# Pairing with nonzero spin in layered and in quasi-one-dimensional superconductors

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Layered and quasi-one-dimensional superconductors are considered in which the attraction between electrons located on neighboring layers or filaments is the strongest. A superposition of states with spin 0 and spin 1 is realized in such superconductors. The influence of a magnetic field and electron hopping on the properties of these superconductors is investigated. A comparison is made with experiment.

## 1. INTRODUCTION

It has been observed in certain layered superconductors that the superconductivity does not vanish in magnetic fields, several times exceeding the paramagnetic limit.[1] One possible explanation of this phenomenon is that the electrons in such superconductors form pairs with total spin equal to one. Such pairing arises if the attraction between electrons located in different conducting layers is the strongest. Polarization of the molecules of the dielectric spaced between the conducting layers is a possible mechanism for this attraction. Below we shall not investigate specific mechanisms for an attraction between electrons located in different layers, but we shall ascertain what consequences such an attraction induces. It leads to the result that the electrons appearing in a given pair are located in different layers. For a small probability of electron hopping between the layers, the Pauli exclusion principle is unimportant, and the total spin of the pair may be equal to zero or one. The most favorable situation is a superposition of these two states. Only the pairing with spin one remains in the presence of sufficiently strong magnetic fields, parallel to the layers. An analogous phenomenon may appear in quasi-one-dimensional superconductors. In this case the fluctuations destroy the long range order.

## 2. EQUATIONS FOR THE GAP IN LAYERED SUPERCONDUCTORS

Let us consider a system of parallel planes. The interaction between the electrons is described by an effective potential $V(\mathbf{r}-\mathbf{r}')$, including the Coulomb repulsion. We assume that the electrons on neighboring planes are attracted to each other. Neglecting electron hopping from one plane to another, let us write down the Gor'kov equations for such a system:

$$(i\omega_n-\xi(\mathbf{p})+\mu\boldsymbol{\sigma}\mathbf{H})\hat{G}_{ij}(\omega_n,\mathbf{p})+\sum_k\hat{\Delta}_{ik}\hat{F}_{kj}^+(\omega_n,\mathbf{p})=\delta_{ij}, \tag{1}$$

$$(i\omega_n+\xi(\mathbf{p})-\mu\boldsymbol{\sigma}\mathbf{H})\hat{F}_{ij}^+(\omega_n,\mathbf{p})-\sum_k\Delta_{ik}^*\hat{G}_{kj}(\omega_n,\mathbf{p})=0,$$

$$\hat{\Delta}_{ij}^*=TV_{ij}\sum_{\omega_n}\int F_{ij}^*(\omega_n,\mathbf{p})\,d\xi. \tag{2}$$

In Eqs. (1) and (2)

$$(\hat{G}_{ij})_{\alpha\beta}=-\langle T_\tau a_{i\alpha}(\mathbf{p})a_{j\beta}^+(\mathbf{p})\rangle,$$
$$(\hat{F}_{ij}^+)_{\alpha\beta}=-\langle T_\tau a_{i\alpha}^+(\mathbf{p})a_{j\beta}^+(\mathbf{p})\rangle,$$

$\alpha$ and $\beta$ are spin indices; i and j label the number of the planes; $\mu$ is the Bohr magneton; $\sigma_x$, $\sigma_y$, and $\sigma_z$ are the Pauli matrices; $\xi(\mathbf{p}) = v_0(|\mathbf{p}| - p_0)$, and $v_0$ and $p_0$ denote the velocity and momentum at the Fermi surface. Summation is carried out over repeated indices, and the vector $\mathbf{p}$ is parallel to the planes. Furthermore,

$$V_{ij}=\rho\int V(\mathbf{r}-\mathbf{r}')\,e^{i(\mathbf{p}-\mathbf{p}')(\mathbf{r}_\parallel-\mathbf{r}_\parallel')}\,d^2\mathbf{r}_\parallel'\,\frac{dn'}{2\pi}, \tag{3}$$

where $\rho$ is the density of states at the Fermi surface and $\mathbf{n}' = \mathbf{p}'/|\mathbf{p}|$. The magnetic field in Eq. (1) is parallel to the planes and does not affect the orbital motion.

