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ON THE POSSIBILITY OF COOPER PAIRING AND SUPERCONDUCTIVITY IN THE SURFACE BANDS OF NOBLE METALS

We discuss the possibility of a superconducting transition in island films of noble metals, in particular gold, on a semiconductor substrate. As is well known, bulk superconductivity is absent in noble metals, but, in this case, superconductivity may be possible due to the surface states. Such states are well studied for the noble metal–vacuum interface and arise from the peculiarities of the spectra of such metals near the L-point of the Brillouin zone. Similar states should also occur at the metal–semiconductor interface. In the latter case, the interaction of electrons leading to Cooper pairing can be provided by bosonic surface excitations of both phonon and collective electron nature at the metal–semiconductor interface. The interaction with surface phonons is effective at low energies, while, at energies of the order of $E_{\rm F}$ *(the Fermi energy of the surface band), the electron–exciton interaction becomes predominant, the optimal conditions for the latter arise at close values of the penetration depths for electrons and excitons, both in the metal and in the semiconductor. We argue that the superconducting transition in such island films should be governed by a phonon mechanism, although not similar to the Bardin–Cooper–Schrieffer (BCS) one, but that non-phonon interactions also play a significant part, providing a BCS-like contribution into the equation for the superconducting gap. Recently, there have been reports on the experimental observation of superconductivity at almost room temperature in Au–Ag nanostructures. In our opinion, the role of silver in these structures, due to the difference in electronegativity, is reduced to the role of an electron donor to the surface states associated with Au, which increases the value of* $E_{\rm F}$ *and leads to an increase in the BCS-like contribution of the non-phonon interactions.*

Keywords: noble metals, island film, surface states, room-temperature superconductivity.

1. Introduction

The authors would like to dedicate this paper to the memory of two outstanding theoretical physicists who passed away in recent years, Petro Mykhailovych Tomchuk and Ernst Anatolievich Pashitskii. P.M. Tomchuk, starting with his pioneering work at the Institute of Physics of the National Academy of Sciences of Ukraine (see e.g., [1, 2], also [3] and references therein), repeatedly turned to the theoretical analysis of various experimentally observed exotic phenomena related to the quantization effects in gold nanoparticles and the heating of electron gas in gold island films in external fields. E.A. Pashitskii, who also devoted his entire life to the Institute of Physics of the NAS of Ukraine and who had outstanding results in the field of non-phonon models of superconductivity, in particular the high-temperature one, as well as in the theory of low-dimensional systems, was our teacher and longtime collaborator. In our present work, we also attempt to explore the possibility of the existence of rather unusual superconducting properties of gold island films. Interestingly, the only joint publication by P.M. Tomchuk and E.A. Pashitskii [4] dealt with hypothetical possibility of non-phonon superconductivity in thin film of degenerated semiconductor decorated with arbitrary metal nanoparticles.

Lately, there have been reports [5, 6] on the experimental observation of superconductivity at near-

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room-temperature in Au–Ag nanostructures. In the previous experiments, summarized in [5], signatures of superconductivity (such as sharp drops in resistivity influenced by magnetic field) were observed in a rather rare cases among a lot of fabricated specimens, with highly divergent values of T_c, which varied during thermocycling, and where very unstable at ambient conditions.

However, in the last report [6], an improved technology of atmospherically stable Au–Ag mesoscopic thin film deposition was proposed, with the claim of the repeated near-ambient values of T_c with moderate scattering. The essential step in this technology is the chemical deposition of some ion-conducting metal oxide layer with consequent implantation of Ag^+ ions and thermal deposition of Au nanoparticles of the order of 10 nm in size, forming a thin island film. In the case of thicker (continuous) films, there was no evidence of superconductivity, as in the case of bulk noble metals.

Such a mysterious phenomenon motivated us to theoretically investigate the possibility of Cooper-pair superconductivity in the nanoparticle surface states associated with the band structure of the noble metals.

