FeBS in the case in which there is a spatial inhomogeneity in the Josephson coupling, but this theory [12] is based on phenomenological boundary conditions [11] in the diffuse case. The theory [13] has considered the proximity effect in contacts between a conventional s-wave superconductor and FeBS and concluded that this proximity effect is negative in the case of \( s_{\pm} \) pairing symmetry in FeBS, but also this theory is based on phenomenological boundary conditions [11].

The theory [14] has considered the Josephson effect in contacts with three-band superconductors, but this consideration is based on phenomenological boundary conditions [10] in the clean case. The theory [15] has considered the density of states and the Josephson current in contacts with unconventional \( s_{\pm} \) superconductors, based on phenomenological boundary conditions [11], and concluded that the Josephson current-phase relations themselves are not indicative of \( s_{\pm} \) symmetry. Our microscopic consideration will show below that this is not so. The theories of [16,17], as well as theories [3–6,8,9,12–15], do not take into account the important difference in the directions of the Josephson current relative to crystallographic axes of the superconductor [7]. The importance of the investigation of the Josephson transport in different directions relative to crystallographic axes of FeBS was emphasized in [18]; however, the current-phase dependences of the Josephson current were not presented in [18]. In addition the important case, as will be demonstrated below, of the Josephson junction with a thick insulating layer was not considered. Our theory eliminates all the above-mentioned disadvantages of the previous investigations. Based on our calculations, we demonstrate the possibility to determine the symmetry of the order parameter in FeBS.

Model for the Josephson junction with an FeBS.

- We consider a model clean planar superconducting \( S/I/S_p \) junction with perfectly flat interfaces in the tight-binding approximation like the one depicted in fig. 1. One can see a two-dimensional crystallographic plane of a conventional spin-singlet s-wave superconductor \( S \) (blue filled circles on left side of fig. 1), \( N \) atomic layers of an insulator (circles in the middle of fig. 1) and multiorbital superconductor \( S_p \) (in the right part of fig. 1). We consider the application of our method for the case in which a multiorbital superconductor \( S_p \) is FeBS.

The minimal model to reproduce Fermi surfaces in FeBS is a two-band model considering \( d_{xz} \) and \( d_{yz} \) orbitals in iron [19]. There are four hopping parameters \( t_1, t_2, t_3 \) (intraorbital hopping parameters) and \( t_4 \) (interorbital hopping parameter) in this model, as shown in fig. 1. For the pair potential, the intraorbital \( s_0 \) and \( s_{++} \) models are considered [20]. We consider the case of zero misorientation angle of the crystallographic axes of FeBS with respect to the interface as shown in fig. 1. The hopping parameter between the sites of a usual superconductor \( S \) on the left side and the sites of an insulator \( I \) is described by \( \gamma \), and the hopping parameters between the sites of an insulator \( I \) and \( d_{xz} \) (\( d_{yz} \)) orbitals on the right side are described by \( \gamma_1 \) (\( \gamma_2 \)).

For simplicity, we assume that the periods of the crystal lattices in a normal metal and FeBSs are the same and equal to \( a = 1 \). To calculate the Josephson current across a \( S/I/S_p \) junction we should construct a coherent Green’s function of the whole system. The simplest way to do it is to construct the Green’s functions of \( S, I, S_p \) and then match them at the boundaries [21]. Let us define the temperature matrix Green’s function \( G \) in the tight-binding approximation for FeBS in the framework of the two-orbital model in the following form:

\[
G_{\{n\},\{j\}}(\tau_1, \tau_2) = \begin{pmatrix}
\hat{G}_{\{n\},\{j\}}(\tau_1, \tau_2) & \hat{F}_{\{n\},\{j\}}(\tau_1, \tau_2) \\
\hat{F}_{\{n\},\{j\}}(\tau_1, \tau_2) & \hat{G}_{\{n\},\{j\}}(\tau_1, \tau_2)
\end{pmatrix},
\]

