Shape-resonant superconductivity in nanofilms: from weak to strong coupling

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Abstract Ultrathin superconductors of different materials are becoming a powerful platform to find mechanisms for enhancement of superconductivity, exploiting shape resonances in different superconducting properties. Here we evaluate the superconducting gap and its spatial profile, the multiple gap components, and the chemical potential, of generic superconducting nanofilms, considering the pairing attraction and its energy scale as tunable parameters, from weak to strong coupling, at fixed electron density. Superconducting properties are evaluated at mean field level as a function of the thickness of the nanofilm, in order to characterize the shape resonances in the superconducting gap. We find that the most pronounced shape resonances are generated for weakly coupled superconductors, while approaching the strong coupling regime the shape resonances are rounded by a mixing of the subbands due to the large energy gaps extending over large energy scales. Finally, we find that the spatial profile, transverse to the nanofilm, of the superconducting gap acquires a flat behavior in the shape resonance region, indicating that a robust and uniform multigap superconducting state can arise at resonance.

Keywords Shape Resonance · Ultrathin Superconductivity · Lifshitz Transitions · BCS-BEC crossover.

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1 Introduction

Superconductivity in strongly confined systems at the nano or atomic scale is attracting a growing interest after the recent observation of a sizable enhancement of the critical temperature in superconducting FeSe systems when reduced to monolayers [1] and the observation of superconductivity above 5 K in graphene doped with Lithium [2]. The multiband nature of the superconductivity in doped FeSe can also lead to amplifications of the superconducting parameters when the chemical potential crosses a Lifshitz transition [3] as well as to BCS-BEC crossover phenomena [4,5] in a multigap configuration [6,7,8,9]. Furthermore, since 2004 the observation of shape resonances in superconducting metallic nanofilms of Pb [10,11,12] and first evidences
of shape resonances in the superconducting critical temperature in metallic nanowires of Sn and Al [13,14,15] clearly established the importance of the interplay between quantum size effects and superconductivity when the lateral dimensions of the system are reduced to the order of the interparticle distance or the pair correlation length [10,11]. It is important to note that current experiments on nanofilms reporting shape resonances in the gap and in the critical temperature [10,11,12] do not find evidences for multigap superconductivity close to the shape resonances. In this paper we identify the parameter regime in which future experiments should directly detect multiple gaps. Strong enhancement of superconductivity has been also predicted and observed when all the lateral dimensions of a bulk superconductor are reduced to the nanoscale, as in nanoparticles, nanoclusters, and nanocubes [20,21,22,23]. Predictions of large amplifications of the superconducting critical temperature and of multigap BCS-BEC crossover phenomena point toward superstripes, i.e. a system of periodic stripes organized in a superlattice, as an ideal candidate system to control and enhance superconductivity at the nanoscale [24,25,26]. Motivated by the fact that many different bulk superconducting materials can be used as a starting system to realize nanostructures, for instance by nanosculpting lithography [27], here we investigate theoretically the nature of the superconducting shape resonances in metallic nanofilms, tuning the parameters of the pairing interaction from weak to strong coupling, and considering different values of the energy scale of the pairing. For a review on theory and experiments discussing the multiband and multigap physics of superconducting nanofilms, see Refs. [17,18]. The shape resonances in the superconducting gaps at zero temperature are characterized in terms of the amplification with respect to the bulk value of the gap and the width of the resonance, where formation of a multicondensate with multiple gaps can be observed. We will show that the amplification is controlled by the pairing strength, while the width of the resonance depends on the energy cutoff of the pairing interaction. Note that recently a heterostructure of superconductors and insulating barriers has been proposed to generate multigap superconductivity also outside the shape resonance region [28]. The chemical potential renormalization at fixed density is also explored, which is important when the system is close to a shape resonance and the gap becomes a large energy scale with respect to the distance of the chemical potential to the Lifshitz transitions [29,30]. In this situation, a mixture of BCS-like and crossover BCS-BEC pairs is realized [31,32,33], providing the best condition to stabilize the detrimental superconducting fluctuations [34] which can be strong in reduced dimensionality [35]. Finally, we investigate the spatial profile of the superconducting gap parameter and of the density of electrons. We find that the shape resonant superconductivity is characterized by a flat behavior of the gap profile, to be contrasted with a many-peak gap outside resonance. Resonant superconductivity is therefore the most robust phase of superconducting nanofilms in the strong quantum confinement regime.

