Fulde–Ferrell state in superconducting core/shell nanowires: role of the orbital effect

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Abstract
The orbital effect on the Fulde–Ferrell (FF) phase is investigated in superconducting core/shell nanowires subjected to the axial magnetic field. Confinement in the radial direction results in quantization of the electron motion with energies determined by the radial $j$ and orbital $m$ quantum numbers. In the external magnetic field, the twofold degeneracy with respect to the orbital magnetic quantum number $m$ is lifted which leads to the Fermi wave vector mismatch between the paired electrons, $(k, j, m, \uparrow) \leftrightarrow (-k, j, -m, \downarrow)$. This mismatch is transferred to the nonzero total momentum of the Cooper pairs, which results in a formation of the FF phase occurring sequentially with increasing magnetic field. By changing the nanowire radius $R$ and the superconducting shell thickness $d$, we discuss the role of the orbital effect in the FF phase formation in both the nanowire-like ($R/d \ll 1$) and nanofilm-like ($R/d \gg 1$) regime. We have found that the irregular pattern of the FF phase which appears for the case of the nanowire-like regime, for the nanofilm-like geometry evolves towards the regular distribution in which the FF phase stability regions emerge periodically between the BCS states. The transition between these two different phase diagrams is explained as resulting from the orbital effect and the multigap character of superconductivity in the core/shell nanowires.

Keywords: FFLO phase, superconducting nanowires, core/shell nanowires

(Some figures may appear in colour only in the online journal)

1. Introduction

Over the last decade, unconventional superconductivity with a nontrivial Cooper pairing has attracted growing interest due to fascinating superconducting properties which are not observed for the standard BCS state. Among the wide class of unconventional superconductors including high-$T_c$ cuprates [1–3], iron-pnictides [4], or heavy fermion materials [5], recently, special attention has been drawn to systems with a spatially varying energy gap [6]. The existence of such a superconducting phase with the order parameter oscillating in real space was proposed in the mid-1960s by Fulde and Ferrell [7] (FF phase) as well as independently by Larkin and Ovchinnikov (LO phase) [8]. According to their original concept, superconductivity can survive in the magnetic field substantially higher than the critical field $H_c$, due to the creation of an inhomogeneous paired state with a non-zero total momentum of the Cooper pairs $(k \uparrow, -k + q \downarrow)$. This so-called FFLO state results from the pairing between electrons from Zeeman-splitted parts of the Fermi surface.

In spite of the straightforward nature of the theoretical prediction and many ongoing theoretical investigations regarding the appearance of the FFLO state in different materials [9–13], the experimental evidence of the non-zero momentum pairing has been reported only recently in heavy fermion systems [5, 14–16] and 2D organic superconductors [17–20]. Both of these material classes are characterized by reduction of the orbital pair breaking mechanism which is a crucial physical limitation for the experimental realization of the FFLO phase. Significance of the orbital pair braking is described by the Maki parameter [21, 22] defined as $\alpha = \sqrt{2}H_{orb}^c / H_{c2}^p$, where $H_{orb}^c$ is the upper critical field calculated without Zeeman splitting and $H_{c2}^p$ is the critical field in paramagnetic limit [23, 24]. It has been established that the FFLO phase can exist at a finite temperature if $\alpha > 1.8$ [25]. This criterion can be
met in ultrathin metallic nanofilms in which confinement in the direction perpendicular to the film strongly reduces the orbital effect for the in-plane magnetic field.

The theoretical model describing the FFLO phase in metallic nanofilms, besides the possibility of the non-zero momentum pairing, should also contain the multiband character of superconductivity in these systems. In metallic nanostuctures with size comparable to the electron wavelength, the Fermi surface splits into a set of discrete subbands leading to many interesting effects which are not observed in the bulk limit, e.g. the formation of Andreev states [26] or oscillations of superconducting properties [27–36]. As reported in our recent paper [37], due to the multiband nature, the FF phase in metallic nanofilms splits into subphases, number of which corresponds to the number of subbands participating in the formation of the paired state. Similar behavior has also been reported for Pauli-limiting two-band superconductors [38]. In both of these reports the FF phase has been induced by the Zeeman effect for the magnetic field \( H > H_c \).

The multiband character of superconductivity is even more pronounced in metallic nanowires. Studies of superconductor to normal-metal transition driven by the axial magnetic field has led to discovery of a very interesting phenomenon [36]. It turns out that the magnetic field does not destroy superconductivity simultaneously in all subbands participating in the paired phase but the transition to the normal state occurs gradually. The magnetic field suppresses superconducting correlations step by step in subsequent subbands. It reveals itself as a cascade of jumps in the order parameter with increasing magnetic field. Such anomalous behavior has inspired our recent study [39]. Surprisingly, we have found that in cylindrical nanowires subjected to the axial magnetic field, the orbital effect, which so far has been regarded as detrimental to the FFLO phase formation, can in fact induce the non-zero momentum paired state. As shown in [39], the Fermi wave vector mismatch induced by the orbital effect between the subbands with opposite orbital momenta is transferred to the non-zero total momentum of the Cooper pairs which results in formation of the sequentially occurring FF and BCS phases with increasing magnetic field. In this context, understanding the physical mechanism behind the change of the phase diagrams from the Pauli-limit, in which the FF phase occurs in the vicinity of \( H_c \) as for nanofilms, to the orbital limit, in which the stable FF phases appear between the BCS-paired states for \( H < H_c \), still remains an unexplored issue. This can be done by considering superconducting core/shell nanowires, in which by the control of the ratio \( R/d \), where \( R \) is the core radius and \( d \) is the shell thickness, we can switch from the FFLO phase formation, can in fact induce the non-zero total momentum of the Cooper pairs.

between these two different phase diagrams is explained as resulting from the orbital effect and the multiband character of superconductivity in the considered nanostructures.

