SIZE-RESONANCE EFFECT IN CYLINDRICAL SUPERCONDUCTING NANOWIRE

M.D. Croitoru, A.A. Shanenko and F.M. Peeters

Department of Physics, University of Antwerp, 171, Groenenborgerlaan, B-2020, Antwerp, Belgium
(Received 29 March 2007)

Abstract

We investigate the dependence of the basic superconducting quantities - the order parameter, energy gap and critical temperature - on the quantum confinement of a nanoscale superconductor in the clean limit. The Bogoliubov-de Gennes equations are solved numerically for a Pb nanowire in the clean limit with cylindrical cross section. Strong size superconducting resonances are found in the critical temperature of a Pb nanowire.

1.Introduction

In recent years, due to remarkable experimental progress, the study of the superconducting properties in nanostructures has attracted a lot of interest. Characteristic feature of such structures is quantum confinement of charge carriers. The control of the size and shape of the carrier confinement that favors the enhancement of superconductivity and leads to an oscillatory behavior of the superconducting properties is an important issue. With this in mind, the study of superconductivity in nanostructures like nanowires is of fundamental interest, due to possibility of tuning superconducting characteristics by changing the cross section.

As far back as the sixties of the last century, Blatt and Thompson [1] calculated a remarkable sequence of peaks in the thickness dependence of the energy-gap parameter of superconducting nanofilms in the clean limit. They called these spikes size resonances. It was not possible to produce highly crystalline superconductors with nanoscale dimensions at that time. Only very recently the thickness-dependent oscillations of T_c were observed experimentally in Pb nanofilms [2]. For decades atomic nuclei were the only system where the interplay between quantum confinement and pairing of fermions could be studied experimentally and where the expectations of Blatt and Thompson were confirmed as a series of size resonances in the pairing energy gap of nuclei [3]. Recent advances in fabrication technology for nanosized structures can make breakthroughs in the fundamental question about the size dependence of the properties of nanoscale superconductors [2, 4-14]. For example, in Ref. 13 it was shown that the latter method supplemented by ion-beam sputtering allows one to reduce the nanowire width down to 10nm. In Ref. 15 it is strongly suggested that the recent experimental observations of the width-dependent increase in the superconducting-transition temperature of clean Al and Sn nanowires [14, 16, 17] are the first observations of the size-dependent resonances in quasi-one-dimensional superconductors.

In the present paper we investigate the dependence of the basic superconductive quantities on the confinement of Pb nanowires. Since the dimensions of nanostructures are comparable or even smaller than the typical superconductor coherence lengths, a correct analysis of superconductivity phenomena in such systems requires a detailed quantum-mechanical treatment. Quantum-size effects strongly influence the physical (and superconducting) properties of highly-crystalline nanoscale metallic structures. In order to take these effects into account, we numerically solve the Bogoliubov-de Gennes (BdG) equations for a clean metallic nanowire with a cylindrical cross section uniform over the whole wire length. Note that the BdG equations are designed to describe the superconducting condensate for an arbitrary confining potential. In contrast to the Ginzburg-Landau theory, there is no need to introduce effective boundary conditions for the order parameter.

The paper is organized as follows. In Sec. 2, we outline the formalism of the BdG equations written for a crystalline nanowire in the parabolic band approximation. Details of our numerical method for solving these equations self-consistently are presented in this section. Section 3 presents numerical results of the BdG equations solved in a self-consistent manner for a Pb nanowire. We discuss size superconducting resonances resulting in quantum oscillations of the critical temperature and effect of such resonances on the superconducting order parameter. The conclusions are presented in Sec. 4.

