PAPER ID: 0009
DOI: 10.18462/iir.cryo.2019.0009

The Enhancement of the Critical Temperature in Thin Aluminium Films

Egor SEDOV(a), Ilya GOLOKOLENOV(a,b), George KONSTANTINIDIS (c), Antonis STAVRINIDIS(c), George STAVRINIDIS(c), Vitaliy ZAVIALOV(b), Konstantin ARUTYUNOV(a,b)

(a) National Research University Higher School of Economics
Moscow, 101000, Russia, esedov@hse.ru
(b) P. L. Kapitza Institute for Physical Problems RAS
Moscow, 119334, Russia
(c) Institute of Electronic Structure & Laser (IESL), Foundation for Research & Technology Hellas (FORTH),
Heraklion, 70013, Greece

ABSTRACT

It is well known that new effects appear in superconductors with the reduction of their size. Among them one is the most interesting – phenomenon of changing of critical temperature. It can be both decrease and increase in different metals, however, despite the number of existing works, there is still no generally accepted conception of what is exactly the origin of this effect. At the moment it is more or less clear, that this is a rather complicated mechanism, which is influenced by many factors, particularly connected with the sample's manufacturing. Nevertheless, we suppose even after minimization of all impacts, the temperature of superconducting transition shifts anyway because of quantum size effect. We present here the results of the investigation of high-quality polycrystalline aluminum films and demonstrate the presence of quantum-confinement process that was not considered earlier.

Keywords: superconductivity, nanofilms, quantum-size effects, quantum confinement.

1. INTRODUCTION

The investigation of electric properties of different metallic structures with the reduction of their dimensions has been started in the middle of the last century. It was studied as typical conductor like tin (Shal'nikov A. I., 1938), lead (Lock J. M., 1951), indium (Toxen A. M., 1961), aluminum (Strongin M. et al, 1965, Shanenko A. A. et al, 2006), niobium (Stromberg T. F., and Swenson C. A., 1962) and different sandwiches structures (Astrakharchik E. G. and Adkins C. J., 1998, Strongin M., et al, 1967). Despite the diversity of characteristics, one of the most attractive is the critical temperature Tc of superconductors. The fact that still there are no room temperature superconductors effective for use in computers, and the tendency towards miniaturization of electronic devices makes the variation of Tc in superconductive nanostructures is very promising field of study. Nevertheless, the question about the nature of critical temperature shift in superconductors with decreasing of their dimensions remains unsolved, and the scientific community has not received a generally accepted opinion concerning the main factor responsible for this effect.

Initially, several mechanisms have been considered as main factors influencing alteration of the critical temperature: disorder-driven and phonon effects. Thus, couple of works were made keeping in mind those two factors, for review see refs. (Dynes R. C., et al, 1978, Jaeger H. M., et al, 1989, Liu Y., et al, 1991) and (Abeles B., et al, 1966, Naugle D. G., et al, 1973, Strongin M., et al, 1965), respectively. Nevertheless, even at that time it become clear that one should take into account the existence of another phenomenon which play a no less role then the others. For example, in the article (Toxen A. M., 1961) the author noticed that in contrast to the earlier studies (Lock J. M., 1951) there is not only the impact of stresses in the films, but also the quantum size effect. However, the importance of the influence of the manufacturing conditions was not denied. Later, V. L. Ginzburg suggested that in extremely pure, monoatomic superconductive film the temperature of transition would be the same as in bulk (Ginzburg V. L., 1964). Unfortunately, no one could prove or disprove that thesis due to absence of appropriate objects of study. After the beginning of 2000th the new impulse in studying of low-dimensional structures was driven by recent advances in fabrication technology (Savolainen M., et al, 2004, Zgirski, et al, 2005). The dimension in any direction of new systems can reach now a few nanometers opening new possibilities for tuning of electrical properties of various objects. Particularly, there is a huge field for investigation of quantum size effect.