First let us consider the case when the magnetic field is equal to zero. A nonzero solution of Eqs. (1) and (2) for $\hat{\Delta}_{ij}$ first appears at a temperature $T_c$ which is determined from the usual relationship

$$1=TV\sum_{\omega_n}\int\frac{d\xi}{\omega_n^2+\xi^2} \tag{4}$$

In formula (4) $V = \max\{-V_{ij}\}$. We assume below that electrons located in neighboring planes are most strongly attracted to each other. In this connection the interaction of the electrons in one plane, which may be repulsive, does not affect the obtained solution.

The values of $\hat{\Delta}_{ij}$, which are the solutions, differ from zero only in the case when i and j denote the labels of neighboring planes. (The contribution from the interaction of electrons, located in planes with labels differing by more than two, is small and is not taken into consideration below.)

We shall seek the form of $\hat{\Delta}_{ij}$ at temperatures below $T_c$. From Eqs. (1) we obtain the following equation for $\hat{F}_{ij}^+$:

$$(\omega_n^2+\xi^2)\hat{F}_{ij}^+-\sum_{k,l}\hat{\Delta}_{ik}^*\hat{\Delta}_{kl}\hat{F}_{lj}^+=-\hat{\Delta}_{ij}^*. \tag{5}$$

As a consequence of the antisymmetric nature of the electron wave functions, the quantities $\hat{F}_{ij}$ and $\hat{\Delta}_{ij}$ must change sign upon simultaneous interchange of the coordinates and transposition of the spin indices. Here $\hat{\Delta}_{ij} = 0$ if i and j do not denote neighboring planes. The general expression for $\hat{\Delta}_{ij}$ has the following form for neighboring planes:

$$\hat{\Delta}_{ij}=a_{ij}\sigma_y+b_{ij}(\boldsymbol{\sigma}\mathbf{n}_{ij})\sigma_y(i-j), \tag{6}$$

where $\mathbf{n}_{ij}$ is a unit vector which may depend on the number of the plane; $a_{ij}$ and $b_{ij}$ are functions of the numbers used to label the planes, symmetric with respect to interchange of the subscripts.

The first term in Eq. (6) corresponds to pairing with the total spin of the pair equal to zero, and the second corresponds to the total spin being equal to one.

  

Equations (2) and (5) admit the following obvious solutions: a and b are simultaneously equal to zero, or else a or b is separately equal to zero. However, for temperatures smaller than $T_c$ these solutions do not correspond to a minimum of the energy. The smallest free energy corresponds to the solution in which

$$a_{ij}=b_{ij}={}^1/_2\Delta\exp(i\varphi_{ij}),\qquad \mathbf{n}_{ij}=\mathbf{n}. \tag{7}$$

With this choice we have

$$\sum_k \hat{\Delta}_{ik}{}^*\hat{\Delta}_{kl}=-\Delta^2\delta_{il}. \tag{8}$$

We can easily find $F_{ij}$ from Eq. (5). Substituting $F_{ij}$ into Eq. (2), we obtain the condition that determines $\Delta$:

$$1=TV\sum_{\omega_n}\int\frac{d\xi}{\omega_n^2+\xi^2+\Delta^2}. \tag{9}$$

The solution found for $\hat{\Delta}_{ij}$ has a simple form if the Z axis in spin space coincides with the direction of the vector $\mathbf{n}$. In such a representation, the matrix $\hat{\Delta}_{ij}$ is given by

$$\Delta_{ij}=\Delta\begin{pmatrix}0 & \theta(i-j)\\ -\theta(j-i) & 0\end{pmatrix}\exp(i\varphi_{ij}),$$
$$\theta(x)=1,\ x>0;\qquad \theta(x)=0,\quad x<0. \tag{10}$$

This means that an electron with spin directed along the Z axis is paired with an electron having the opposite projection of the spin on the Z axis and located on only one of the two neighboring planes.