The paper is organized as follows. In Section II we discuss the coupled mean-field equations for the gaps and the chemical potential, where we report the expressions of the gap and density profiles in the Anderson’s approximation to the Bogoliubov-de Gennes equations. In Section III we report and analyze the numerical results at \( T = 0 \) for the gaps, the chemical potential and the spatial profile of the gap and the electronic density. In Section IV we summarize our results and we conclude with predictions for key experiments to detect the effects reported in this paper.

## 2 Methods

The approach employed in this paper to investigate the ground state of superconducting nanofilms in the strong quantization regime is based on a full self-consistent solution of the coupled gaps and density equations, arising from the Anderson's prescription [36] to solve the Bogoliubov-de Gennes equations for non uniform superconductors [37]. It is the same approach introduced in the seminal paper by Thompson and Blatt [16]. Here, we consider a system of electrons confined in a thin metallic slab with infinite potential walls. In the direction parallel to the film, the electrons have a parabolic dispersion with an effective mass equal to the bare mass of the electrons. In the direction perpendicular to the film the motion of the electrons is quantized, with formation of discrete single-particle energy levels, as given by the solution of the uni-dimensional Schrödinger equation [14,16]. Hence, the electronic subbands have the following form:

\[
\xi_n(k) = \frac{|k|^2}{2m} + E_n - \mu; \quad E_n = \frac{1}{2m} \left( \frac{n\pi}{L} \right)^2 ,
\]

where \( k \) is the wave-vector of the electrons parallel to the film, \( m \) is the effective mass, \( \mu \) is the chemical potential, and \( E_n \) are the discrete energies of the subband bottoms. The index \( n=1,2,... \) labels the electronic subbands. For a given chemical potential, the Fermi surface exhibits a number of concentric circular 2D Fermi sheets. The reduced Planck constant \( (\hbar/2\pi) \) is taken equal to unity throughout the paper.
The electrons interact via an effective attraction characterized by an interaction strength $V^0$ and an energy cutoff $\omega_0$. The effective pairing attraction of the bulk system is taken in a separable form, as in Ref. \[16\]. Moreover, because in the case of nanofilms the motion along the $z$-axis is tightly bound, the bare strengths of the potential that control the intraband pairings and the interband exchange (Josephson-like) pairing between the two subbands are related by $V_{nm}^0 = V^0(1 + \frac{1}{2} \delta_{nm})$. This expression is due to the overlap integral of the single-particle wave-functions, as arising from the Anderson approximation to the full Bogoliubov-de Gennes (BdG) equations (a detailed comparison between the Anderson approximation and the exact BdG solution is available in Ref. \[8\]). Therefore, the quantum confinement in superconducting nanofilms is able to generate different intraband and pair exchange interactions, but the partial condensates of each subband are strongly coupled by the pair exchange terms, being the intraband term only 50% larger than the pair exchange. As we will see below, this behavior is at the origin of a not too evident multigap structure in single superconducting nanofilms. We note also that this large pair-exchange interaction will prevent the resonant condensate to enter the BEC regime at strong coupling \[9\].

In the limit of large thicknesses. At a mean field level the form originally introduced for two-band superconductors can be written as

$$V_{nm}(k, k') = -V^0(1 + \frac{1}{2} \delta_{nm}) \Theta (\omega_0 - |e(k)|) \times \Theta (\omega_0 - |e(k')|), \quad (2)$$

where $V^0$ is the (positive) strength of the attractive potential. The $k$-dependence of the (isotropic s-wave) gaps is a consequence of the separable form of the interaction of Eq. (2) and it is given by

$$\Delta_n(k) = \Delta_0 \Theta (\omega_0 - |e(k)|). \quad (3)$$

where the factor 2 is the spin degeneracy of the electrons and $v_n(k)$ is the BCS weight of the occupied states

$$v_n(k)^2 = \frac{1}{2} \left[ 1 - \frac{\xi_n(k)}{\sqrt{\xi_n(k)^2 + \Delta_n(k)^2}} \right]. \quad (6)$$

The sums over $k$ are replaced by two-dimensional integrals over momenta and then by integrals over the energy variable, after introducing the 2D density of states $N_{2D} = m/(2\pi)$. The integrals of Eqs. (4-5) can be expressed in a closed form, as shown in Ref. \[14\].