2. Surface States

The schematic representation of the noble metal Fermi surface (FS) in the face-centered cubic first Brillouin zone (BZ) is shown in Fig. 1. In vicinities of the L-points (the regions of hyperbolic equal-energy surfaces or "necks" of the FS), the effective mass in the direction normal to the BZ edge is negative. On the other hand, there is a band gap for the bulk electron states in the $\Gamma - L$ direction due to $s-p$ hybridization, where the surface electron states can emerge.

The surface electron states were predicted by Tamm as early as in 1932 [7] and, since then, were observed experimentally by angle-resolved photoemission for various noble metals (see, e.g., [8–10]). Their existence can be explained by a simple model in the effective mass representation illustrated in Fig. 2, where the (111) surface of the noble metal is described by a rectangular potential barrier U. The periodic potential inside the crystal is converted into an effective mass tensor,

$$\begin{pmatrix} m_{\|} & 0 & 0 \\ 0 & m_{\|} & 0 \\ 0 & 0 & -m_z \end{pmatrix}, \tag{1}$$

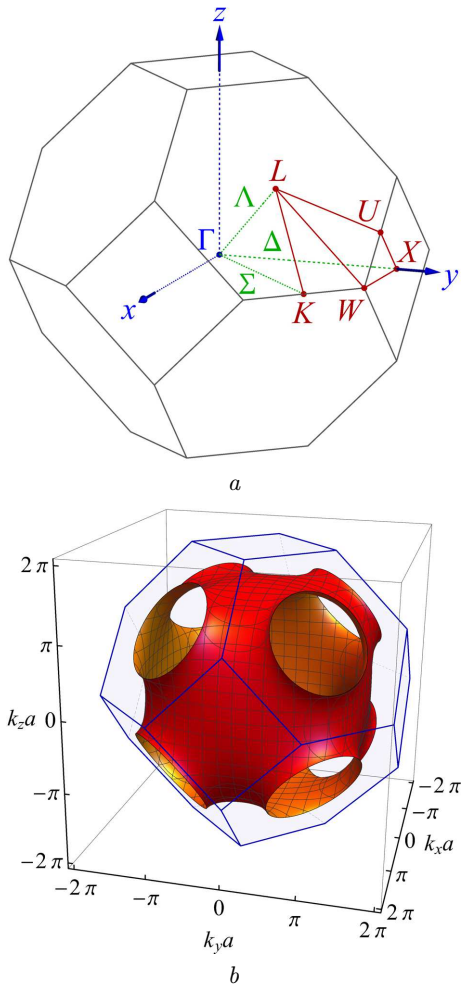

Fig. 1. The first Brillouin zone (*a*) and the Fermi surface (*b*) in the tight-binding approximation for a metal with *fcc* crystal structure

where z direction is chosen along the normal to the interface. On the other side of the interface, the mass is assumed to be isotropic and equal m.

In this case, for the electronic wave function, we have the usual requirement of its continuity at the interface,

$$\psi(z \to -0) = \psi(z \to +0), \tag{2}$$

while the second boundary condition, the continuity of the derivative of the wave function, is replaced in the case of the effective mass approach by the so-called BenDaniel–Duke condition [11], stemming from the requirement of the continuity of the proba-

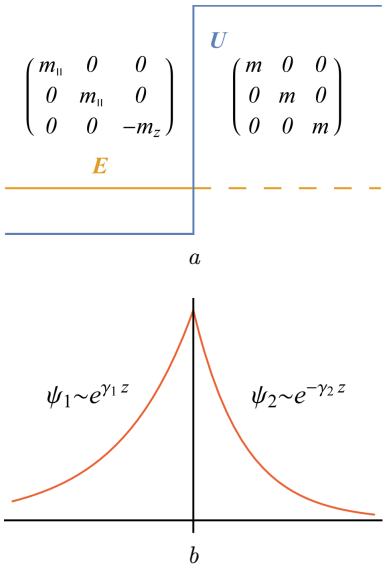

Fig. 2. The potential barrier, associated with the interface (a) and the wave function on both sides of the barrier (b)

bility flow:

$$-\frac{1}{m_z}\left.\frac{d\psi_1}{dz}\right|_{z\to-0} = \frac{1}{m}\left.\frac{d\psi_2}{dz}\right|_{z\to+0}. \tag{3}$$