## 3. MAGNETIC FIELD PARALLEL TO THE LAYERS

An investigation of paramagnetic effects in an external, homogeneous magnetic field **H** is of interest because of the anomalous nature of the pairing. In the absence of electron hopping between the planes, no diamagnetic currents exist in the quasi-two-dimensional case in a field parallel to the planes. The magnetic field only acts on the spins. At temperatures close to $T_c$, one can obtain equations of the Ginzburg-Landau type from Eqs. (1) and (2) by an expansion in terms of $\Delta$ and H

$$\begin{aligned}a(\tau-2B\mu^2H^2)&=a(3a^2+b^2)B,\\ b\tau\sigma\mathbf{n}_\perp&=b(a^2+3b^2)B\sigma\mathbf{n}_\perp,\\ b(\tau-2B\mu^2H^2)\sigma\mathbf{n}_\parallel&=b(a^2+3b^2)B\sigma\mathbf{n}_\parallel,\end{aligned} \tag{11}$$

where

$$\tau=\frac{T_c-T}{T_c},\qquad B=\frac{7\zeta(3)}{8\pi^2T_c^2}, \tag{12}$$

$\mathbf{n}_\parallel$ is the component of the vector **n** parallel to the field **H**; $\mathbf{n}_\perp$ is the component perpendicular to the field **H**. The state with the least energy corresponds to $\mathbf{n}_\parallel = 0$. For $T < T_c$ we find the following results from Eqs. (11):

$$\begin{cases}a^2=\tau/4B-{}^3/_4\mu^2H^2\\ b^2=\tau/4B+{}^1/_4\mu^2H^2\end{cases}\quad\text{for }\mu^2H^2<\frac{\tau}{3B},$$
$$\begin{cases}a=0\\ b^2=\tau/3B\end{cases}\quad\text{for }\mu^2H^2>\frac{\tau}{3B}. \tag{13}$$

Only the trivial solution exists for $T > T_c$.

The solutions (13) completely determine the behavior of the system near $T_c$ in the presence of a magnetic field, which does not cause diamagnetic currents. In sufficiently strong magnetic fields a = 0. It is physically clear that a sufficiently large field completely suppresses the spinless state at arbitrary temperatures, since the electrons in a pair cannot have opposite spins. The electron spins are parallel in the state with spin one; therefore, one can anticipate that the field does not affect this state. For a = 0, $F_{ij}$ from Eqs. (1) has the form

$$F_{ij}=\prod_{i\le k\le j}(\sigma\mathbf{n}_{ik}\sigma_y)\int_0^{2\pi}F(q)e^{iq(i-j)}\frac{dq}{2\pi}, \tag{14}$$

where

$$F(q)=-\frac{2ib\sin q}{\omega_n^2+(\xi-\mu\sigma\mathbf{H})^2+4b^2\sin^2q}. \tag{15}$$

Substituting $F_{ij}$ from Eqs. (14) and (15) into Eq. (2), we have

$$1=\frac{TV}{\pi}\sum_{\omega_n}\int\frac{\sin^2q\,d\xi\,dq}{\omega_n^2+(\xi+\mu\sigma\mathbf{H})^2+4b^2\sin^2q}$$
$$=\frac{TV}{\pi}\sum_{\omega_n}\int\frac{\sin^2q\,d\xi\,dq}{\omega_n^2+\xi^2+4b^2\sin^2q}. \tag{16}$$

Thus, in the presence of a sufficiently large field H the value of the gap ceases to depend on H, but becomes strongly dependent on the transverse momenta. The gap vanishes for momenta parallel to the planes. One can determine the quantity b(0) at T = 0 from Eq. (16):

$$b(0)=\Delta(0)/\sqrt{e}. \tag{17}$$

A gap exists in the excitation spectrum in the region where $a_{ij}\neq 0$. Therefore, the system undergoes a phase transition at the point where $a_{ij}$ vanishes. The phase transition line between the two superconducting states is determined by the equation

$$1=\frac{TV}{\pi}\sum_{\omega_n}\int\frac{d\xi\,dq\sin^2q}{(\omega_n-i\mu\sigma\mathbf{H})^2+\xi^2+4b^2\sin^2q}. \tag{18}$$

In weak fields the transition temperature is close to $T_c$ and, as is evident from formulas (13), is a quadratic function of H. At low temperatures, in order of magnitude the critical field corresponding to this transition is $\Delta(0)/\mu$.