2. Model

As known since the classical papers by Gor'kov [18] and Bogoliubov [19], the superconducting order parameter can be seen as the wave function governing the center-of-mass motion of the Cooper pairs. Hence, in the presence of quantum confinement, the order parameter will be a spatially dependent function ($\Delta = \Delta(\mathbf{r})$) even in the absence of a magnetic field. Such quantum-confinement effects cannot be studied within the Ginzburg-Landau theory and a more complex approach as, e.g., based on the Bogoliubov-de Gennes (BdG) equations [17] is needed. These equations are written as

$$\left[\hat{H}_e\sigma_z + \Delta\sigma_x\right]\psi_i = E_i\psi_i, \tag{1}$$

where

$$\psi_i(\mathbf{r}) = \begin{pmatrix} u_i(\mathbf{r}) \\ v_i(\mathbf{r}) \end{pmatrix}$$

is the two-component (particle and hole) wave function corresponding to the quasi-particle (bogolon) energy E_i; σ_x and σ_z are the Pauli matrices acting in the particle-hole space; the single-electron Hamiltonian H_e reads

$$\hat{H}_e = -\frac{\hbar^2}{2m}\nabla^2 + V(\mathbf{r}) - \mu, \tag{2}$$

with μ the chemical potential and $V(\mathbf{r})=V_B\theta(\rho - R)$ the confining potential (V_B is the barrier parameter assumed to be infinite in the present paper, and $\theta(x)$ is the Heaviside step function). For a cylindrical nanowire $i = \{j, m, k_z\}$, where j label the subbands appearing due to the size quantization of the electron motion in the transverse ρ directions, m is the azimuthal quantum number; and k_z is the wave-vector of the quasi-free electron motion along the z direction.

In order to find the quasi-particle energy spectrum E_i and corresponding wave functions $u_i(\mathbf{r})$ and $v_i(\mathbf{r})$, we need to solve the BdG equations self-consistently together with the relation

$$\Delta(\mathbf{r}) = g\sum_{i\in C} u_i(\mathbf{r})v_i^*(\mathbf{r})\left[1 - 2f(E_i)\right], \tag{3}$$

where g is the coupling constant and $f(E_i) = 1/[\exp(\beta E_i) + 1)$ is the Fermi function ($\beta=1/(k_B T)$ with T the temperature and k_B the Boltzmann constant). It is important that the summation in Eq. (3) is over all the eigenstates that have positive quasi-particle energy E_i and the single-electron energy

$$\xi_i = \int d\mathbf{r}\left[u_i^*(\mathbf{r})\hat{H}_e u_i(\mathbf{r}) + v_i^*(\mathbf{r})\hat{H}_e v_i(\mathbf{r})\right] \tag{4}$$

within the Debye "window" $[-\hbar\omega_D, \hbar\omega_D]$ (ω_D is the Debye frequency). Below we indicate by R the full set of quantum numbers corresponding to all the eigenstates with positive energy whereas C denotes the subset of the eigenstates with $\xi_{i\in C} \in [-\hbar\omega_D,\ \hbar\omega_D]$. For a given mean electron density n the chemical potential μ is determined by

$$n = \frac{2}{V}\sum_{i\in R}\int d\mathbf{r}\left[|u_i(\mathbf{r})|^2 f(E_i) + |u_i(\mathbf{r})|^2 (1 - f(E_i))\right] \quad (5)$$

with $V = \pi R^2 L_z$. Along the z direction we introduce a unit cell of length L_z which is repeated periodically. Due to the quantum confinement in the transverse directions we have to impose the following boundary conditions

$$u_i(\mathbf{r})\big|_{\mathbf{r}\in S} = v_i(\mathbf{r})\big|_{\mathbf{r}\in S} = 0 \quad (6)$$

on the wire surface while in the longitudinal direction periodic boundary conditions are used. Equations (1)-(6) are the basic formulas needed to obtain a self-consistent solution.

In order to numerically solve the BdG equations (1), we expand the two-component wave function $\psi_i(\mathbf{r})$ as

$$\begin{pmatrix} u_i(\mathbf{r}) \\ v_i(\mathbf{r}) \end{pmatrix} = \frac{e^{ik_z z}}{\sqrt{L_z}}\frac{e^{im\varphi}}{\sqrt{2\pi}}\sum_n \varphi_n(\rho)\begin{pmatrix} u_n^i \\ v_n^i \end{pmatrix} \quad (7)$$

in terms of the quantum well states

$$\sum_n \varphi_n(\rho) = \frac{\sqrt{2}}{R\, J_{m+1}(\alpha_{mn})} J_{m+1}\left(\alpha_{mn}\frac{\rho}{R}\right). \quad (8)$$