(1)

where \( \hat{G}_{\{n\},\{j\}}(\tau_1, \tau_2) \), \( \hat{F}_{\{n\},\{j\}}(\tau_1, \tau_2) \), \( \hat{G}_{\{n\},\{j\}}(\tau_1, \tau_2) \), \( \hat{F}_{\{n\},\{j\}}(\tau_1, \tau_2) \) are \( 4 \times 4 \) matrices in orbital space. The components of these matrices have the following form:

\[
\begin{align*}
G^{(a\beta)}_{\{n\},\{j\}}(\tau_1, \tau_2) &= -(T_ \tau c_{\{n\},\{j\}}^{(a)} |(n), \tau_1 c_{\{j\}}^{(\beta)} \rangle \langle \{j\}, \tau_2) , \\
F^{(a\beta)}_{\{n\},\{j\}}(\tau_1, \tau_2) &= (T_ \tau c_{\{n\},\{j\}}^{(a)} |(n), \tau_1 c_{\{j\}}^{(\beta)} \rangle \langle \{j\}, \tau_2) , \\
\hat{G}^{(a\beta)}_{\{n\},\{j\}}(\tau_1, \tau_2) &= -(T_ \tau c_{\{n\},\{j\}}^{(a)} |(n), \tau_1 c_{\{j\}}^{(\beta)} \rangle \langle \{j\}, \tau_2) , \\
\hat{F}^{(a\beta)}_{\{n\},\{j\}}(\tau_1, \tau_2) &= (T_ \tau c_{\{n\},\{j\}}^{(a)} |(n), \tau_1 c_{\{j\}}^{(\beta)} \rangle \langle \{j\}, \tau_2).
\end{align*}
\]

(2)
In eqs. (1), (2) indices \( \alpha \) and \( \beta \) run through all values 1,2, where index 1 corresponds to the \( d_{xz} \) orbital and index 2 corresponds to the \( d_{yz} \) orbital; \( t^{(1)}_{\{n\},\{l\}} \) (\( t^{(2)}_{\{n\},\{l\}} \)) is the creation operator of an electron belonging to the \( d_{xz} \) (\( d_{yz} \)) orbital with spin \( \sigma \) on the \( \{n\} = (n_x, n_y) \) site, \( \tau_i \) is an imaginary “time”, and \( T_\tau \) is an imaginary “time” ordering operator.

Green’s functions of a conventional superconductor \( G^S \) and an insulator \( G^I \) have the same form as in eqs. (1), (2), but without the upper orbital indices.

Gor’kov’s equations in the discrete case for an arbitrary model of the intraorbital superconducting pairing have the following form:

\[
\text{see eq. (3) above}
\]

In eq. (3) \( \alpha \neq \beta \), \( t^{(1)}_{\{n\},\{l\}} \) and \( t^{(2)}_{\{n\},\{l\}} \) are the hopping parameters between the same \( d_{xz} \) (\( d_{yz} \)) orbitals, and \( t^{(12)}_{\{n\},\{l\}} \) and \( t^{(21)}_{\{n\},\{l\}} \) are the hopping parameters between the different orbitals, \( \omega_m = \pi T(2m+1), m \) is the integer value, \( T \) is the temperature.

Discrete Gor’kov’s equations for the Green’s function of a conventional superconductor \( G^S \) and an insulator \( G^I \) have the same form as in eq. (3), but without the orbital indices, the third and fourth terms on the left side of eq. (3) for \( G^I \) and without the third term on the left side of eq. (3) for \( G^S \) [21]. It can be shown that in order to calculate the Josephson current in the structure under consideration it is enough to solve only eq. (3) with \( \alpha = 1 \) or \( \alpha = 2 \), because the remaining system of the equations gives the same results.

To construct the coherent Green’s function of the whole \( S/I/S_p \) junction one should match Green’s functions of \( S \), \( I \) and \( S_p \) regions at the boundaries. The boundary conditions for matching of wave functions in multiorbital metals were proposed in [2]. For temperature Green’s functions these boundary conditions for the case \( \alpha = 1, \beta = 2 \) have the following form:

\[
\begin{align*}
\left\{ \begin{array}{l}
tG^S_{\alpha \beta,j} = \gamma tG^I_{\alpha \beta,j}, \\
tF^S_{\alpha \beta,j} = \gamma tF^I_{\alpha \beta,j}, \\
\gamma G^S_{\alpha \beta,j} = t'G^I_{\alpha \beta,j}, \\
\gamma F^S_{\alpha \beta,j} = t'F^I_{\alpha \beta,j},
\end{array} \right.
\end{align*}
\]

Due to the translational invariance of the structure in the direction parallel to the interface, the \( k_y \) component of the quasimomentum is conserved and the subscripts corresponding to the coordinate of a site in this direction are omitted. We neglect the self-consistency of the pair potential at the above-outlined procedure of construction of the coherent Green’s function of the \( S/I/S_p \) junction since, as was shown in [7], it is allowed in theoretical investigation of the Josephson current in junctions with unconventional superconductors.