The paper is organized as follows. In the next section we introduce the basic concepts of the theoretical model based on the modified BCS theory, in which the superconducting gap acquires the non-zero total momentum of the Cooper pairs. We explain in detail how the angular-momentum-induced Fermi-surface splitting generates the FF phase. In section 3 we discuss our results considering the contributions of both the orbital and Zeeman effect to the FF state. Finally, section 4 is devoted to conclusions and a short discussion on the possibility of the experimental verification of the phenomena presented in the paper.

2. Theoretical method

Let us consider a core/shell nanowire consisting of a core of radius \( R \) surrounded by a superconducting shell of thickness \( d \) (figure 1(a)). Recently, analogous systems of semiconductor nanowires covered by a superconducting layer have attracted growing interest due to their potential application in topologically protected quantum computing using the Majorana zero modes [42, 43]. For simplicity, let us assume that the core is an ideal insulator and electrons cannot penetrate the region of the core which allows us to neglect the proximity effect at the superconductor/insulator interface. We start from the general form of the BCS Hamiltonian

\[
\hat{H} = \sum_{\sigma} \int d^3r \hat{\Psi}^\dagger(\mathbf{r}, \sigma) \hat{H}_0 \hat{\Psi}(\mathbf{r}, \sigma) + \int d^3r \left[ \Delta(\mathbf{r}^\uparrow) \hat{\Psi}^\dagger(\mathbf{r}, \uparrow) \hat{\Psi}(\mathbf{r}, \downarrow) + \text{h.c.} \right] + \int d^3r \frac{\left| \Delta(\mathbf{r}) \right|^2}{g},
\]

where \( \sigma \) denotes the spin state \((\uparrow, \downarrow)\), \( g \) is the phonon-mediated electron–electron coupling constant and the gap parameter in the real space is given by

\[
\Delta(\mathbf{r}) = -g \left( \hat{\Psi}(\mathbf{r}, \downarrow) \hat{\Psi}(\mathbf{r}, \uparrow) \right).
\]

Figure 1. (a) Scheme of the superconducting core/shell nanowire. (b) Schematic illustration of the FF pairing in the nanowire. In the presence of the magnetic field the two-fold degeneracy with respect to the orbital magnetic quantum number \( m \) and the two-fold degeneracy with respect to the spin \( \sigma \) are lifted. The Fermi vector mismatch is compensated by the non-zero center-of-mass momentum of the Cooper pairs.
Choosing the gauge for the vector potential as $\mathbf{A} = (0, eHr/2, 0)$, where the magnetic field $H$ is directed along the nanowire axis, the single-electron Hamiltonian $H_0$ in the cylindrical coordinates $(r, \varphi, z)$ is given by

$$H_0 = \frac{\hbar^2}{2m_e} \left[ -\frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial}{\partial r} \right) + \frac{m^2}{r^2} + \frac{\hbar^2 k^2}{2m_e} + (m + \sigma) \mu_B H - \mu \right],$$

(3)

where $\sigma = \pm 1$ for spin-up and spin-down electrons, $\mu$ is the chemical potential and $e, m_e$ is the electron charge and mass, respectively.

If we assume the azimuthal invariance and neglect the diamagnetic term $\sim A^2$, whose energy for nanowires is one order of the magnitude lower than the order parameter, $H_0$ can be reduced to the 1D form

$$H_{0,1D} = \frac{\hbar^2}{2m_e} \left[ -\frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial}{\partial r} \right) + \frac{m^2}{r^2} + \frac{\hbar^2 k^2}{2m_e} + (m + \sigma) \mu_B H - \mu \right],$$

(4)

with the corresponding single-electron wave functions

$$\psi_{j,m}(r, \varphi, z) = \frac{1}{\sqrt{2\pi L}} \phi_{j,m}(r) e^{im\varphi} e^{ikz},$$

(5)

where $L$ is the nanowire length, $j$ is the radial quantum number, $m$ is the orbital magnetic quantum number and $k$ is the wave vector along the nanowire axis $z$. By assuming the hard-wall boundary conditions in the shell, $\phi_{j,m}(R) = \phi_{j,m}(R + d) = 0$, the radial wave function $\phi_{jm}(r)$ can be written as [41]

$$\phi_{jm}(r) = \sqrt{\frac{2}{\sqrt{J_m}} J_m(\chi_m R) J_m(\chi_m r) - J_m(\chi_m R) Y_m(\chi_m r)},$$

(6)

where $J_m(r)$ and $Y_m(r)$ are Bessel functions of the first and second kind of $m$th order and $J$ is the normalization constant. The parameter $\chi_m$, related to the single-electron energy $\xi_{j,m,\sigma}$ by

$$\xi_{j,m,\sigma} = \frac{\hbar^2}{2m_e} (\chi_m^2 + k^2) + (m + \sigma) \mu_B H - \mu,$$

(7)

is a solution of the equation

$$J_m(\chi_m R) J_m(\chi_m (R + d)) - J_m(\chi_m (R + d)) Y_m(\chi_m R) = 0.$$  

(8)