At the present time, there are several works where the key role is attributed to the quantum confinement of charge carriers occurring in nano-dimensional structures as a consequence of quantum size effect (Eom D., et al, 2006, Chen Y., et al, 2012, Shanenko A. A., Croitoru M. D., and Peeters F. M., 2006). In contrast to conventional considering of quantum confinement as the effect influenced on system through the electronic density of states, we demonstrate manifestation of another quantum mechanism which increases the electron-electron attraction by modification of electron wave function. The basis of our research is based on highly accurate nanofabrication, microscopic analyses and experimental study of Tc dependence on thickness of high-quality polycrystalline thin aluminum films, and a theoretical exploration of the Bogoliubov-de Gennes equations. Cooperation among phonons induced by surface-substrate effect and quantum mechanical effect can noticeable rise the critical temperature of films compared with bulk sample (Shanenko A. A. and Croitoru M. D., 2006)

2. MAIN SECTION

The critical temperature increases in thin film superconductors as aluminum (Khukhareva I. S., 1962, Strongin M., et al, 1965, Shanenko A. A. et al, 2006), indium (Toxen A. M., 1961, Lock J. M., 1951), tin (Shal'nikov A. I., 1938) and decreases in mercury (Khukhareva I. S., 1961), lead (Tinkham M., 1963), niobium (Khlyustikov I. N. and Moskvin S. I., 1985). Among these metals we have chosen aluminum not only because of the pronounced size dependence of Tc, but also because of its utilization in numerous nanoelectronic devices, for instance, quantum computer (Yohannes D. T., et al., 2015). Historically, aluminum has demonstrated pronounced dependence between the dimensions of the structure and its critical temperature, and more importantly Tc is higher in nanostructrures (thin films Strongin M., et al, 1965, Khukhareva I. S., 1962, Chubov P. N. et al, 1968 and nanowires Shanenko A. A. et al, 2006) than in bulk samples.

2.1. Experiment

During the examination process of previous experimental works, we encountered a problem of the absence of structured data. The electronic characteristics of the film varied strongly even between samples fabricated in one series: the temperature of the substrate during the deposition process, deposition rate, grain size, etc. could be different. As the result, it was sometimes impossible to connect the experimental data and theoretical interpretation leading to ambiguity in completed investigation. Thus, our main task was to produce high-quality aluminum nanofilms with grain size comparable to the thickness of the film, and moreover the manufacturing conditions should be the same in all cases of the fabrication. Thus interpretation of results can be reduced solely to a size effect eliminating possible fabrication artifacts.

As the result, the set of aluminum thin films was fabricated with thickness *d* varying from 5 nm to 100 nm. The single-crystalline GaAs was selected as the material of substrate, and aluminum films was made by e-beam evaporation at the vacuum 10^{-9} mBar. The quality of the films, their sizes and the size of crystallites was controlled by high resolution TEM analyses. Surely, there is a presence of the oxide layer on the surface of the film (approximately 2 nm) and the amorphous layer on the border between substrate and aluminum (about 3 nm). Thereby the precise thickness of the specimens can be varied by these two affects. Nevertheless, the TEM analyses in conjunction with previous studies on aluminum nanostructures (Savolainen M., et al, 2004, Zgirski, et al, 2005) enables us to claim the high-purity of the films.

We present typical dependence of R(T) of 70 nm thick film in figure 1 a) and the image of this sample b). We defined the critical temperature as the point on the R(T) dependence where the resistance of the sample has reduced by a factor of two compared to its value in normal state. The curve presented in fig. 1 was measured between "legs" D+ and A+, the value at room temperature was 11.1 Ohm and at 1.4 K temperature was 0.9 Ohm, thus demonstrating the quality of the films. In order to make sure that the experimental results are reproducible we used two type of cryostats for measurement. One is direct-pumped 4He bath setup with the lowest temperature about 1.28 K and the second is more complicated setup – 3He sorption cryostat reaching the much lower temperature down to 0.4 K. To measure the resistance of the samples, in former case the DC current configuration was used and in the latter the lock-in AC current scheme. In both cases the 4-probe measurements were made, the value of current was between 0.1 uA and 1 uA, and the connection lines were highly filtered (Zavyalov V. V. et al, 2018). Thermometers in these systems were calibrated against the 4He pressure with ~ 1 mK precision, also we used superconducting bulk materials as tantalum, indium, aluminum and titanium for reference points, the transition temperature of the pure aluminum bulk was 1.19 K, corresponding to literature sources.