After inserting Eq. (7) into the BdG equations we obtain the following equations for the expansion coefficients:

$$\left(T_n^i - E_i\right)u_n^i + \sum_{n'}\Delta_{nn'}v_{n'}^i = 0 \quad (9a)$$

$$\left(E_i - T_n^i\right)v_n^i + \sum_{n'}\Delta_{nn'}u_{n'}^i = 0 \quad (9b)$$

where ($i = \{j, m, k_z\}$)

$$T_n^i = \frac{\hbar^2}{2m}\left[\frac{\alpha_{mn}^2}{R^2} + k_z^2\right] - \mu \quad (10)$$

and

$$\Delta_{nn'} = \int d\rho\, \varphi_n(\rho)\Delta(\rho)\varphi_{n'}(\rho). \quad (11)$$

Then, Eqs. (9) are readily converted into a matrix form so that the eigenvalues and eigenfunctions of the problem can be found by means of diagonalizing the relevant matrix.

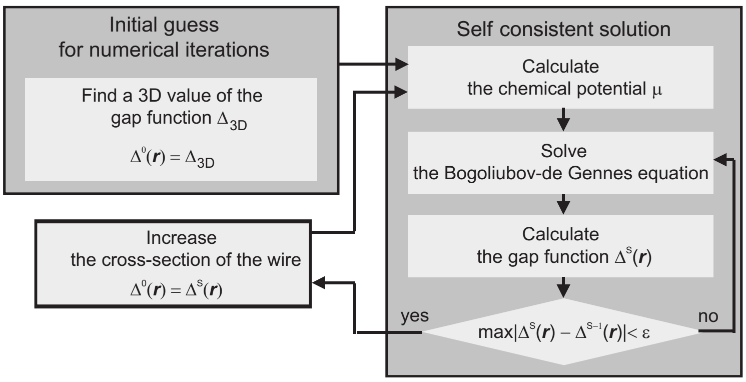


Fig.1. Numerical flow diagram for the self-consistent solution of the BdG equations.

Our procedure of solving the BdG equations consists of three steps (see Fig. 1). At the first step, a 3D bulk value of the gap function Δ_{bulk} is used as the initial guess of our iteration, and the BdG equations are solved by substituting $\Delta_{bulk} \rightarrow \Delta(\mathbf{r})$. At the second step, the obtained eigenfunctions $u_i(\mathbf{r})$ and $v_i(\mathbf{r})$ together with the corresponding excitation energies E_j are inserted into Eq. (3). At the third step, solving the BdG equations, with the order parameter found at the previous step, yields new eigenfunctions and corresponding quasi-particle energies that are used in the next iteration. For a given radius R the second and third steps are repeated till the maximum difference $\delta = \max|\Delta^s(\mathbf{r}) - \Delta^{s-1}(\mathbf{r})|$ between the order parameters at consecutive iterations is sufficiently small to satisfy the adopted accuracy requirement, we took typically $\delta < \varepsilon = 10^{-3}$. When increasing the cross section of the nanowire, the order parameter, obtained for a smaller cross section can be used as an initial guess in the iteration process. If T is significantly lower than T_c (T_c is found as the temperature above which there is only a zero solution to the BdG equations), our algorithm is stable during all simulated iterations and is rapidly convergent. For example, $\delta \sim 10^{-3}$ is usually reached after 20-30 iterations for $T = 0$. Even for $T \approx 0.95\ T_c$ the convergence is reasonably fast (the number of iterations needed increases only by a factor of 3) and does not depend very much on the initial guess for $\Delta(\mathbf{r})$. However, very near to T_c the algorithm becomes very sensitive to this guess and more and more iterations are required to reach convergence.

3. Results and discussion

In the present model the bulk density of single-electron states per unit volume and per spin projection can be written as

$$N(0) = \frac{mk_F}{2\pi^2\hbar^2}, \tag{12}$$

with k_F the 3D Fermi wave vector. In order to simulate an infinite nanowire we have chosen the length $L_z = 1000$ nm $\gg \lambda_F = 2\pi/ k_F$ (the Fermi wavelength) for the periodic boundary in the z direction. The real ρ-space grid sampling was chosen to be 0.2 Å. In our numerical investigations we restrict ourselves to a Pb nanowire with the Debye temperature $\hbar\omega_D / k_B$=96K and the bulk density of states $N(0)$ multiplied with the interaction constant is equal to 0.39.