From equation (7) we can see that for $H = 0$, each single-electron state is fourfold degenerate (two-fold degeneracy with respect to the orbital magnetic quantum number $m$ and two-fold degeneracy with respect to the spin $\sigma$). In the presence of the external magnetic field both these degeneracies are lifted resulting in a shift between the subbands corresponding to $m$ and $-m$ as well as $\uparrow$ and $\downarrow$. Since in the superconducting state the pairing appears between particles with opposite spins, momenta and orbital momenta: $(k, j, m, \uparrow) \leftrightarrow (-k, j, -m, \downarrow)$, the Fermi-wave vector mismatch induced in the magnetic field can be transferred into the non-zero momentum of the Cooper pairs ($q \neq 0$ along the $z$ axis) giving rise to the FF phase. Schematic illustration of this process is sketched in figure 1(b). Using the field operators in the form

$$\hat{\Psi}(r, \varphi, z, \sigma) = \sum_{k,j,m} \psi_{k,j,m}(r, \varphi, z) \hat{c}_{k,j,m,\sigma},$$

$$\hat{\Psi}^\dagger(r, \varphi, z, \sigma) = \sum_{k,j,m} \psi_{k,j,m}^*(r, \varphi, z) \hat{c}_{k,j,m,\sigma}^\dagger,$$

(9)

where $\hat{c}_{k,j,m,\sigma}(\hat{c}_{k,j,m,\sigma}^\dagger)$ is the annihilation (creation) operator, the BCS Hamiltonian with the possibility of non-zero momentum pairing is given by

$$H_q = \sum_{k,j,m,n} \hat{f}_{k,j,m,n} \hat{c}_{k,j,m,n} \hat{c}_{-k,j,-m,n}^\dagger + \sum_{k,j,m} \xi_{k,q,j,m,n}^\dagger \hat{c}_{k,j,m,n}^\dagger \hat{c}_{-k,j,-m,n} + \sum_{j,m} \frac{\Delta_{j,m,n}^2}{g^2},$$

(10)

where $\hat{f}_{k,j,m,n} = (\hat{c}_{k,j,m,\uparrow}^\dagger \hat{c}_{-k,j,-m,\downarrow} + \hat{c}_{k,j,-m,\downarrow}^\dagger \hat{c}_{-k,j,m,\uparrow})$ is the composite vector operator and

$$\hat{H}_{k,j,m,n} = \begin{pmatrix} \xi_{k,j,m,\sigma} & \Delta_{j,m,n} \\ \Delta_{j,m,n}^\dagger & -\xi_{k,j,-m,\sigma} \end{pmatrix},$$

(11)

In the above equation, for simplicity, we limit ourselves to the situation in which all the Cooper pairs have the momentum $q$. This assumption corresponds to the FF phase. In equation (11), $\Delta_{j,m,n}$ is the superconducting energy gap in the subband $(j, m)$ defined as

$$\Delta_{j,m,n} = \frac{g}{4\pi^2} \sum_{k,j,m} C_{j,m,n} \langle \hat{c}_{-k,q,j,-m,n} \hat{c}_{k,j,m} \rangle,$$

(12)

with the interaction matrix

$$C_{j,m,n} = \int_R \right R \left( r \phi_{j,m}(r) \partial r \phi_{j,m}^*(r) \right).$$

(13)

Hamiltonian (10) can be diagonalized by the Bogoliubov–de Gennes transformation

$$\begin{pmatrix} \hat{c}_{k,j,m,\uparrow} \\ \hat{c}_{-k,j,-m,\downarrow} \end{pmatrix} = \begin{pmatrix} U_{k,j,m,n} & V_{k,j,m,n} \\ -V_{k,j,m,n} & U_{k,j,m,n} \end{pmatrix} \begin{pmatrix} \hat{c}_{k,j,m}^\dagger \\ \hat{c}_{-k,j,-m} \end{pmatrix},$$

(14)

where

$$U_{k,j,m,n} = \frac{1}{2} \left( 1 + \frac{\xi_{k,j,m,\sigma} + \xi_{-k,j,-m,\sigma}}{\sqrt{(\xi_{k,j,m,\sigma} + \xi_{-k,j,-m,\sigma})^2 + 4\Delta_{j,m,n}^2}} \right),$$

$$V_{k,j,m,n} = \frac{1}{2} \left( 1 - \frac{\xi_{k,j,m,\sigma} + \xi_{-k,j,-m,\sigma}}{\sqrt{(\xi_{k,j,m,\sigma} + \xi_{-k,j,-m,\sigma})^2 + 4\Delta_{j,m,n}^2}} \right),$$

(15)

are the Bogoliubov coherence factors. As a result, one obtains the following form of the quasiparticle energies

$$E_{k,j,m,n} = \frac{1}{2} \left( \xi_{k,j,m,\sigma} + \xi_{-k,j,-m,\sigma} \right) \pm \sqrt{\frac{1}{4} (\xi_{k,j,m,\sigma} + \xi_{-k,j,-m,\sigma})^2 + \Delta_{j,m,n}^2 + (m + \sigma) \mu_B H},$$

(16)

By substituting equation (14) into equation (12) we derive the self-consistent equations for the superconducting gaps
\[ \Delta_{j,m} = \frac{g}{4\pi^2} \int \frac{d\ell}{\sqrt{\xi_{j,m}^2 + \xi_{j+1,m}^2 + 4\Delta_{j,m}^2}}. \]

where \( f(E) \) is the Fermi-Dirac distribution. The summation in equation (17) is carried out only over the single-energy states \( \xi_{j,m} \) inside the Debye window \( \xi_{j,m} < 4\hbar\omega_D \), where \( \omega_D \) is the Debye frequency. Since the chemical potential in nanostructures strongly deviates from that assumed in the bulk, for each shell thickness we determine \( \mu \) assuming constant electron concentration,

\[
n_{e} = \frac{1}{\pi^2[(R+d)^2 - R^2]} \int d\ell \sum_{j,m} \int_{R}^{R+d} dr \, r \times \left\{ |U_{j,m}(r)|^2 f(E_{j,m}^+) + |V_{j,m}(r)|^2 [1 - f(E_{j,m}^-)] \right\}. \tag{18} \]