This self-consistent system of equations for the multiple gaps and the chemical potential in superconducting nanofilms has been investigated recently both at an analytical and numerical level in Refs. \[40,41\], with their focus on the role of different boundary conditions of the nanofilms and the continuity of the shape resonances as a function of thickness.

In this work we will evaluate also the spatial profile of the total superconducting gap $\Delta(z)$ and the total density of conduction electrons $n_e(z)$ along the direction transverse to the nanofilm. Within the Anderson approximation we have,

$$\Delta(z) = \sum_n |\Psi_n(z)|^2 \Delta_n \int \frac{dE}{2\sqrt{(E + E_n - \mu)^2 + \Delta_n^2}}, \quad (7)$$

$$n_e(z) = 2 \sum_n |\Psi_n(z)|^2 \int dE v_n^2(E). \quad (8)$$

where $\Psi_n(z) = \sqrt{2/L} \sin(n\pi z/L)$ is the single particle wave-function along the $z$ direction corresponding to the energy level $E_n$, solution of the Schrödinger equation in the transverse direction with infinite wall potential. The extremes of the integral are the same of the coupled self-consistent gap and density equations, determined by the entering and exiting of each subband bottom $E_n$ from the Debye energy window. In the density profile of Eq. (8) the contribution of the free electron density outside the Debye energy window (hence, with zero gaps) is also included.

### 3 Results

In Figure 1 we report the superconducting gap in the first subband as a function of the nanofilm thickness for different 3D couplings, from weak ($\lambda = 0.3$) to very strong coupling ($\lambda = 2.0$), at fixed energy cutoff of the pairing interaction ($\omega_0 = 300\, K$). The gaps are normalized to their bulk value, obtained in the limit of large thickness ($k_F L >> 1$).

In Table \[1\] we report the bulk values of the gap for different couplings, from the weak ($\lambda < 0.6$) to the strong ($\lambda > 1.0$) coupling regime. In the third
Fig. 1 Superconducting gap in the first subband \( \Delta_1 \) as a function of the film thickness \( L \) for different couplings. \( \Delta_1 \) is normalized to the corresponding bulk values of the gap \( \Delta_{\text{bulk}} \) determined for different couplings (see text).

Table 1 Bulk values of the gap normalized to \( \omega_0 \) and the amplification factors \( A \) at \( L = 0.8 \) nm for the different couplings \( \lambda \) here considered.

| \( \lambda \) | \( \frac{\Delta_{\text{max}}}{\omega_0} \) | \( A = \frac{\Delta_{\text{max}}}{\Delta_{\text{bulk}}} \) |
|-------|-----------------|------------------|
| 0.3   | 0.073           | 2.98             |
| 0.6   | 0.394           | 1.85             |
| 1.0   | 0.855           | 1.61             |
| 1.5   | 1.399           | 1.54             |
| 2.0   | 1.926           | 1.50             |

column of Table 1 we show the amplification factor \( A = \Delta_{\text{max}}/\Delta_{\text{bulk}} \) of the gap at the shape resonance, taken in the ultrathin regime at \( L = 0.8 \) nm. For weak coupling the gap amplification is large, while it approaches values of order unity for stronger couplings. Therefore, weakly coupled superconductors are the best candidate to observe quantum size effects and shape resonances in the superconducting gaps (and in the critical temperature).

Note that increasing the thickness \( L \) the amplification \( A \) becomes less dependent on the coupling. Interestingly for experimental detection, even for the large thickness \( L = 5 \) nm, the amplification of the gap is approximately 1.25, which is a measurable effect in all practical cases.