If we take the wave function on the sides of the energy barrier U to be exponentially decaying away from the boundary,

$$\psi_1 \propto e^{\gamma_1 z}, \quad \psi_2 \propto e^{-\gamma_2 z}, \tag{4}$$

then the Schrödinger equation gives us

$$\frac{k_\parallel^2}{2m_\parallel} + \frac{\gamma_1^2}{2m_z} = E = \frac{k_\parallel^2}{2m} - \frac{\gamma_2^2}{2m} + U, \tag{5}$$

where E is the energy of the quasiparticle and $k_\parallel$ – it's quasimomentum parallel to the interface, and with

$$\frac{\gamma_1}{m_z} = \frac{\gamma_2}{m}, \tag{6}$$

which is the consequence of (3), we obtain the dispersion of the surface band:

$$E\left(k_\parallel\right) = \frac{m_z}{m+m_z}\left[U + \frac{k_\parallel^2}{2m_z}\left(\frac{m}{m_\parallel} + \frac{m_z}{m}\right)\right]. \tag{7}$$

In the case of a noble metal surface in vacuum, the existence of electronic surface bands has been firmly established experimentally by angle-resolved photoemission spectroscopy. Experimental data, illustrating the surface band dispersion (7), may be found, for example, in Fig. 11 from [8], where the surface states were investigated for the copper (111) surface. It should be noted that the above crude model, where the step potential barrier is determined by the metal's work function W ($U = W + E_\mathrm{F} - E_L$) with $W \approx (5.0 \pm 0.2)$ eV and $(E_\mathrm{F} - E_L) \approx (0.8 \pm 0.1)$ eV, $m_z \approx (0.3 \pm 0.05) \cdot m$ for copper [8] and gold [10] (111) surfaces, greatly overestimates in (7) the splitting of the surface band bottom from the bulk state in the L-point (for quantitative estimates at least the image-force potential must be taken into account), but gives a correct qualitative estimate of the dependence on all the parameters. Surface electronic states can also exist on some other surfaces of noble metals–see, e.g., [12] and Fig. 3 therein, showing projections of the gold 3D BZ on several planes, such as (111), (100), (110), and (112), and respective 2D BZs, with shaded regions representing the areas of existence of surface states. Contrary to the (111) case, when filled surface states are located in the centrum of the 2D BZ, for all other cases, surface states appear near the edges of the respective 2D BZs. However, below, all evaluations will be performed for the (111) surface.

3. Superconductivity on Surface States. Bosons Mediating Electron–Electron Interaction

The problem of surface or interface superconductivity has a rather long history. The idea of superconductivity (SC) in the Tamm surface states was proposed by Kirzhnits and Ginzburg [13] within the usual Bardeen–Cooper–Schrieffer (BCS) scheme, but with surface phonons and the 2D density of electron states. Then Ginzburg [14,15] and Allender, Bray and Bardeen [16] paid attention to the excitonic mechanism of SC, but for the bulk states, reflected from the metal–semiconductor interface. The plasmonic mechanism was considered by Pashitskii [17] for the interface of two semiconductors—one with degenerate light electrons and the other with heavy holes.

Here, we assume that a gold nanoparticle has an interface with some non-degenerate n-type semiconductor with positive effective mass, satisfying the condition of the existence of interface electron states, and consider bosonic interface states, possible in

this case and necessary for the emergence of attractive electron–electron interaction. These are acoustic phonons of Rayleigh type (at the surface) or Stoneley type (at the interface), normally polarized with respect to the surface (the interface), consisting of the longitudinal (l) and transverse (t) parts (linked by the boundary conditions of the stress and displacement tensors' continuity) and having the respective penetration depths [18]:

$$\lambda_{l,t}^{\pm} = \left[q_{\parallel}^2 - \left(\frac{\omega}{s_{l,t}^{\pm}} \right)^2 \right]^{-1/2}, \tag{8}$$

where $s_{l,t}^{\pm}$ are the sound velocities of the bulk l and t waves in medium for $z > 0$ (+) and $z < 0$ (−), respectively.