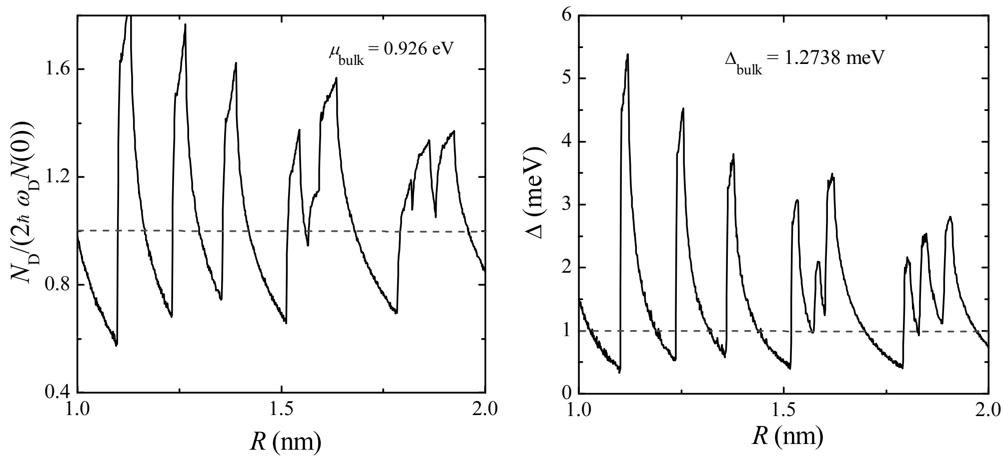


Fig.2. The radius dependent relative mean density of the single-electron states in the Debye window per unit volume and per spin projection $N_D/(2\hbar\omega_D V)$ and energy gap Δ at zero temperature for a Pb nanowire.

Quantitative description of the recently observed quantum-size oscillations in the critical temperature of superconducting nanofilms [2] as well as a detailed understanding of the size-dependent variations in the work function, the surface energy and the thermal stability [21-25] requires the knowledge of the crystal band structure in the presence of quantum confinement. As shown in Ref. 26, this procedure, to a certain extent, can be avoided by using the band-mass approximation together with a change of the reference point in the band structure. This can be realized by the introduction of an effective Fermi level [28]. Following this scheme, we employ the effective Fermi level for a Pb nanowire $\mu_{\rm bulk} = 0.926$ eV, which follows from the recent results found in Ref. 6.

The mean density of single-electron states in the Debye "window" per volume unit and spin projection $N_{\rm D}/(2\hbar\omega_D V)$ is a good start for the understanding of the superconductive properties [15, 27]. Here $N_{\rm D}$ is the number of single-electron states (for one spin projection) situated in the Debye "window". Fig. 2 illustrates the size-dependence of $N_{\rm D}/(2\hbar\omega_D V)$ for a Pb nanowire as a function of radius R in units of the bulk density of states near the Fermi level $N(0)$. In the calculations R is varied with steps of $\Delta R = 0.001$Å. From Fig. 1 one can see that the Van Hove singularities have a strong effect on the mean density of states, which results in a sequence of profound peaks. These peaks appear due to quantum confinement of the electron motion in the transverse directions of the nanowire, which results in the formation of discrete electronic states and, so, in a splitting of the total band of single-electron states in a series of subbands. With an increase (a decrease) of wire radius, the bottom of a given subband moves up (down) in energy. When it passes through the Fermi surface, a sharp increase in the mean density of relevant states occurs.