In the considered nanowires, the spatial dependence of the superconducting gap results not only from the creation of the FF phase \( \Delta(r, \varphi, z) = \Delta(r, \varphi) e^{i\varphi z} \) but it is also induced by the quantum confinement. The spatial dependence of the order parameter in the radial direction can be expressed as

\[
\Delta_{r}(r) = \frac{g}{4\pi^2} \int \frac{d\ell}{\sqrt{\xi_{j,m}^2 + \xi_{j+1,m}^2 + 4\Delta_{j,m}^2}} \times \left\{ |U_{j,m}(r)|^2 f(E_{j,m}^+) - |V_{j,m}(r)|^2 [1 - f(E_{j,m}^-)] \right\}. \tag{19} \]

To obtain the phase diagram, the superconducting gaps \( \Delta_{j,m} \) and the chemical potential \( \mu \) are calculated by solving equations (17) and (18) self-consistently. The wave-vector \( q \) is determined by minimizing the free energy of the system [44].

Calculations presented in the paper have been carried out for the material parameters typical of aluminum: \( \hbar\omega_D = 32.31 \text{ meV}, gN(0) = 0.18 \), where \( N(0) = mk_b/\pi^2\hbar^2 \) is the bulk density of states at the Fermi level, \( \Delta_{\text{bulk}} = 0.25 \text{ meV} \) and the chemical potential \( \mu_{\text{bulk}} = 0.9 \text{ eV} \) which corresponds to the electron density \( n_e = 3.88 \times 10^{21} \text{ cm}^{-3} \). The assumed low value of the chemical potential, relative to that measured in the bulk, results from the parabolic band approximation (for more details, see [35]). Its value has been determined to obtain good agreement with the experimental data reported in [45].

The self-consistent procedure has been carried out for the constant electron concentration which implies a gradual increase of the chemical potential with decreasing shell thickness. Moreover, we do not include a thickness-dependent change in the electron-phonon coupling [46], as it can only result in quantitative effects and do not alter the qualitative picture of the FF phase creation presented in the paper.

### 3. Results and discussion

#### 3.1. Quantum-size oscillations of the superconducting order parameter

To determine the geometrical parameters appropriate for the analysis of the non-zero total momentum pairing, we have calculated the spatially averaged superconducting order parameter \( \Delta \), defined as

\[ \Delta = \frac{2}{d(2R + d)} \int_{R}^{R+d} d\ell \, r \Delta(r), \tag{20} \]

as a function of the shell thickness for different core radii (figure 2).

The \( \Delta(d) \) oscillations presented in figure 2 are due to the quantum size effect which arises when the system size becomes comparable to the electron Fermi wave length [35]. In the core/shell nanowires, reduction of the electron motion in the radial direction implies the quantization of the electronic states with energies determined by the quantum numbers \( j, m \) and \( k \) (equation (7)). Subsequent peaks in \( \Delta(d) \) correspond to the subsequent subbands \( (j, m) \) passing through the Fermi level while increasing the shell thickness. When the bottom of a subband is getting close to the Fermi energy, the density of states at the Fermi level rapidly increases. Since the Cooper-pair condensation concerns electrons from the energy window \( [\mu - 4\hbar\omega_D, \mu + 4\hbar\omega_D] \), each time this condition is satisfied we observe a peak in \( \Delta(d) \).

As seen, the \( \Delta(d) \) oscillations presented in figures 2(a)–(d) differ significantly from each other. The irregular oscillations for \( R = 1 \text{ nm} \) (figure 2(a)), reminiscent of these predicted for superconducting nanowires [35], evolve with increasing \( R \) towards the regular oscillations characteristic for superconducting nanofilms (figure 2(d) [47]). The crossover from the irregular pattern to the regular regime was explained in details in [35]. It is related to the centrifugal term, \( \hbar^2 m^2/2m_r r^2 \), which for \( R/d \leq 1 \) contributes significantly to the single electron energy leading to the energetically well-separated states for different \( m \). The irregular oscillations of \( \Delta(d) \) presented in figure 2(a) reflect the irregular distribution of states \( (j, m) \).
Consequently, the sequence of resonances exhibits a more regular pattern characteristic for nanofilms. Therefore, by the appropriate choice of the geometrical phase in both of the considered regimes.

Now we discuss in detail the phase diagram for $d = 1.11$ nm presented in figure 3(a). For the chosen resonant thickness, the full spectrum of $q$ in the whole range of the magnetic field is plotted in figure 4(a). For completeness, in figures 5(a) and (e) we present the quasiparticle dispersions $E_{kj}$ versus $k$ and the superconducting order parameter $\Delta(r)$ for $H = 0$.

As one can see, for the chosen geometrical parameters, there are twenty relevant subbands participating in the superconducting state: $(0,0) - (0,\pm 8)$ and $(1,0) - (1,\pm 2)$. Their contributions $P_{jm}(r)$ to $\Delta(r, H = 0)$ are displayed in figure 5(c), where

$$P_{jm}(r) = \frac{g}{4\pi^2} \int dk |\phi_{jm}(r)|^2 \frac{\Delta_{jm,q}}{\sqrt{(\xi_{k,j,m,q} + \xi_{k-\omega,j,-m,q})^2 + 4\Delta_{jm,q}^2}} \times \left[ 1 - f(E_{kj,m,q} - f(E_{kj,m,q})) \right].$$

As presented, the contributions of the individual subbands to the superconducting order parameter, $P_{jm}(r)$, vary significantly. Due to the enhanced density of states, they are the

**Figure 3.** Magnetic field dependence of the averaged superconducting gap $\Delta(H)$ for the shell thicknesses $d$ denoted by the red squares in figure 2(a) (see also insets). Gray areas represent the FF phase stability regions between which the conventional BCS phase, displayed as white regions, is stable. In panel (a), the values of the magnetic field corresponding to depairing in subsequent subbands $(j, m)$ are marked by the arrows.