Formulas (1), (2), and (6) enable us to find the average spin **S** in the magnetic field, which determines the Knight shift:

$$\mathbf{S}={}^1/_2\operatorname{Sp}\sigma G_{ii}(\mathbf{r}_\parallel,\mathbf{r}_\parallel). \tag{19}$$

In a sufficiently strong field, where a = 0, the Knight shift is the same as in the normal metal. In weak fields $a = b = \Delta/2$, and in the linear approximation with respect to the field the average spin **S** is given by

$$\mathbf{S}=\rho\mu\mathbf{H}T\sum_{\omega_n}\int\frac{\xi^2-\omega_n^2}{(\xi^2+\omega_n^2+\Delta^2)^2}d\xi. \tag{20}$$

In expression (20) it is more convenient to carry out the integration over $\xi$ before the summation over $\omega_n$. In this connection a difference is obtained between the average spin in the superconducting and normal metal. The final answer for the spin susceptibility $\chi = \partial S/\partial H$ takes the form

$$\chi=(\chi_s+\chi_n)/2, \tag{21}$$

where $\chi_n = \rho\mu$ is the spin susceptibility of the normal metal and $\chi_s$ is the spin susceptibility of a superconductor with the usual pairing. At T = 0 the value of the spin susceptibility is equal to half the spin susceptibility of the normal metal. Near $T_c$ formula (21) gives

$$\chi=\chi_n(1-\tau). \tag{22}$$

Expression (21) indicates that only half of the "superconducting" electrons does not give a contribution to the spin susceptibility.

## 4. EFFECT OF ELECTRON HOPPING

Hopping of the electrons from plane to plane leads to the appearance of diamagnetic currents even in a field parallel to the planes. The hoppings can be taken into account if the energy of the single-electron state is written in the form

$$\xi(\mathbf{p}, q) = \xi(\mathbf{p}) + 2W(1 - \cos q). \qquad (23)$$

in Eqs. (1). As before the vector $\mathbf{p}$ is parallel to the planes. We assume everywhere that $W \ll \epsilon_F$. The solution $\Delta_{ij} = b_{ij}(\sigma \cdot \mathbf{n}_{ij})\sigma_y(i-j)$ corresponding to pairing with spin one, appeared previously in the presence of a magnetic field. Let us consider what effect a small probability of hopping has on the temperature at which such a solution first appears. In accordance with this, below we assume that $a = 0$. We neglect the nonlinear terms in $\Delta_{ij}$. Near $T_c$ the equation for $\Delta_{ij}$ takes the form

$$[-1+\xi^2(i\nabla+2e\mathbf{A}_\|)^2]\Delta_{ij} + \frac{W^2B}{\tau}(2\Delta_{ij} - \Delta_{i+1,j+1}\exp(-i\chi_{ij}) - \Delta_{i-1,j-1}\exp(i\chi_{ij})) = 0, \qquad (24)$$

where

$$\mathbf{A}_\| = (A_x, A_y), \quad \nabla = \left(\frac{\partial}{\partial x}, \frac{\partial}{\partial y}\right),$$

and $\chi_{ij} = 2eA_z(i, j)d$, where d is the distance between the layers. In Eq. (24) B is determined by formula (12) and $\xi$ denotes the coherence length in the plane. This quantity depends on the temperature and on the mean free path in the usual way.

Let us choose $\mathbf{A}$ in the form

$$\mathbf{A} = (Hy\sin\theta, 0, -Hy\cos\theta) \qquad (25)$$

so that $H_\| = H\cos\theta$ and $H_\perp = H\sin\theta$.