At higher frequencies there are interface p-polarized polaritons of the optical phonon type and of the exciton (or hybridized plasmon–exciton) type with $|\mathbf{H}| \sim (\omega/qc)\,|\mathbf{E}|$ (here ω and q are the frequency and momentum of the bosonic excitation, and c is the frequency of light). Let us estimate their spectrum in the simplest approximation. It can be deduced in the important short wavelength (with respect to $q_{\parallel}$) region, where magnetic field may be neglected in a quasistatic approximation

$$q^2 \equiv q_{\parallel}^2 + q_z^2 = \left(\frac{\omega}{c} \right)^2 \to 0 \quad \left(q_z^2 < 0 \right), \tag{9}$$

from the single Maxwell equation for the electric displacement field $\operatorname{div} \mathbf{D} = 4\pi\rho$ with the boundary condition $D_z^+ - D_z^- = 4\pi\,\sigma \equiv 4\pi e n_s$, where ρ and σ are the 3D and 2D charge densities, respectively. If we assume additional boundary condition of free interface (with zero oscillations of the bulk electron state density on it) then n_s is solely determined by the small deviation from the equilibrium density δN_s of the surface electron states. If we also assume isotropic dielectric function in both media $\mathbf{D}^{\pm} = \varepsilon^{\pm}\mathbf{E}^{\pm}$ and neglect dissipation and bulk (quadratic in q) spatial dispersion in semiconductor, the dispersion of the quasistatic surface excitations is determined from the following equation:

$$\varepsilon^{+}(\omega, q \to 0) + \varepsilon^{-}(\omega, q \to 0) = \frac{4\pi e^2 N_s q_{\parallel}}{m_s \omega^2}. \tag{10}$$

Here

$$\varepsilon^{-} = 1 - \frac{\omega_p^2}{\omega^2}, \tag{11}$$

with ω_p being the bulk plasma frequency of gold, and

$$\varepsilon^{+} = \varepsilon_{\infty} + \sum_{j=1,2} \frac{\alpha_j}{1 - \frac{\omega^2}{\omega_{0j}^2} + i\delta_j \frac{\omega}{\omega_{0j}^2}}, \tag{12}$$

where $j = 1$ stands for the optical phonon (only one is assumed for simplicity) and $j = 2$ for the electron–hole exciton, ω_{0j} and α_j are the frequencies of the bulk transverse excitations and their oscillator strengths, respectively, ε_{∞} is the high-frequency ($\omega \gg \omega_{0j}$) limit of ε^{+}, and m_s is the effective mass of the surface electron state. For the simple model given above (see (7)), we have

$$m_s = m_{\parallel} \frac{m \cdot (m + m_z)}{m^2 + m_{\parallel} \cdot m_z}. \tag{13}$$

The penetration depth of these surface excitations is determined by the imaginary z-component of the wave vector and equals approximately $\lambda_{ex} \equiv |q_z|^{-1} \approx \approx q_{\parallel}^{-1}$ (see (9)).

Let us estimate what kinds of excitations on a semiconductor–gold interface may contribute to the Cooper pairing. Their frequencies must be less than the Fermi energy of interface electron states. Obviously, these are interface acoustic excitations of Stoneley type mentioned above and interface optical phonons with negligible bulk spatial dispersion, well separated from higher electronic excitations for $\omega_{01} \ll \omega_{02}$, ω_p and possessing frequency (see (10)):

$$\omega_1 \approx \omega_{01} \left(1 - \frac{\alpha_1 \omega_{01}^2}{2\omega_p^2 \left(q_{\parallel} \right)} \right), \tag{14}$$

where

$$\omega_p^2 \left(q_{\parallel} \right) = \omega_p^2 \left(1 + \frac{4\pi e^2 N_s}{m_s} q_{\parallel} \right). \tag{15}$$