**Figure 4.** (a) Total Cooper-pair momentum $q$ which minimizes the free energy as a function of the magnetic field $H$ together with (b) an exemplary difference between free energy in the superconducting and normal state $(F_{sc} - F_{n})(q)$. Minimum of $(F_{sc} - F_{n})(q)$ corresponds to the stable FF phase.

The phase diagrams presented in figure 3 exhibit the sandwich structure where the FF phase stability regions alternate with the conventional BCS states. This behavior can be explained as follows. Each time the magnetic field $H$ becomes slightly higher than $H_{c}^{jm}$, the Fermi wave-vector mismatch between the paired electrons $(k,j,m,\uparrow) \leftrightarrow (-k,j,-m,\downarrow)$ is partially compensated by the non-zero total momentum of the Cooper pairs, to sustain superconductivity in the subband $(j,m)$. As shown in figure 4(b), the formation of the FF phase minimizes the free energy of the system. At this point, we should note that in the presence of the magnetic field the Fermi vector mismatch for each of the subbands $(j,m)$ is different, which means that each of them has its own favorable total momentum $q_{jm}$. However, the situation where several values of $q_{jm}$ appear in the system is impossible due to the coupling between all the branches participating in the superconducting state (see equation (17)). Hence, the value of $q$ which minimizes the free energy is usually a result of the Cooper pair breaking processes occurring in several subbands and we can not distinguish between individual contributions to the total momentum $q$ coming from each of them. Note that the further increase of $H$, well above $H_{c}^{jm}$, causes that the paired state with the non-zero total momentum becomes energetically less favorable and the standard BCS pairing is restored.

Now we discuss in detail the phase diagram for $d = 1.11$ nm presented in figure 3(a). For the chosen resonant thickness, the full spectrum of $q$ in the whole range of the magnetic field is plotted in figure 4(a). For completeness, in figures 5(a) and (e) we present the quasiparticle dispersions $E_{kj}$ versus $k$ and the superconducting order parameter $\Delta(r)$ for $H = 0$. As one can see, for the chosen geometrical parameters, there are twenty relevant subbands participating in the superconducting state: $(0,0) - (0,\pm 8)$ and $(1,0) - (1,\pm 2)$. Their contributions $P_{jm}(r)$ to $\Delta(r, H = 0)$ are displayed in figure 5(c), where

$$P_{jm}(r) = \frac{g}{4\pi^2} \int dk |\phi_{jm}(r)|^2 \frac{\Delta_{jm,q}}{\sqrt{(\xi_{k,j,m,q} + \xi_{k-\omega,j,-m,q})^2 + 4\Delta_{jm,q}^2}} \times \left[ 1 - f(E_{kj,m,q} - f(E_{kj,m,q})) \right].$$

As presented, the contributions of the individual subbands to the superconducting order parameter, $P_{jm}(r)$, vary significantly. Due to the enhanced density of states, they are the
Figure 5. (a) and (b) Quasiparticle dispersions $E_{jm}$ versus $k$, (c) and (d) contributions to the paired phase coming from different subbands $P_{jm}$ and (e) and (f) the position dependent superconducting order parameter $\Delta(r)$. Results for the resonant shell thickness $d = 1.11$ nm (left panels) and the non-resonant shell thickness $d = 1.39$ nm (right panels), for $H = 0$. In panels ((a)-(d)) lines plotted in color correspond to the subbands with the major contribution to the superconducting state.

largest for subbands situated in the vicinity of the Fermi surface. For $d = 1.11$ nm, the major contribution to $\Delta(r)$ comes from the states $(0, \pm 8)$ and $(1, \pm 2)$. With increasing magnetic field the superconducting correlations are suppressed successively in the individual subbands. For the subband $(j, m)$, the critical field $H_{c}^{jm}$ depends not only on the orbital magnetic quantum number $m$ but also on the energy gap of excitation $\Delta_{jm}$. The latter is considerably affected by the quantum confinement and the Andreev mechanism, which appears due to the spatial variation of the superconducting order parameter [41]. One should note that in the nanowire-like regime, $\Delta_{jm}$ may be different for different quantum numbers leading to the multigap superconductivity. Therefore, the condition $H_{c}^{jm} < H_{c}^{m}$ for $|m_1| > |m_2|$ do not have to be satisfied. This agrees with our numerical results showing that $H_{c}^{2,0} > H_{c}^{0,2}$ (see figure 3(a)). Consequently, the subband $(0, 7)$ is the first one in which the superconducting phase is destroyed as the magnetic field increases. The Cooper pair breaking in this single branch entails the formation of the FF phase with the total momentum $q$ which increases with increasing magnetic field (figure 4(a)). This FF phase region, shown in figure 3(a) as the gray area, is stable up to the magnetic field value at which the Cooper pair breaking takes place in the next two states $(0, 6)$ and $(0, 8)$. Their critical magnetic fields $H_{c}^{6,0}$ and $H_{c}^{8,0}$ are almost equal leading to the substantial jump in $\Delta(H)$.

Preservation of superconductivity in these branches requires to adjust a new value of the Cooper pair momentum $q$ which is shown as the sharp dip in $q(H)$ (figure 4(a)), after which $q$ starts to increase again up to the magnetic field value at which the ordinary BCS phase is restored.