Let us take the solution which doesn't depend on the number of the plane

$$\Delta_{ij} = b(\sigma\mathbf{n})(i-j). \qquad (26)$$

Then we have the following equation for b:

$$\left[-1-\xi^2\frac{d^2}{dy^2} + 4e^2\xi^2H^2\sin^2\theta y^2 + \frac{2W^2B}{\tau}(1-\cos(2eH\cos\theta yd))\right]b = 0. \qquad (27)$$

An analogous equation for the usual pairing in layered superconductors was obtained by Bulaevskiĭ.[2] However, the term allowing for paramagnetic destruction of the superconductivity is absent from Eq. (27), which substantially changes the temperature dependence of the critical field. The condition

$$\tau \ll W^2/T_c^2. \qquad (28)$$

is satisfied in a narrow range near the transition temperature $T_c$ or for a sufficiently large probability of hopping. In this connection small values of y are important in Eq. (27), and it goes over into the equation determining $H_{c2}(\theta)$ in anisotropic superconductors:

$$H_{c2}(\theta) = H_{c2}(\pi/2)\left(\sin^2\theta + \frac{W^2Bd^2\cos^2\theta}{\tau\xi^2}\right)^{-1/2}, \qquad (29)$$

where $H_{c2}(\pi/2)$ is determined by the same formula as in isotropic superconductors:

$$H_{c2}(\pi/2) = \Phi_0/2\pi\xi^2. \qquad (30)$$

Formula (29) shows that near the transition point the field $H_{c2}(\theta)$ is proportional to $\tau$, just as in ordinary isotropic superconductors.

In the opposite limiting case, for $\tau \gg W^2/T_c^2$, the last term in Eq. (27) can be treated as a perturbation for arbitrary fields. The critical field is determined by the component perpendicular to the layers

$$H_{c2}(\theta) = H_{c2}(\pi/2)/\sin\theta. \qquad (31)$$

A field parallel to the layers only leads to a certain reduction of $T_c$. In weak fields this reduction is determined by formula (29) and is proportional to the field. In strong fields,

$$H \gg W\Phi_0/T_c d\xi(0), \qquad (32)$$

the cosine term in Eq. (27) oscillates rapidly and can be neglected. In these fields the transition temperature $T_c(H)$ doesn't depend on the field and is given by

$$T_c(H) = T_c(0)(1-2W^2B). \qquad (33)$$

The electron hoppings are essential in connection with an estimate of the influence of fluctuations on the existence of an order parameter in layered superconductors. In the absence of hopping, the average value of the order parameter would tend to zero. The presence of hopping leads to the result that this value decreases by the amount

$$\frac{\delta\Delta}{\Delta} \sim \frac{T_c}{\varepsilon_F\tau}\ln(T_c/W). \qquad (34)$$

For any reasonable values of W, the fluctuations become important only in a very narrow range near $T_c$.

## 5. QUASI-ONE-DIMENSIONAL SYSTEMS

Let us consider a system of parallel, conducting filaments. As in the preceding case, we assume that the attraction between electrons located on neighboring filaments is the strongest. In such a model there is an instability in the zero-sound channel (the Peierls instability) in addition to an instability in the superconducting channel. We shall assume below that each filament has a large number of conducting bands. In this connection the Peierls instability does not appear, and the system may be described by the usual formulas of the theory of superconductivity.[3] In particular, Eqs. (1) and (2) are valid if the substitution

$$V_{ij} \to nV_{ij}. \qquad (35)$$

is made. The subscripts i and j determine the number of the filament. At a temperature close to $T_c$, one can expand Eq. (2) in powers of $\Delta$ and the Ginzburg-Landau equation can be written down. In this approximation and in the absence of hopping, the expression for the free energy has the form

$$F = \frac{1}{2}n\,\mathrm{Sp}\int\Big[-A\sum_{i,j}\hat\Delta_{ij}\hat\Delta_{ji}^* - \sum_{i,j}C\left(\frac{\partial}{\partial Z}\hat\Delta_{ij}\right)\left(\frac{\partial}{\partial Z}\hat\Delta_{ji}^*\right) + \frac{B}{2}\sum_{i,j,k,l}\hat\Delta_{ij}\hat\Delta_{jk}^*\hat\Delta_{kl}\hat\Delta_{li}^*\Big]dZ, \qquad (36)$$

where $\Delta_{ij}$ is defined by expression (6). The minimum free energy for a square lattice corresponds to the solution

$$a_{ij} = b_{ij} = \frac{\Delta}{2\sqrt{2}}\exp(i\varphi_{ij}), \quad \mathbf{n}_{ij} = \mathbf{n}. \qquad (37)$$