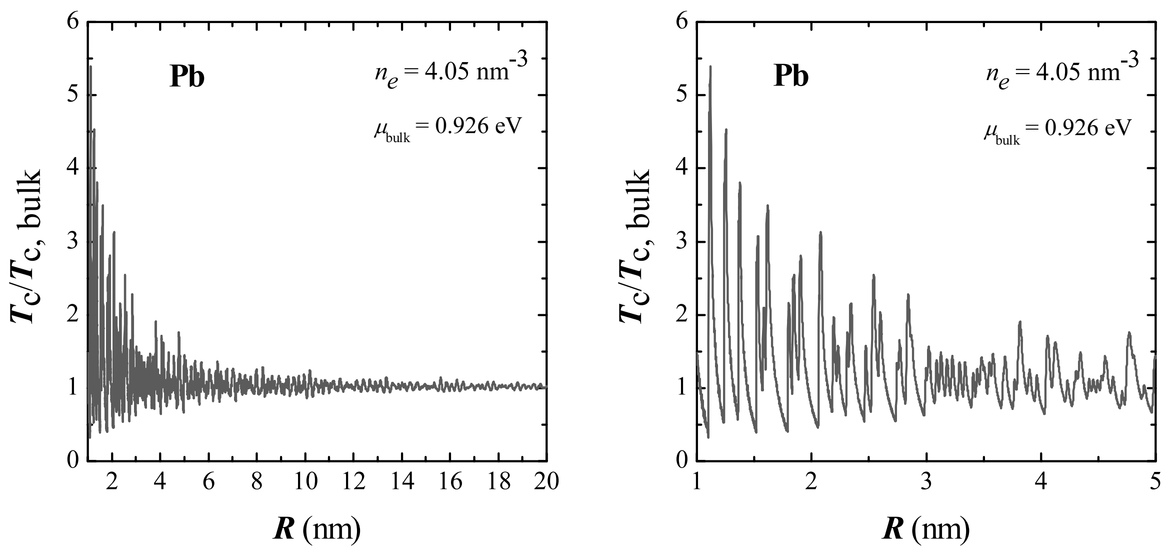


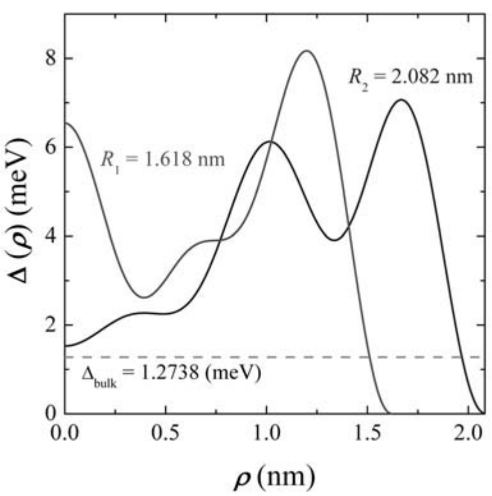
Fig.3. Left: The critical temperature T_c in units of $T_{c,\rm bulk}$ as a function of nanowire radius for a cylindrical Pb nanowire. Right: the same dependence but in the small-R region.

The peaks in the mean density of single-electron states result in pronounced size resonances of the superconducting critical temperature as seen in Fig. 2, where $T_c/T_{c,\rm bulk}$ is plotted as a function of nanowire radius in the same radius interval. It is noteworthy that the critical temperature increases well above its bulk value at a resonant point but, then drops down until a sequent resonance comes into play. As a rule, the resonance enhancements with respect to

$T_{c,\text{bulk}}$ are much more pronounced than the drops between two neighboring resonances. Moreover, the larger the resonance magnitude, the larger the drop below $T_{c,\text{bulk}}$ between these resonances. Resonances in $T_c/\ T_{c,\text{bulk}}$ are rather sensitive to the governing parameters ω_D, g and μ_{bulk} (see Refs. 15, 27). Comparing the obtained results for Pb with those for Al and Sn given in Refs. 15, 27 one can see that the superconductive resonances weaken with increase of any of these parameters. For example, as follows from Refs. 15, 27, aluminium nanowires exhibit the most significant resonances. Enhancement in the electron-phonon coupling in tin and lead results in a considerable suppression of the multi-spike pattern in Sn and Pb. Such dependences are explained by the fact that the superconducting resonances result from a competition between quantum-confinement energy and superconducting "condensation" energy: the stronger the quantum confinement, the more pronounced are the radius-dependent resonances.