In Figure 2 we show the superconducting gaps in the first subband and in the last subband contributing to the pairing as a function of the nanofilm thickness tuned around a shape resonance \( \left( N_{\text{res}} = 10 \right) \) for different values of the energy cutoff \( \omega_0 \) at fixed coupling strength \( \lambda \). As in Figure 1, the gaps are normalized to their bulk value, obtained in the limit of large thickness \( (k_F L \gg 1) \), see Table 1. As one can see, the multigap regime of the superconducting condensate is present only in the shape resonant region, and for the here-considered cases we have all the gaps equal \( (\Delta_1 = \Delta_2 = ... = \Delta_9) \), except the gap of the last subband \( (\Delta_{10}) \). For \( \omega_0 = 300 \) K, the largest difference between these gaps is found at the anti-resonance, with a factor 1.05 of difference for the resonance \( N_{\text{res}} = 10 \) at \( L = 4.56 \) nm, and the width of the resonance having multigap character is found over width-span \( \delta L = 0.07 \) nm. Increasing the energy cutoff to \( \omega_0 = 1500 \) K, the width-span of the resonance showing multiple gaps increases to \( \delta L = 0.37 \) nm, which is now a range of thicknesses realizable in current nanofilm deposition processes. It is therefore crucial to consider systems with large energy cutoffs and in the weak coupling regime to amplify in size and in width the multigap resonant character of the confined superconductors, in order to be able to access experimentally the interesting multigap regime, never observed in the single superconducting nanofilms.

In Figure 3 the chemical potential as a function of thickness for different couplings and two different cutoff energies is reported. The chemical potential \( \mu \) is normalized with respect to the Fermi energy of the 3D bulk non interacting system \( E_F \), value that is approached in the limit \( (k_F L) \gg 1 \). Since we work at fixed conduction electron density, the chemical potential is renormalized by the discrete structure of the electronic levels and by the superconducting gap opening. The main effect is the discreteness of the levels, while the gap opening, both in value and in energy extension \( (2\omega_0) \), determines differences only around the shape resonant region, differences which become sizable when \( \omega_0 \) and the gaps increases in the strong coupling regime. We find that in the ultrathin limit \( L < 3 \) nm, the solution of the coupled gaps and density equations is important
and it is not possible to work at fixed chemical potential to get the precise locations in $L$ of the shape resonances.

Figure 4 shows the total gap profile $\Delta(z)$ along the direction transverse to the film ($z$), evaluated according to Eq. (7). For the case $\lambda = 0.6$ and $\omega_0 = 300$ K we consider three cases: the thickness $L = 2.210$ nm at the $N = 4$ anti-resonance, $L = 2.222$ nm very close to the shape resonance, and $L = 2.270$ nm outside and above the shape resonance.

The results shown in Figure 4 indicate an interesting behavior of the superconducting gap profile: $\Delta(z)$ outside resonance displays the Friedel oscillations, as already discussed in Ref. [14], together with the vanishing of the gap profile at the boundaries due to infinite-wall boundary conditions. The new interesting property reported here is a quite flat behavior of $\Delta(z)$ when the nanofilm thickness $L$ is tuned very close to the shape resonance, see the case $L = 2.222$ nm in Figure 4. We have also analyzed other shape resonances for larger $L$, finding an even flatter behavior at resonance, owing to the larger number of harmonics entering in the calculation of $\Delta(z)$. Regarding the electron density profile $n_e(z)$ of Eq. (8), we have found a very flat dependence of $n_e(z)$ at the center of the nanofilm, with tiny oscillations as a function of $z$ (less than 10% of the maximal density). Therefore, close to shape resonances the superconducting ground state of the nanofilms in the quantum-size regime appears to be quite uniform, with the exception of the boundaries (for more realistic boundary conditions, see [40]), together with sizable amplifications of the gaps, and hence it points toward an optimized shape-resonant superconductivity.

4 Conclusions

In this paper we have shown that the ground state properties of shape-resonant superconductivity in ultranarrow nanofilms strongly depend on the microscopic details of the pairing interaction. The amplification of the superconducting gap is the largest in the ultrathin limit and in the weak-coupling regime of pairing. The same amplification is progressively reduced when the coupling is increased toward strong coupling. The width of the shape resonance is instead governed by the energy cutoff of the pairing interaction: the range of thicknesses of the nanofilms in which superconductivity is shape-resonant increases for increasing energy cutoff, allowing the formation of a multicondensate and multi-gap superconducting phase in the shape-resonant region. Interestingly, the gap profile along the transverse direction of the nanofilm indicates a uniform and robust superconducting state at resonances. The multigap properties at resonance may be detected by next generation nano-ARPES [42,43] or nano-STM measurements [44], which are in construction to investigate structural and electronic complexity in high-T$_c$ superconductors. Therefore, we conclude that the optimal shape resonant superconductors can be realized starting from intermediate to weak-coupling bulk superconductors having large energy cutoffs, as in FeSe monolayers or doped graphene systems, reducing one or more dimensions to the nano or atomic scale.

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