From figure 3(a) we can see that all of the FF phase stability regions are extended over the magnetic field range in which the superconductivity is destroyed in several consecutive subbands. The widest one, starting with depairing in the subbands $(0, -4)$ and $(0, 2)$ extends up to $H = 11.5$ T which is the critical field for the states: $(0, -3), (0, 1), (1, 1)$ and $(1, 2)$. The Cooper pair breaking occurring simultaneously in four subbands is accompanied by the highest jump in $\Delta(H)$ which is largely caused by the fact that $(1, 2)$ is the resonant state with the highest contribution to the superconducting order parameter (see figure 5(c)). As shown in figure 4(a), the formation of the FF phase for this particular case requires to adjust the Cooper pair momentum $q$ which is almost four times greater than that observed in other FF stability regions. Its high value is mainly determined by the Fermi wave vector mismatch in the resonant subband $(1, 2)$. The last FF phase stability region presented in figure 3(a) is related to the onset of depairing in the states $(0, -2)$ and $(0, 0)$. Note that the orbital effect does not exist for states with $m = 0$ and so the FF phase related to depairing in the subband $(0, 0)$ is solely induced by the Zeeman effect. For this reason, the corresponding Cooper pair momentum $q$ is twice smaller than in the regions with the dominant role of the orbital effect (figure 4(a)).

Calculations carried out for different values of $d$ show that the similar phase diagram, in which the FF phase stability regions are sandwiched between the standard BCS state stability ranges, is characteristic for each resonant thickness. As an example, in figure 3(b) we present $\Delta(H)$ for the neighboring resonant point $d = 1.19$ nm. Interestingly, the FF phase is not formed for the non-resonant thickness, $d = 1.39$ nm (figure 3(c)), when the superconductor to normal-metal transition has a more BCS-like character without noticeable jumps in $\Delta(H)$. For $d = 1.39$ nm, the spatially averaged value of the superconducting order parameter $\Delta = 0.142$ meV $< \Delta_{bulk}$ (figure 5(f)). All subbands are far away from the Fermi level having almost equal contributions to the superconducting state. As a result, depairing in the individual subbands is less energy-consuming and consequently the formation of the FF phases is unfavorable.

3.3. Nanofilm-like regime, $R/d \gg 1$

Now, let us discuss how the phase diagrams change if we increase the nanowire radius $R$ up to the nanofilm-like regime, where $R/d \gg 1$. In figure 6 (right panels) we present the magnetic field dependence of the spatially averaged superconducting gap $\Delta(H)$ for the resonant shell thickness $d = 1.09$ nm (see figure 2) and nanowire radii (a) $R = 4$ nm, (c) $R = 8$ nm and (e) $R = 15$ nm. As previously, the FF phase stability regions are displayed as gray areas while the corresponding values of the Cooper pair momentum $q$ are plotted in the left panels (b), (d), (f)).
for any two states with

\[ \Delta \] for the resonant shell thicknesses \( d = 1.09 \text{ nm} \) and the nanowire radii \((a)\) and \((b)\) \( R = 4 \text{ nm} \), \((c)\) and \((d)\) \( R = 8 \text{ nm} \) and \((e)\) and \((f)\) \( R = 15 \text{ nm} \). In the right panels, the gray areas represent the FF states between which the conventional BCS phases, displayed as the white regions, are stable.

The phase diagrams in figure 6 differ considerably from those calculated for the nanowire-like regime, for \( R = 1 \text{ nm} \) (figure 3). The irregular pattern of the FF phase occurrence from figure 3 evolves towards the regular distribution, in which the FF phases appear periodically between the BCS state stability ranges. As aforementioned, with increasing nanowire radius the centrifugal term of the single electron energy is suppressed, which in turn leads to the formation of bundles of subbands with the same radial quantum number \( j \) and different \( m \). The number of subbands \( N \) which take part in the superconducting phase increases significantly. For \( R = 4 \text{ nm} \), shown in figure 6(a), \( N = 58 \) and the subbands \((0, 0) - (0, \pm 23)\) as well as \((1, 0) - (1, \pm 5)\) make a contribution to the paired state (see dispersion \( E_{km} \) versus \( k \) in figure 7(a)). Such a large number of states causes the contribution of an individual subband less significant. Consequently, the magnetically-induced depairing in a single subband is not so energy-consuming and it is not accompanied with the jump in \( \Delta(H) \) as in the nanowire-like regime. Contrarily, as show in figure 6, \( \Delta \) decreases rather smoothly with increasing magnetic field up to \( H_c \), at which the superconductor to normal-metal transition is of the first order. In figure 6, the regions of the smooth decrease of \( \Delta(H) \) are divided into FF phases which appear quasi-periodically alternating with the ordinary BCS paired state stability ranges. This periodicity is the more noticeable, the closer to the nanofilm-like regime we approach—compare figures 6(a) and (e). In the intermediate regime, for \( R = 4 \text{ nm} \) and \( R = 8 \text{ nm} \), the quasi-periodic pattern is disturbed in the vicinity of the small jump in \( \Delta(H) \), where the corresponding total momentum of the Cooper pair \( q \) shows the distinct peak—see figure 6(b). The reason for this is the simultaneous Cooper pair breaking in three subbands which requires to adjust the Cooper pair momentum \( q \) which is almost six times larger than those obtained in the other FF stability regions.