We would like to emphasize that the drops in gap function occur in nanowires with a cross section uniform over the whole nanowire length. In real samples there are unavoidable cross-section variations so that the critical temperature measured experimentally cannot be compared directly with T_c presented in Fig. 2. Hence, the theoretical results obtained for a uniform nanowire should be averaged over the particular cross-section variations and such averaging will be influenced by the superconducting resonances. Indeed, as shown in Ref. 15, the experimental data for nearly clean Al [12, 13] and Sn [14] nanowires exhibit a monotonous increase in T_c with a decrease of R and follow the average trend of the size-dependent superconducting resonances calculated via the BdG equations for a cylindrical uniform nanowire [15].

If one recall from the textbooks [20, 29] that in bulk the BCS critical temperature depends on the density of single-electron states at the Fermi level as $T_{c,bulk} = 1.13\hbar\omega_D \exp(-1/gN(0))$, one would expect that this formula is still applicable for nanowires but with $N(0)$ replaced by $N_D/(2\hbar\omega_D V)$. This is not the case. For example, application of this straightforward guess results in a 20-30% underestimation of the resonant critical temperatures in a Pb nanowire.



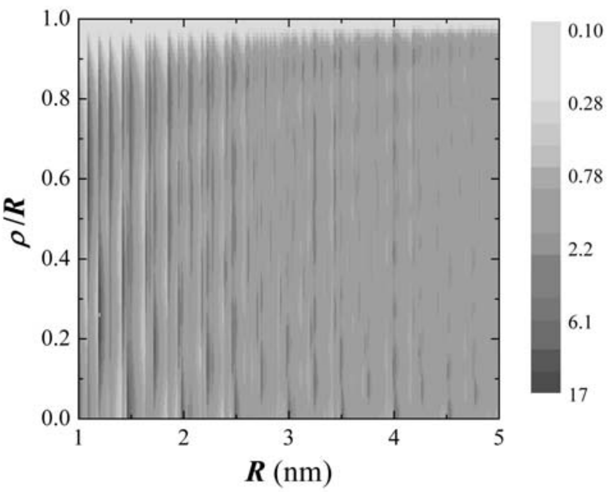


Fig.4. Left: The superconducting order parameter $\Delta(\rho)$ versus radius of a Pb nanowire with cylindrical cross section at two resonant points R =1.618 nm and 2.082nm. Right: The contour plot of the superconducting order parameter profile $\Delta(\rho/R)$ as a function of the normalized radial coordinate ρ/R and of the radius of the Pb nanowire.

An obvious consequence of the confinement in a nanoscale superconducting wire is the appearance of a nonuniform spatial distribution of the superconducting condensate. Any profound size resonance is accompanied by strong spatial variations of the superconducting order parameter and by a giant increase of its mean value (this value is close to the energy gap parameter). For example, the superconducting order parameter calculated at the resonant points R = 1.618 nm and 2.082 nm is plotted in Fig. 3(a). As seen from this figure, the spatial condensate structure is strongly inhomogeneous. This is very similar to the behavior of the order parameter in a superconducting nanofilm [28]. But now, contrary to nanofilms, the curves do not exhibit regular spatial oscillations.

From Fig. 3(b), where the order parameter Δ(ρ/R) as a function of the normalized radial coordinate ρ/R and of the nanowire radius is plotted as a contour plot, it is seen that the spatial variations in the superconducting order parameter become less pronounced with rise of the nanowire radius. At R > 2 nm the electron subbands appearing due to the size quantization are not well separated any more: a new size resonance appears when the previous one has not yet decayed. From Fig. 3(b) one can notice that any superconductive resonance demonstrates its unique spatial structure which is never repeated at another resonant point. In the areas, where the superconducting order parameter is enhanced, it significantly exceeds the bulk gap Δ_{bulk} = 1.2738meV. In general, the smaller the nanowire width, the more profound is the increase of the order parameter (the critical temperature) at a resonant point.