Here, bulk frequency $\omega_p \approx 2.5$ eV for gold, and we assume $\omega_{01} \approx 0.05$ eV and $\omega_{02} \approx 2$ eV for semiconductor. The Fermi energy of the surface (111) states is near 0.45 eV for copper [8] as well as for gold [10]. But, in the case of interface states, if the bottom of semiconductor conduction band is much closer in energy to the Fermi level than the vacuum work function, the splitting of the bulk and interface electron states in the L-point should be, according to (7), also much smaller than in the free surface case (of the order of 0.1 eV). On the other hand, higher electronegativity of silver may be the cause of electron doping of

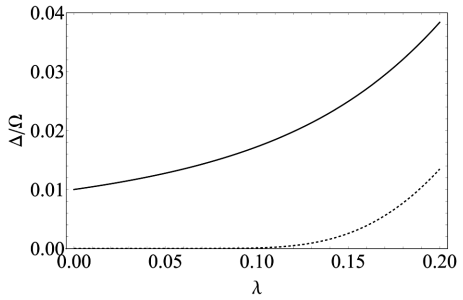

Fig. 3. Superconducting gap dependence on the interaction constant λ according to (19), with $\nu V_0/\Omega = 0.01$ and $\omega/\Omega = 0.01$ (solid curve), and the usual BCS gap dependence on λ (dashed curve)

gold nanoparticle due to the contact with silver ones, which, in turn, should increase the Fermi level in the former. These two factors (lowering of the band bottom and rising of the Fermi level) may increase Fermi energy of interface states up to the values ≤ 1 eV.

As to the bosonic interface excitations of collective electronic nature, there are two hybridized plasmon–exciton branches (labeled 2,3 below), separated from optic phonon solution (labeled 1) of Eq. (10):

$$\omega_{2,3}^2 = \frac{1}{2}\left\{\left[\omega_{02}^2\left(1+\frac{\alpha_2}{1+\varepsilon_\infty}\right)+\frac{\tilde{\omega}_p^2\left(q_\parallel\right)}{1+\varepsilon_\infty}\right]\times\right.$$

$$\left.\times\left[1\pm\sqrt{1-\frac{4\tilde{\omega}_p^2\left(q_\parallel\right)\cdot\omega_{02}^2}{(1+\varepsilon_\infty)\left[\omega_{02}^2\left(1+\frac{\alpha_2}{1+\varepsilon_\infty}\right)+\frac{\tilde{\omega}_p^2(q_\parallel)}{1+\varepsilon_\infty}\right]^2}}\right]\right\}, \tag{16}$$

where

$$\tilde{\omega}_p^2\left(q_\parallel\right) = \omega_p^2\left(1+\alpha_1\omega_{01}^2+\frac{4\pi e^2 N_s}{m_s}q_\parallel\right). \tag{17}$$

The upper branch ω_3 (sign "+" in (16)) is always higher than bulk transversal exciton frequency ω_{02} and is not interesting from the pairing possibility view, while the lower branch ω_2 (sign "−") for a reasonable range of parameters ($\varepsilon_\infty \approx 10\div20$, $\alpha_2 \approx 2\div3$) may be less than the Fermi energy of the interface electron states ($\omega_2 \approx 0.7\div0.8$ eV) and can contribute to their Cooper pairing. We can see from (10)–(12) that 2D electron states on the metal surface/interface do not support their own low-frequency plasmon with characteristic square-root dispersion (contrary to the case of a conducting 2D system sandwiched between two dielectric media), but contribute to the linear spatial dispersion of the optical phonon and hybridized plasmon–exciton interface waves.