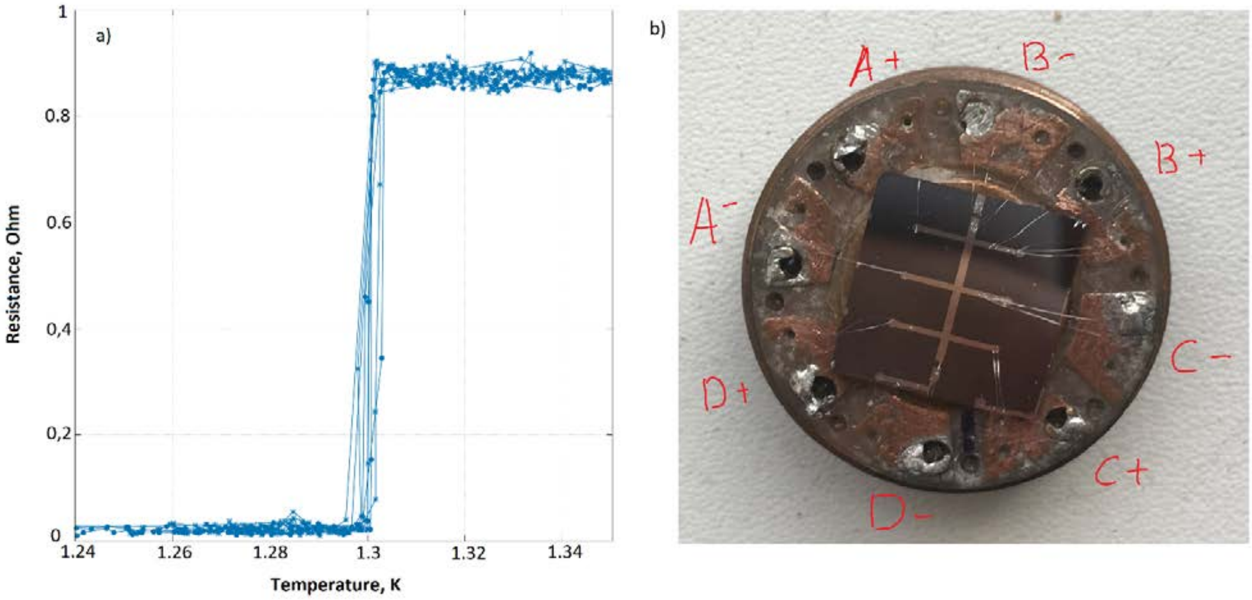

Figure 1: a) Typical R(T) dependence of the 70 nm thick aluminium film measured with 1 uA excitation current. b) Optical microscope image of the sample holder with typical sample. Usually contacts B- and D- were used for current injection and the others were voltage probes.

2.2. Theory

According to BCS theory, the critical temperature Tc depends exponentially on the electronic density of the states on the Fermi level N(E$_F$) and electron-phonon interaction potential V:

$$T_c \sim \exp\left\{-\frac{1}{N(E_F)V}\right\}$$

In the article (Shanenko A. A., et al, 2008) it is shown that both parameters change non-monotonically with the decrease of sample's thickness because of quantum size effect being the consequence of shape-resonant theory, developed by Blatt and Thomson several decades ago (Blatt J. M. and Thomson C. J., 1963).