The Josephson current through an insulating region is given by

\[
I = \frac{eT\tau}{\hbar} \int_{\omega_m} \sum_{j,j+1} (G^I_{j,j+1} - G^I_{j+1,j}) + (G^I_{j,j+1} - G^I_{j+1,j}) dk_y. \tag{6}
\]

It can be shown that previous relations for the Josephson current in junctions with both conventional and unconventional superconductors [7] follow from eqs. (3)–(6). Equations (3)–(6) provide the possibility to calculate microscopically the Josephson current in the \( S/I/S_p \) junction for different directions of current relative to the crystallographic axes of FeBS and different symmetries of the order parameter in it.

**Numerical results.** – The phase dependences of the averaged over \( k_y \) Josephson current in the (100) oriented \( S/I/S_p \) junction (fig. 1) are depicted in fig. 2 for the case of the \( s_\pm \) symmetry of the order parameter in FeBS. In our calculations we use the following values of hopping parameters and chemical potential in FeBS: \( t_1 = -0.1051 \), \( t_2 = 0.1472 \), \( t_3 = -0.1909 \), \( t_4 = -0.0874 \) and \( \mu_y = -0.081 \) (eV), according to [22], and suppose that the \( S/I \) interface is transparent: \( \gamma = t \). We consider the \( s_\pm \) model of FeBS with momentum-dependent order parameter \( \Delta = 4\Delta_0 \cos k_x \cos k_y \) with \( \Delta_0 = 0.008 \) (eV), in a superconductor \( S \) we choose the magnitude of the isotropic
order parameter \( \Delta_0 = 0.002 \) (eV) and assume a relatively low temperature \( T/T_c \approx 0.02 \). We choose the normal excitation spectrum in \( S \) in the form of \( \epsilon_N = 2t\left(\cos k_x + \cos k_y\right) + \mu_N \) with hopping parameter \( t = -0.3 \) (eV) and chemical potential \( \mu_N = 0.05 \) (eV) in order to provide a large size of the Fermi surface in \( S \). Consequently, areas with large \( k_y \) in FeBS contribute to the Josephson current. In the insulating region we choose the normal excitation spectrum in the form of \( \epsilon_{\|} = 2t'\left(\cos k_x + \cos k_y\right) + \mu_{\|} \) with hopping parameter \( t' = -0.3 \) (eV) and chemical potential \( \mu_{\|} = 1.2 \) (eV). In all four panels (a)–(d) in fig. 2 the solid lines correspond to the atomically sharp boundary without layers of an insulator (the number of insulators layers \( N = 0 \), lines with crosses correspond to \( N = 3 \) layers of an insulator. Panels (a)–(d) in fig. 2 differ from each other by the choice of the set of the \( I/I_p \) interface hopping parameters, which determines the transparency of the \( I/I_p \) interface [2].

One can see from fig. 2 that for different sets of the \( I/I_p \) interface hopping parameters and atomically sharp \( I/I_p \) boundary \( S/I_p \) Josephson junction can achieve a ground state (which corresponds to the minimum of the free energy of the Josephson junction) at the phase difference \( \varphi = \pi \) (fig. 2(a), (d)), \( \varphi = 0 \) (fig. 2(c)) and \( \varphi = \phi_0 \), where \( 0 < \phi_0 < \pi \) (fig. 2(b)). This ground state corresponding to the \( S/I_p \) Josephson junction with atomically sharp boundary is denoted by filled circles in fig. 2. Such a variety of the current-phase dependences is explained by the sign-changing in different bands the \( s_\pm \) order parameter in FeBS and the contribution from all values of \( k_y \) to the total Josephson current in this case.

Taking into account an insulating layer in the \( S/I_p \) Josephson junction leads to the suppression of the contributions to the average current from regions with large \( k_y \); therefore the regions with small \( k_y \) dominate [23]. In this case (lines with crosses in fig. 2) the current-phase dependence becomes very close to the sinusoidal with ground state at \( \varphi = \pi \) (fig. 2(a)–(c)) and \( \varphi = 0 \) (fig. 2(d)). This ground state corresponding to the \( S/I_p \) Josephson junction with \( N = 3 \) layers of an insulator is denoted by unfilled circles in fig. 2. This situation differs from the case of \( S/I_p \) Josephson junctions with \( d \)-wave superconductor with nonzero misorientation angle, when \( \pi \)-contact survives with the increase of the length of an insulator layer [7].

Our calculations of the phase dependence of the Josephson current in the \( S/I_p \) junction with the \( s_{+} \) symmetry of the order parameter in FeBS demonstrate that in all cases this junction has a ground state at \( \varphi = 0 \).