More detailed analysis of the FF phase formation in the nanofilm-like regime will be given based on figures 6(e) and (f) for \( R = 15 \text{ nm} \), where \( R/d \gg 1 \). The regular occurrence of the FF phases presented in figure 6(f) can be explained based on the same arguments as used in the nanowire-like regime. Each of the FF stability regions is due to the Cooper pair breaking in the individual subbands while increasing magnetic field. Since in the nanofilme-like regime the values of \( \Delta_{j,m}(H = 0) \) are equal for each of the subbands \((j, m)\) (in contrast to the nanowire-like regime), \( H_c^{j,m} = H_c^{j,m-2} \) and \( H_c^{j,m} > H_c^{j,m-2} \) for any two states with \( |m_1| < |m_2| \). It means that the Cooper pair breaking starts from the states \((0, M)\) and \((0, -M - 2)\), where \( M \) is the highest positive orbital magnetic quantum number, and subsequently it takes place in the subbands \((j, m)\) and \((j, -m - 2)\) with \( m = M, M - 1, \ldots, 0 \).
It is of interest that, regardless of $N$, the first FF phase region is derived by the Cooper pair breaking in the subbands $(0,38)$ and $(0,−40)$. The FF phases corresponding to depairing in the states with higher $|m|$ do not occur. As an example, for $R=15$ nm, $N=200$ (see figure 7(e)) and although depairing in the states with high $|m|$ is observed at $H \approx 0.34$ T, at which $\Delta$ starts decreasing (see figure 6(e)), the first FF phase occurs for $H \approx 0.47$ T, where the Cooper pair breaking takes place in the subbands $(0,38)$ and $(0,−40)$. It explains the gradual shift of the FF phase stability regions towards $H_c$ for larger $R$. We expect that in the limit $R/d \to \infty$, all these regions will move to the close vicinity of $H_c$ and, due to the induced degeneration with respect to $m$, they will merge into one wide region. This picture is consistent with the ordinary FF phase diagrams predicted for nanofilms [5].

In figure 6(e) we can also observe that the subsequent FF phase regions become narrower with decreasing magnetic field up to the value $H_0$, below which the FF phases do not occur. Simultaneously, the corresponding value of $q$ tends to zero for $H = H_0$ (see figure 6(f)). This characteristic behavior can be explained as resulting from the difference in the orbital energy acquired from the magnetic field by the states with different orbital magnetic quantum numbers. Since the orbital term $m_j q_j H$ is proportional to the quantum number $m$, for a given magnetic field $H$, the Fermi wave vector mismatch $q_{j,m}$ between the paired electrons $(k,j,m,\uparrow) \leftrightarrow (-k,j,−m,\downarrow)$ is larger for states with higher $|m|$.

It means that if the Cooper pairs are broken in the state with high $|m|$, the wave vector mismatch $q_{j,m}$ in states with low $|m|$ is still very small. Since the value of $q$ is a result of the Fermi vector mismatches $q_{j,m}$ in all states contributing to the superconductivity, the formation of a phase with nonzero $q$ is energetically unfavorable. The critical point is depairing in the subbands $(0,38)$ and $(0,−40)$, when the Fermi vector mismatches in all superconducting states become sufficiently large to gain the small value of $q$ by all of the subbands. The FF phase with such a small value of $q$ is very susceptible to the magnetic field and even a slight increase of $H$ destroys this phase and the system switches back to the BCS state—note that the first FF phase stability region corresponding to depairing in the states $(0,38)$ and $(0,−40)$ is extremely narrow. The same behavior is repeated each time when the Cooper pairs are broken in subsequent subbands while increasing magnetic field. However, for the states with lower $|m|$, the Fermi wave vector mismatches become larger. Consequently, the reduction of the so-called depairing region on the Fermi sphere requires a larger momentum vector $q$, as shown in figure 6(f). Since the larger $q$ requires the higher magnetic field needed to destroy the FF phase, we observe the gradual extension of the FF phase stability regions for higher magnetic field.

4. Conclusions and outlook

The orbital effect on the Fulde–Ferrell phase has been investigated in the superconducting core/shell nanowires subjected to the axial magnetic field. The energy quantization induced by the confinement of the electron motion in the radial direction leads to the multiband superconductivity, similar to that found in novel superconductors, e.g. MgB$_2$. It results in the quantum-size oscillations of the superconducting order parameter. The character of $\Delta$ variations changes considerably with increasing $R$. From irregular pattern typical for nanowires, it evolves to the regular oscillations characteristic for nanofilms. As discussed above, in the superconducting core/shell nanowires, the orbital effect which so far has been considered as detrimental to the FF phase formation, can in fact induce the non-zero momentum paired state. In the presence of the magnetic field, the degeneracy with respect to the orbital magnetic quantum number $m$ is lifted which leads to the Fermi wave vector mismatch between the subbands with opposite orbital momenta in the paired state. Therefore, as the magnetic field increases, the superconductivity is destroyed in the subsequent subbands which manifests itself as a cascade of jumps in $\Delta(H)$. To sustain the Cooper pairing $(k,j,m,\uparrow) \leftrightarrow (-k,j,−m,\downarrow)$ in the corresponding subband, the non-zero total momentum state (FF phase) is formed, which in turn leads to the phase diagram of alternating FF and BCS stability regions.