In general, the physics of the shape-resonance effect in nanofilms can be understood as follows. In the presence of quantum confinement, the energy band $\mathcal{E}(\mathbf{k})$ splits into a series of sub-bands. If the dimension of perpendicular projection of the film changes, both rise/lower, these sub-bands move down or up in energy, respectively. Every time, when the bottom of a sub-band crosses the Fermi energy, the density of states increases abruptly and hence there is an enhancement of superconductivity. As a result, any superconducting property shows the thickness-dependent oscillations (Blatt J. M. and Thomson C. J., 1963, Shanenko A. A., et al, 2008, Chen Y., et al, 2012). The period of the calculated oscillating dependence Tc(d) is of the order of one monoatomic layer (Shanenko A. A., Croitoru M. D., Zgirski M., Peeters F. M., and Arutyunov K., 2006). The inevitable surface roughness of real samples is noticeable larger. Hence, only the envelope of the rapidly oscillating Tc(d) function is observed in experiment.

We do not take into account the surface or substrate contribution because the scale of our films lies far beyond the ultrathin regime, where these effects play the key role. Another important thing is that in a first approximation one can ignore the impact of different stresses and effect of disordering due to the high-quality of the specimens. Thus, we suppose that the increase of the superconducting transition temperature caused by quantum-confinement and phonon effects. See refs. (Arutyunov K. Yu., et al, 2018 or Shanenko A. A., et al, 2008) for more detailed theoretical considerations.

2.3. Results

The cumulative result of our measurements is presented by solid line with triangles in the figure 2, here, in addition, we provide the data from other papers. In work (Khukhareva I. S., 1962), two types of films were considered: films evaporated at room temperature and in liquid helium with continuous annealing to 573 K. In picture 2 the latter data is not depicted because of high statistical error. The Tc(d) enhancement between the two types of films was not dramatic and was associated by the author of that paper with variation of the mean free path between samples.

In another article (Chubov P. N. et al, 1968), the authors also noticed the increase of the critical temperature of aluminum films with the reduction of their thickness. Nevertheless, they have not suggested any new explanations of this phenomenon and just meant that the change of Tc was influenced by different factors as temperature of annealing, technic of the samples' fabrication, grains size, the presence of stresses, quantum size effects etc.

We imply that the shift of the critical temperature is connected with the quantum confinement effect on account of the equal conditions of the manufacturing and comparable size of crystallites with the thickness of the film in each case. It can be seen the general trend is follows: the thinner the aluminum film, the higher is the transition temperature. The quantitative discrepancy between the results of our groups is likely the difference between the conditions of fabrication, leading to noticeable variations of the characteristic size of crystallites.