The situation changes in the case of the investigation of the Josephson current in \( S/I_p \) junctions along the \( z \)-axis. In this direction at each fixed \( k_{\|} = (k_x, k_y) \) the contribution to the Josephson current is effectively just from one of the FeBS bands because another band is significantly far from the Fermi level. The phase dependences of the averaged over \( k_{\|} \) Josephson current along the \( z \)-axis in the \( S/I_p \) structure are depicted in fig. 3(a) and fig. 3(b) with \( S_p \), describing by \( s_\pm \) and \( s_{+} \) model of the superconducting pairing, respectively. The solid line and the left axis correspond to an atomically sharp boundary, the line with crosses and the right axis correspond to the case of an insulating layer containing \( N = 3 \) atoms. In these calculations we choose the normal excitation spectrum in \( S \) in the form of \( \epsilon_N = 2t\left(\cos k_x + \cos k_y + \cos k_z\right) + \mu_N \) with hopping parameter \( t = -0.3 \) (eV) and chemical potential \( \mu_N = 0.6 \) (eV). Such values of the hopping parameter and chemical potential provide a sufficiently large size of the Fermi surface in \( S \), so both electronic and hole packets.
contribute to the Josephson current. For FeBS along the $z$-axis we take into account only hopping $t_{ij} = -0.1$ (eV) between the same orbitals on the nearest-neighbor sites. We considered a transparent $S/I$ interface and the following values for hopping parameters across the $I/S_p$ interface: $\gamma_1 = \gamma_2 = 0.17$. Our calculations in the case of the $s_{\pm}$ model of the superconducting pairing demonstrate that for atomically sharp boundaries the contribution to the total Josephson current from electron pockets dominates and the resulting $S/I/S_p$ junction is the $\pi$-junction (solid line in fig. 3(a)). This ground state corresponding to the $S/I/S_p$ Josephson junction with atomically sharp boundary is denoted by filled circles in fig. 3(a). The presence of an insulating layer leads to the suppression of the contributions to the averaged current from regions with large $k_{ij}$, that is from the electron pockets, so the considered structure with nonzero insulating layer has a ground state at 0 phase difference (line with crosses in fig. 3(a)). This ground state corresponding to the $S/I/S_p$ Josephson junction with $N = 3$ layers of an insulator is denoted by unfilled circles in fig. 3(a). It should be noted that similar results for the Josephson tunneling in the $z$-direction have been recently obtained using a different technique [24]. In the case of the $s_{++}$ model of the superconducting pairing for atomically sharp boundary and for $N = 3$ layers of an insulator the $S/I/S_p$ Josephson junction has a ground state at zero phase difference (fig. 3(b)). The modern technology permits to create the loop of the normal superconductor, one end of which is oxidized and the other is not, to connect it with $z$-oriented FeBS and to create dc SQUID. If one observes in this experiment a $\pi$ phase shift, it will be the crucial evidence in favor of the presence of the $s_{\pm}$ symmetry in FeBS. The same experiment was suggested recently in [23]. The crucial experiment suggested in [23] is based on ideas which were previously discussed in [25]. For the case of Josephson tunneling in the $x$-$y$ plane the evidence of the implementation of the $s_{\pm}$ symmetry of the order parameter is the presence of the second harmonic with significant amplitude for the case of an atomically sharp boundary, as follows from fig. 2. It is also necessary to note the significant suppression of the magnitude of the Josephson current in the case with a long insulator layer compared to atomically sharp layers (right and left axis in fig. 3(a)). This result can be one of the explanations of the Josephson critical current suppression in the recent Josephson tunneling experiment with $z$-oriented FeBS [26-28]. Our theory also predicts the reduction of the $I_c R_{th}$ product in the case of $s_{\pm}$ symmetry in FeBS for Josephson tunneling in the $x$-$y$ plane compared to $s_{++}$ symmetry. This reduction was observed in [29].

Conclusion. – In conclusion, we have proposed a microscopic theory describing Josephson tunneling in junctions with multiband superconductors. Our theory takes into account the complex excitation spectrum of these superconductors, their multiband Fermi surface, as well as interband scattering at the boundaries. This theory has been applied to the calculation of the Josephson current-phase relations in junctions of FeBS described by $s_{\pm}$-wave and $s_{++}$-wave order parameter symmetries with a conventional superconductor for different directions of the current relative to the crystallographic axes of FeBS and different length of an insulator layer. We have demonstrated the possibility of the ultimate determination of the symmetry of the order parameter in FeBS by the investigation of the Josephson transport. The microscopic approach proposed in this paper can be a basis of the microscopic consideration of the Josephson current in topological superconductors, because many of them are multiband and multiorbital materials.

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