In the present paper, by controlling the ratio $R/d$ we have switched from the nanowire-like $(R/d \ll 1)$ to the nanofilm-like $(R/d \gg 1)$ scenario, strengthening or suppressing the centrifugal energy, respectively. We have found that the phase diagrams differ considerably in both regimes. The irregular pattern of the FF phase occurrence in the nanowire-like regime evolves towards the regular distribution, in which the FF phase stability regions appear periodically between the BCS states, in the nanofilm-like regime. As we have presented, the crossover between these two different phase diagrams can be explained as resulting from the orbital effect and the multigap character of superconductivity. In the nanowire-like regime the centrifugal term, $\hbar^2 m^2/2m_e r^2$, contributes significantly to the single electron energy, which leads to the well separated states for different $|m|$. Due to the quantum confinement and the Andreev mechanism, induced by the spatially dependent superconducting order parameter, the system exhibits multigap superconductivity, in which $\Delta_{jm}$ may vary for different quantum numbers. The critical magnetic field for the subband $(j,m)$ is given by $H_{c}^{(m)} = \Delta_{jm}/(|m| + 1)\mu_B$. We have shown that the Cooper pair breaking in each of the subbands entails the formation of the FF phase. Therefore, due to the multigap character and the irregular position of the states with different $m$ on the energy scale, the FF phase occurrence shows irregular pattern. This picture changes considerably if we increase $R$ up to the limit of $R/d \gg 1$. In the nanofilm-like regime the centrifugal term is negligibly small and the states with different $|m|$ are almost degenerate for $H = 0$, forming the bands labeled by the radial quantum number $j$. The multigap character of the superconductivity vanishes, i.e. all subbands $(j,m)$ have the same value of $\Delta_{jm}$. Consequently, the FF phases start to occur quasi-periodically and each of the FF stability regions is related to the Cooper pair breaking in subsequent subbands with decreasing $|m| = M, M−1, \ldots$. The main findings of the paper can be summarized as follows:
(i) In the core/shell nanowires, the non-zero momentum paired state can be induced by the orbital effect.
(ii) The phase diagrams have the form of alternating FF and BCS stability regions and strongly depend on the geometrical parameters.
(iii) While increasing the ratio \( R/d \), the irregular pattern of the FF phase occurrence in the nanowire-like regime \( (R/d \leq 1) \) evolves towards the regular distribution, in which the FF phase stability regions appear periodically between the BCS states \( (R/d \gg 1) \).
(iv) The crossover between both of the phase diagram regimes is determined by the interplay between the orbital effect and the multigap character of superconductivity.

The present paper is the first synthetic analysis of the orbital effect on the FFLO phase in low dimensional superconductors. In contrary to the previous papers [5, 48], which were usually restricted to the single-band model and studied either nanowires [48] or nanofilms [5], in the present paper we take into account the multiband character of superconductivity induced by the quantum size effect. Moreover, the proposed model gives us the possibility of continuous transition from the the quasi-1D to the quasi-2D regimes. Surprisingly, it turns out that the orbital effect, so far considered as detrimental to the FFLO phase [25], can in fact generate the non-zero momentum paired state. It leads to the characteristic phase diagrams not observed in the previous single-band models, in which the FF phases alternate with the BCS state stability regions. Interestingly, in the limit \( R/d \rightarrow \infty \), our results are consistent with the phase diagrams calculated for nanofilms for which a single FF phase stability region is observed above \( H_c^2 \) and exhibits the complex structure with subphases corresponding to the different momentum of the Cooper pairs [37, 38].

The last remark in this paragraph should be concentrated on the possible experimental signature of the FF phase appearance. We expect that by carrying out the specific heat measurements one could detect the transitions between the FF and BCS phases which appear with the increasing magnetic field. The fact that the existence of the non-zero momentum of the Cooper pairs corresponds to the supercurrent along the nanowire axis can be also used as a signal in the experiential detection of the proposed effect. Moreover, since the existence of the FF phase usually increases the critical field above the paramagnetic limit, we suggest that its appearance can be indirectly detected from the study of the anisotropy of the upper critical field.

Although the proposed experimental methods give a realistic chance to confirm all findings of the paper, the serious limitation in the experimental studies of the FF phase in metallic nanostructures is the appearance of impurities since the FF state can be readily destroyed by scattering. Even in ultra-clean nanowires, the surface scattering can destroy the FF state. However, as discussed above, the phase diagram of the alternating FF and BCS stability regions emerges only for the resonant thicknesses when the superconducting order parameter is controlled by the single-electron subbands whose bottom is situated in the vicinity of the Fermi level. Its characteristic location on the energy scale causes that the corresponding longitudinal wave vector \( k < 0.5 \text{ nm}^{-1} \). It corresponds to the electron wave length grater than 50 nm. Propagation of such long waves should be insensitive to the local surface imperfection with the size of the unit cell. This argument is even stronger if we consider the wave length corresponding to the FF phase for which the total momentum of the Cooper pairs \( q < 0.01 \text{ nm}^{-1} \) gives the wave length greater than 600 nm.

Special attention should be also paid to the magnetic field which should be applied very precisely along the nanowire axis. Otherwise the small by finite perpendicular field can change the presented phase diagrams.

Some additional remarks are also needed in the context of the theoretical model and its assumptions. First, for the sake of simplicity, in this work we consider the FF phase which assumes the single Cooper pair momentum \( q \) for all subbands contributing to the superconducting state. Since the magneto-locally-induced Fermi wave vector mismatch is different for different subbands, due to the orbital effect, it would be interesting to study the case when the superconducting order parameter is a combination of many components with different \( q \) vectors. In the simplest form, it could be the FFLO state being the combination of the Cooper pairs with a total momentum \( q \) and \( -q \). In this manner, we would be able to reduce the so-called depairing region on the Fermi sphere to a greater extent, which in effect would minimize the free energy of the system to the value much lower than that obtained for the FF phase. In fact, this energetically more favorable multimomentum Cooper pair state could be observed in experiment but its appearance does not alter the qualitative picture of the non-zero momentum phase creation presented in the paper.

Finally, we would also like to address fluctuations which may appear in superconducting low dimensional structures. Thermally activated phase slip and quantum phase slip are known to play a serious role in superconducting nanowires making the use of the mean field theory questionable. However, as shown by recent experimental studies of the superconducting Pb nanofilms, the use of the mean-field theory seems to be justified as it gives surprisingly good agreement with experiment for the nanofilm thickness down to 2–5 monolayers [29]. For the superconducting nanowires, the predicted diameter limit is 5–8 nm [49], below which the quantum-phase slip can suppress the superconductivity. However, recent experiments for nanowires with diameter 5–6 nm [50] do not show any signature of the phase fluctuations. Thus, we may expect that the mean-field approach used in the paper is reasonable for the considered geometry of the core/shell nanowires for which the phase fluctuations are assumed not to occur.

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