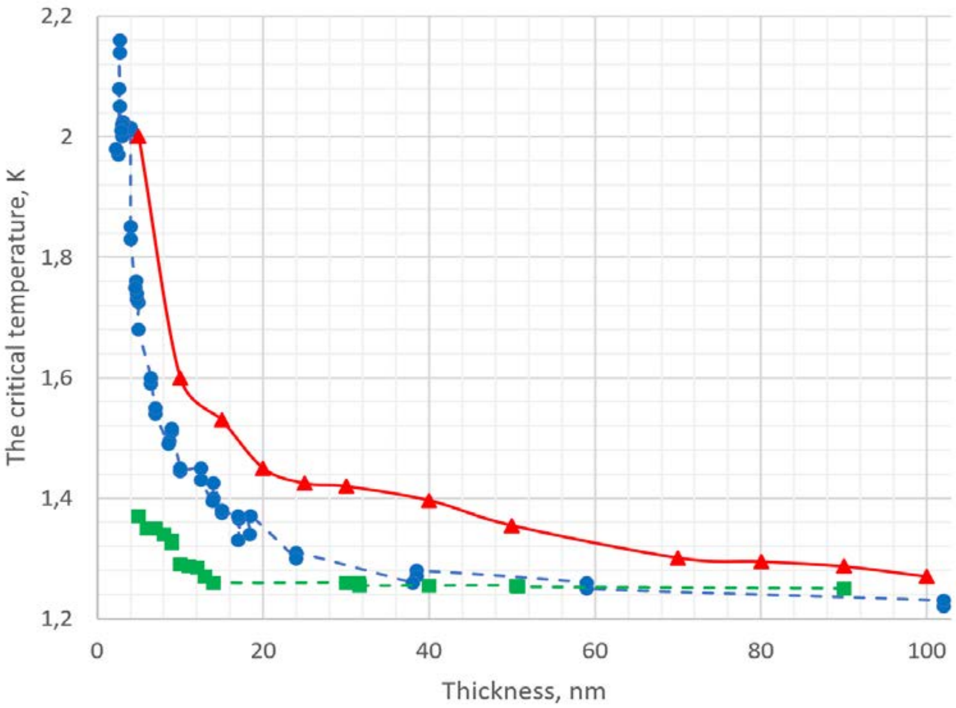

Figure 2: Experimental dependence of the critical temperature of thin aluminium films as function of its thickness. Green squares are result from work Khukhareva I. S., 1962; blue circles are from paper by Chubov P. N. et al, 1968 and the last red triangles are our own results.

3. CONCLUSIONS

In conclusion, we demonstrated the increase of the critical temperature of thin aluminium films connecting this effect with the quantum size effect. The theory calculated by our colleagues (Arutyunov K. Yu., et al, 2018 or Shanenko A. A., et al, 2008) is in good agreement with our experimental data which correlates well with known literature sources.

ACKNOWLEDGEMENTS

This work was supported by the joint Russian-Greek Projects No. RFMEFI61717X0001 and No. T4ΔΡΩ-00031 "Experimental and theoretical studies of physical properties of low dimensional quantum nanoelectronic systems ".

REFERENCES

Abeles B., Cohen R. W. and Cullen G. W. 1966. Enhancement of Superconductivity in Metal Films. Phys. Rev. Lett. 17, 12, 632-635;

Arutyunov K. Yu. Et al. 2018. Nanoarchitecture: Toward Quantum-Size Tuning of Superconductivity. Phys. Status Solidi RPL, 1800317;

Astrakharchik E. G. and Adkins C. J., 1998. Superconductivity Enhancement in Screened Ultrathin Metal Films. J. Phys. Condens. Matter, 10, 4509-4514;

Blatt J. M. and Thomson C. J., 1963. Shape Resonances in Superconducting Thin Films. Phys. Rev. Lett. 10, 8, 332-334;

Chen Yajiang, Shanenko A. A., and Peeters F. M., 2012. Superconducting transition temperature of Pb nanofilms: Impact of thickness-dependent oscillations of the phonon-mediated electron-electron coupling. Phys Rev. B., 85, 224517;

Chubov P. N., Eremenko V. V. and Pilipenko Yu. A., 1968. Dependence of the Critical Temperature. Zh. Eksp. Teor. Fiz. 55, 752-765;

Dynes R. C., Garno J. P. and Rowell J. M., 1978. Two-Dimensional Electrical Conductivity in Quench-Condensed Meatl Films. Phys. Rev. Lett. 40, 479, 479-482;

Eom Daejin, Qin S., Chou M.-Y., and Shih C. K., 2006. Persistent Superconductivity in Ultrathin Pb Films: A Scanning Tunneling Spectroscopy Study. Phys. Rev. Lett. 96, 027005;

Ginzburg V. L., 1964. On Surface Superconductivity. Journ. Exper. Theor. Phys. 47, 2318-2320;

Jaeger H. M., Haviland D. B., Orr B. G., and Goldman A. M., 1989. Onset of superconductivity in ultrathin granular metal films. Phys. Rev. B. 40, 1, 182-196;

Khlyustikov I. N. and Moskvin S. I., 1985. Superconductivity of the Twinning plane of Niobium and a Topological Phase Transition in a Two-dimensional Superconducting System. Zh. Eksp. Teor. Fiz. 89, 1846-1856;