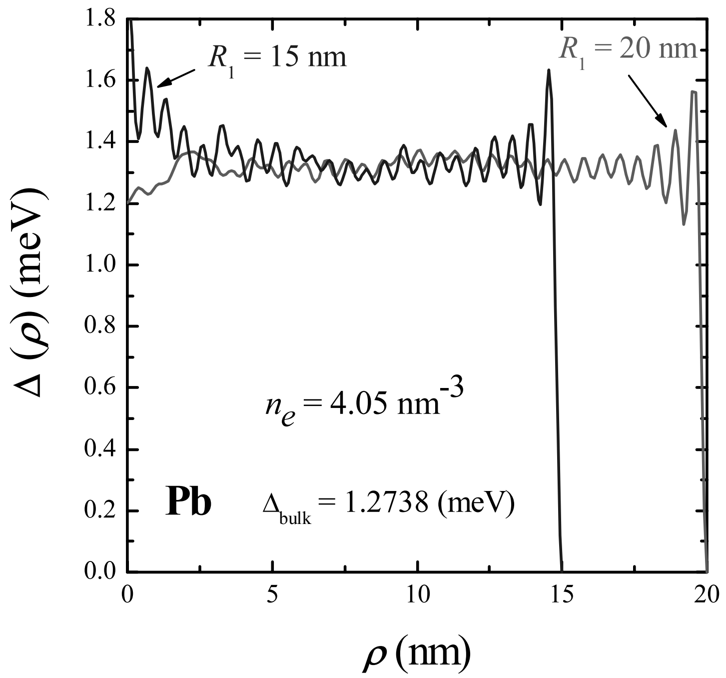


Fig.5. The order parameter Δ(ρ) in Pb nanowires with cylindrical cross section for large values of radii of the nanowires.

When the resonance decays (Fig. 3(b)), the spatially averaged value of the order parameter decreases to 1meV (nearly the bulk value Δ_{bulk} = 1.27meV) and the spatial oscillations in the order parameter become less profound. Nevertheless, they are still significant in the areas close to the boundaries (see Fig. 4). Note that the reason for these boundary effects is the same as in the case of the well-known Friedel oscillations induced by a point charge placed into an electron gas. Our numerical analysis shows that such near-boundary behavior survive for very thick nanowires (with radius larger than 50 nm) when there are no oscillations of the order parameter in the center regions. From Fig. 3 one can clearly see at which values of the radius the number of the oscillations in the order parameter increases. We also remark that the mean distance between two neighboring peaks in T_c given in Figs. 3 and 4 is proportional to $\lambda_F/2$. In the vicinity of $\rho = R$ the order parameter $\Delta(\rho)$ rapidly varies, decreasing to zero also on a scale of order $\lambda_F/2$.

Conclusion

In conclusion, quantum confinement is the main mechanism governing nanoscale superconductivity. Based on a numerical solution of the Bogoliubov-de Gennes equations, we have investigated the sensitivity of the basic superconducting properties - such as the order parameter, critical temperature, energy gap - on the confinement of the Pb nanowire. We have shown that the size-dependent increase of the superconducting temperature in clean Pb nanowires is well explained by the size-resonance effect. In the present paper we showed that the superconducting order parameter can be enhanced (by more than an order of magnitude with respect to bulk) at a resonant point and its spatial distribution exhibits a distinguished nonuniform pattern reflecting the symmetry of the sample.

Acknowledgement

This work was supported by the Flemish Science Foundation (FWO-Vl), the Belgian Science Policy (IAP) and BOF-TOP (University of Antwerp).