4. Dimensional Quantization and Superconductivity on Surface States

As mentioned in the Introduction, the motivation for this work came from superconductivity measurements performed on island films, so, we need to consider the effect of dimensional quantization in nanoparticles. If the nanograins had a regular spherical shape, they would have a set of discrete quantum levels. The energies of the levels with orbital quantum numbers $l = n$ would be roughly proportional to n^2, with $2n+1$ degeneracy. The theory of such finite Fermi systems, including the problem of their superconducting properties, was developed by A.B. Migdal [19]. In a more realistic case of a flattened spheroid, the degeneracy of the energy levels is lifted, but the density of states has sharp maxima determined by the characteristic sizes in the system.

The typical size of the Au particle is about 10÷15 nm, which gives the lower limit of the wave vectors and frequencies of the bosonic modes mentioned above. The upper limit of the relevant momenta for surface states is determined by the diameter $k_{\max} \sim 0.5$ Å^{-1} of the "neck" of the FS. In the case of Au–Ag experiment, the role of Ag with higher electronegativity may consist in the doping of electrons and filling of higher energy levels in Au nanoparticles.

Let us assume that the energy interval ω between the highest partially filled level and all other filled levels is larger than $k_{\max}\cdot s$, where s is the Stoneley wave velocity at the gold–semiconductor interface. In this case such a level makes a separate contribution to the equation for the superconducting gap, which in the first approximation may be written in the form:

$$\Delta = \nu V_0 + \lambda\int\limits_{-\Omega}^{-\omega}\frac{\Delta}{\sqrt{\Delta^2+\xi^2}}d\xi, \tag{18}$$

where Δ is the superconducting gap at zero temperature, Ω is the energy scale of the electron–boson interaction, V_0 represents interaction of electrons on the separate level (with degeneracy ν) with acoustic phonons of Stoneley type, while the sum, corresponding to the contribution of the deeper levels may be

represented as a BCS-like integral with effective interaction constant λ describing the electron–phonon as well as electron–exciton interactions in these levels. This equation may be rewritten in the form

$$\delta\left(1-\lambda\ln\left[\frac{1+\sqrt{\delta^2+1^2}}{x+\sqrt{\delta^2+x^2}}\right]\right)=\frac{\nu V_0}{\Omega}, \tag{19}$$

where $\delta=\Delta/\Omega$ and $x=\omega/\Omega$.

The solution of the equation (19) is shown in Fig. 3 (the parameters for the plot were chosen in accordance with discussion at the end of this section). Here, due to the existence of a separate highest partially filled energy level, the superconducting gap Δ (and critical temperature T_c) for small values of λ is finite, contrary to the usual exponential dependence of BCS. Within the same crude model that led to (18), the critical temperature T_c may be approximated as:

$$T_c\approx\frac{1}{2}\frac{\nu V_0}{1-\alpha\cdot\lambda\ln\left(\frac{\Omega}{\omega}\right)}, \tag{20}$$

where $\alpha\approx 1$ is weekly dependent on ω. The manifestly non-BCS $2\Delta/T_c$ ratio according to (20) and (19) is shown in Fig. 4.

The electron–phonon interaction has the following form (see e.g. [20]):

$$V_0=k_{\max}^2\Xi^2/Ms^2, \tag{21}$$

where Ξ is the matrix element of the deformation potential, M is the atomic mass and s is the sound velocity. Interaction V_0 may be quite large in this case, since it is determined by the acoustic mode, whose velocity squared enters the denominator of the expression for the corresponding interaction constant. For the surface phonons, this velocity should be lower than even the transverse bulk sound velocity, while the deformation potential should be of the same order of magnitude as for the longitudinal bulk modes [21]. Other interactions V_i are determined by the higher-energy modes, such as interface optical phonons and hybridized plasmon-excitons, and the corresponding interaction constants enter the expression for the gap and critical temperature through the effective interaction constant λ, which includes also the effective density of states, corresponding to the low-lying levels.

The estimates of $k_{\max}\cdot s$, as well as of the 2D-electron level splitting ω, give a few millielectronvolts (tens of K), while for $\Omega=\omega_2\left(q_\parallel=k_{\max}\right)$ (see (16)) we have $\Omega\approx 0.7\div 0.8$ eV. So, according to (20), the intrinsically phonon, though non-BCS, mechanism of superconductivity may be significantly enhanced by the non-phonon BCS-like one, even for relatively small values of $\lambda\approx 0.1$.