Khukhareva I. S., 1961. Superconducting Properties of Freshly Deposited Mercury Films. J. Exptl. Theoret. Phys. 41, 728-734;

Khukhareva I. S., 1962. The Superconducting Properties of Thin Aluminum Films. J. Exptl. Theoret. Phys. 43, 1173-1178;

Liu Y., McGreer K. A., Nease B., Haviland D. B., Martinez G., Halley J. W., and Goldman A. M., 1991. Scaling of the insulator-to-superconductor transition in ultrathin amorphous Bi films. Phys. Rev. Lett. 67, 2068;

Lock J. M., 1951. Penetration of Magnetic Fields into Superconductors III. Measurements on Thin Films of Tin, Lead and Indium. Proc. Roy. Soc. (London), A208, 391-408;

Naugle D. G., Baker J. W. and Allen R. E. 1973. Evidence for a Surface-Phonon Contribution to Thin-Films Superconductivity: Depression of Tc by Noble-Gas Overlays. Phys. Rev. B., 7, 7, 3028-3037;

Savolainen M., Touboltsev V., Koppinen P., Riikonen K.-P., and Arutyunov K., 2004. Ion beam sputtering for progressive reduction of nanostructures dimensions. Appl. Phys. A., 79, 1769-1773;

Shal’nikov A. I. 1938. Superconducting Thin Films. Nature. 9, 142;

Shanenko A. A. and Croitoru M. D. 2006. Shape resonances in the superconducting order parameter of ultrathin nanowires. Phys. Rev. B. 73, 012510.

Shanenko A. A., Croitoru M. D., and Peeters F. M., 2006. Quantum-size effects on Tc in superconducting nanofilms. Europhys. Lett. 76, 3, 498-504;

Shanenko A. A., Croitoru M. D., Zgirski M., Peeters F. M., and Arutyunov K., 2006. Size-Dependent Enhancement of Superconductivity in Al and Sn Nanowires: Shape-Resonance Effect. Phys. Rev. B., 74, 052502-1-052502-4;

Shanenko A. A., Croitoru M. D., Peeters F. M., 2008. Nanoscale superconductivity: Nanowires and nanofilms. Phys. Rev. B, 8, 054505;

Stromberg T. F., and Swenson C. A., 1962. Negative Surface Free-Energy Effects in Superconducting Niobium. 9, 9, 370-374;

Strongin M., Kammerer O. F. and Paskin A., 1965. Superconducting Transition Temperature of Thin Films. Phys. Rev. Lett. 14, 23, 949-951;

Strongin M., Kammerer O. F., Douglass D. H. and Cohen M. H., 1967. Effect of Dielectric and High-resistivity Barriers on the Superconducting Transition Temperature of Thin Films. 3, 19, 121-125;

Tinkham M., 1963. Effect of Fluxoid Quantization on Transitions of Superconducting Films. Phys. Rev. 129, 6, 2413-2412;

Toxen A. M., 1961. Size Effects in Thin Superconducting Indium Films. Phys. Rev. 123, 2, 442-446;

Yohannes D. T., et al., 2015. Planarized, Extendible, Multilayer Fabrication Process for Superconducting Electronics. IEEE TRANSACTIONS ON APPLIED SUPERCONDUCTIVITY, 25, 3, 1100405-1100409;

Zavyalov V. V., Chernyaev S. A., Shein K. V., Shukaleva A. G., and Arutyunov K. Yu., 2018. Examination of cryogenic filters for multistage RF filtering in ultralow temperature experiments. J. Phys: Conf. Ser. 969, 012086, 1-6;

Zgirski M., Riikonen K.-P., Touboltsev V., and Arutyunov K., 2005. Size-Dependent Breakdown of Superconductivity in Ultranarrow Nanowires. Nano Letters, 5, 6, 1029-1033.