References

[1] J.M. Blatt and C.J. Thompson, Phys. Rev. Lett. 10, 332, (1963).
[2] Y. Guo, Y.-F. Zhang, X.-Y. Bao, T.-Z. Han, Z. Tang, L.-X. Zhang, W.-G. Zhu, E.G. Wang, Q. Niu, Z.Q. Qiu, J.-F. Jia, Z.-X. Zhao, Q.K. Xue, Science, 306, 1915, (2004).
[3] S. Hilaire, J.F. Berger, M. Girod, W. Satula and P. Schuck, Phys. Lett. B, 531, 61, (2001).
[4] M.M. Özer, J.R. Thomson and H.H. Weitering, Nature Phys., 2, 173, (2006).
[5] X.-Y. Bao, Y.-F. Zhang, Y. Wang, J.-F. Jia, Q.-K. Xue, X.C. Xie and Z.-X. Zhao, Phys. Rev. Lett., 95, 247005, (2005).
[6] Y.-F. Zhang, J.-F. Jia, T.-Z. Han, Z. Tang, Q.-T. Shen, Y. Guo, Z.Q. Qiu and Q.-K. Xue, Phys. Rev. Lett., 95, 096802, (2005).
[7] B.G. Orr, H.M. Jaeger and A.M. Goldman, Phys. Rev. Lett., 53, 2046, (1984).
[8] B.G. Orr, H.M. Jaeger and A.M. Goldman, Phys. Rev. B, 32, 7586, (1985).
[9] A. Bezryadin, C.N. Lau and M. Tinkham, Nature (London), 404, 971, (2000).
[10] M.A. Skvortsov and M.V. Feigel'man, Phys. Rev. Lett., 95, 057002, (2005).
[11] M. Harmele, G. Rafael, M.P.A. Fisher and P.M. Goldbart, Nature Phys., 1, 117, (2005).
[12] M. Savolainen, V. Touboltsev, P. Koppinen, K.-P. Riikonen and K. Arutyunov, Appl. Phys. A, 79, 1769, (2004).
[13] M. Zgirski, K.-P. Riikonen, V. Touboltsev and K. Arutyunov, Nano Letters, 5, 1029, (2005).
[14] M. Tian, J. Wang, J.S. Kurtz, Y. Liu, M.H.W. Chan, T.S. Mayer and T.E. Mallouk, Phys. Rev. B, 71, 104521, (2005).

[15] A.A. Shanenko, M.D. Croitoru, M. Zgirski, F.M. Peeters and K. Arutiunov, Phys. Rev. B, 74, 052502, (2006).
[16] M. Savolainen, V. Touboltsev, P. Koppinen, K.-P. Riikonen and K. Arutyunov, Appl. Phys. A, 79, 1769, (2004).
[17] M. Zgirski, K.-P. Riikonen, V. Touboltsev and K. Arutyunov, NanoLetters, 5, 1029, (2005).
[18] L.P. Gor'kov, Sov. Phys. JETP, 7, 505, (1958).
[19] N.N. Bogoliubov, Sov. Phys. Usp., 67, 549, (1959).
[20] P.G. de Gennes, Superconductivity of Metals and Alloys, W.A. Bejamin, New York, 1966.
[21] A.R. Smith, K.-J. Chao, Q. Niu and C.K. Shih, Science, 273, 226, (1996).
[22] L. Gavioli, K.R. Kimberlin, M.C. Tringides, J.F. Wendelken and Z. Zhang, Phys. Rev. Lett., 82, 129, (1999).
[23] K. Budde, E. Abram, V. Yeh and M.C. Tringides, Phys. Rev. B, 61, R10602, (2000); V. Yeh, L. Berbil-Bautista, C.Z. Wang, K.M. Ho and M.C. Tringides, Phys. Rev. Lett., 85, 5158, (2000).
[24] M. Hupalo, S. Kremmer, V. Yeh, L. Berbil-Bautista and M.C. Tringides, Surf. Sci., 493, 526, (2001); M. Hupalo and M.C. Tringides, Phys. Rev. B, 65, 115406, (2002).
[25] W.B. Su, S.H. Chang, W.B. Jian, C.S. Chang, L.J. Chen and T.T. Tsong, Phys. Rev. Lett., 86, 5116, (2001).
[26] C.M. Wei and M.Y. Chou, Phys. Rev. B, 66, 233408, (2002).
[27] A.A. Shanenko and M.D. Croitoru, Phys. Rev. B, 73, 012510, (2006).
[28] A.A. Shanenko, M.D. Croitoru and F.M. Peeters, Europhys. Lett., 76, 498, (2006); ibid. Phys. Rev. B, 75, 014519, (2007).
[29] A.L. Fetter and J.D. Walecka, Quantum Theory of Many-Particle Systems, Dover, New York, 2003.