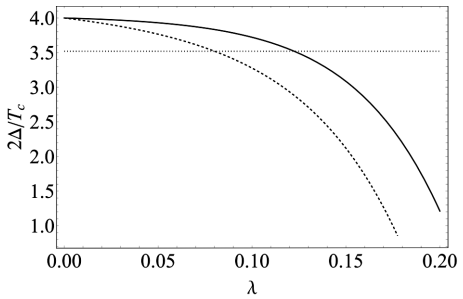

Fig. 4. The $2\Delta/T_c$ ratio according to (20) and (19) with $\nu V_0/\Omega=0.01$ and $\omega/\Omega=0.01$ in function of the interaction constant λ, for $\alpha=1$ (solid curve) and $\alpha=1.14$ (dashed curve), and the usual BCS double gap to T_c ratio ≈ 3.52 (dotted line)

5. Conclusions

We have shown that the dimensional quantization of the surface states can significantly change the character of the Cooper pairing near the interface of the metal with dielectric or semiconductor. The critical temperature of superconducting transition in this case should be proportional to the interaction constant of the 2D electrons with the interface acoustic modes of Stoneley type. The high-frequency modes, such as interface optical phonons and hybridized plasmon-excitons, may appreciably enhance T_c, but their contribution can be described within the BCS-like approach.

It should be noted that, if the Fermi level is close enough to the conduction band of the semiconductor, the penetration depth of the surface electronic wave function may be large enough to ensure percolation between the nanoparticles, with the establishment of superconducting state in the whole island film. Such a situation may emerge in the experiments mentioned in the Introduction due to the doping of electrons by the Ag nanograins to the Au islands.

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ПРО МОЖЛИВІСТЬ КУПЕРІВСЬКОГО ПАРУВАННЯ І НАДПРОВІДНОСТІ В ПОВЕРХНЕВИХ ЗОНАХ БЛАГОРОДНИХ МЕТАЛІВ

Обговорюється можливість надпровідного переходу в острівцевих плівках благородних металів, зокрема золота, на напівпровідниковій підкладинці. Як відомо, об'ємна надпровідність у благородних металів відсутня, але в цьому випадку надпровідність можлива за рахунок поверхневих станів. Такі стани добре вивчені для межі благородний метал–вакуум і виникають з особливостей спектрів таких металів поблизу L-точки зони Бріллюена. Подібні стани мають виникати і на межі метал–напівпровідник. В останньому випадку взаємодія електронів, що приводить до куперівського парування, може забезпечуватися бозонними поверхневими збудженнями як фононної, так і колективної електронної природи на межі метал–напівпровідник. Взаємодія з поверхневими фононами ефективна за низьких енергій, тоді як за енергій порядку $E_{\rm F}$ (енергія Фермі поверхневої зони) переважає електрон–екситонна взаємодія, оптимальні умови для якої виникають за близьких значень глибин проникнення електронів і екситонів, як в металі, так і в напівпровіднику. Ми стверджуємо, що надпровідний перехід у таких острівцевих плівках має відбуватися за фононним механізмом, хоча й не подібним до механізму Бардіна–Купера–Шріффера (БКШ), але суттєву роль відіграють також нефононні взаємодії, які дають БКШ-подібний внесок у рівняння для енергетичної щілини. Нещодавно з'явилися повідомлення про експериментальне спостереження надпровідності за майже кімнатної температури в наноструктурах Au–Ag. На нашу думку, роль срібла в цих структурах, завдяки різниці електронегативностей, зводиться до ролі донора електронів до поверхневих станів, пов'язаних з Au, що збільшує величину $E_{\rm F}$ і приводить до зростання БКШ-подібного внеску нефононних взаємодій.

Ключові слова: благородні метали, острівцева плівка, поверхневі стани, надпровідність за кімнатних температур.