In contrast to the quasi-two-dimensional case considered above, the phases $\varphi_{ij}$ are not arbitrary, but are connected by the relation

$$\varphi_{ij} - \varphi_{jk} + \varphi_{kl} - \varphi_{li} = (2m+1)\pi, \text{ m is an integer}, \qquad (38)$$

where i, j, k, and $l$ are the sites of the elementary lattice enumerated in the order of going around the perimeter.

Expressions (37) and (38) represent the solution of the Gor'kov equations (2) and (5) at arbitrary temperatures. The temperature dependence of the parameter $\Delta$ is determined by the usual equation (9).

Relation (38) can be rewritten in a form which is more convenient for the investigation, if one changes to a new system of coordinates X and Y in which the origin is located in the middle of the side of an elementary square, and the axes are directed parallel to the diagonals of the square. The new coordinate system is rectangular. Any segment, joining two neighboring filaments, is determined by the coordinate of its midpoint. In this notation Eq. (38) takes the following simpler form:

$$\varphi(X,Y)-\varphi\left(X, Y\pm\frac{d}{\sqrt{2}}\right)+\varphi\left(X\pm\frac{d}{\sqrt{2}}, Y\pm\frac{d}{\sqrt{2}}\right) -\varphi\left(X\pm\frac{d}{\sqrt{2}}, Y\right)=(2m+1)\pi, \qquad (39)$$

where d is the period of the lattice.

The general solution of Eq. (39) is written in the following form:

$$\varphi(X,Y)=(2m+1)\pi XY+f_1(X)+f_2(Y), \qquad (40)$$

where $f_1$ and $f_2$ are arbitrary functions.

The influence of a magnetic field can be taken into account in the same way as in the case of layered superconductors. In a sufficiently strong magnetic field, the coefficient $a_{ij}$ in expression (6) for $\Delta_{ij}$ vanishes, but the coefficient $b_{ij}$ doesn't depend on the field.

In order to clarify the role of fluctuations, it is necessary to expand the free energy near its minimum. Here the fluctuations in the modulus of the parameters a and b and in the direction of the vector **n** are three-dimensional. In a model with a large number of bands n, these fluctuations are small in the region not too close to the transition point $\tau \gg n^{-2/3}$, [3] and they will not be taken into consideration below. Only the fluctuations of the phase are important. The free energy has a minimum when the dependence on the coordinates is determined by expression (40). Near this minimum the free energy is given by

$$F=n\int\left[C\Delta^2\left(\frac{\partial\varphi}{\partial Z}\right)^2+\frac{1}{8}B\Delta^4\left(\frac{\partial^2\varphi}{\partial X\,\partial Y}\right)^2\right]dZ\,dX\,dY. \qquad (41)$$

Let us utilize this expression for a calculation of the correlation function

$$\langle\Delta(0)\Delta^*(Z)\rangle=\Delta^2\langle e^{i\varphi(0)-i\varphi(Z)}\rangle=\Delta^2\exp\left(-\frac{\langle(\varphi(0)-\varphi(Z))^2\rangle}{2}\right). \qquad (42)$$

Fluctuations in which the phases of all bonds by a single filament change by the same amount do not violate relation (38) and do not change the free energy (36). Such fluctuations are one-dimensional and lead to an exponential decrease of the correlation function (42).[1)]

The electron hoppings lead to the occurrence of a phase transition into a state with long range order.

## 6. CONCLUSION

The properties of quasi-one-dimensional and layered superconductors essentially depend on the ratio of the hopping amplitude W to the temperature. For $W \gg T_c$ the system is three-dimensional and the difference considered above between such superconductors and ordinary superconductors manifests itself in the value of the spin susceptibility (the Knight shift). The case $W \ll T_c$ is more interesting. Then the quasi-two-dimensional systems may change into the superconducting state for an arbitrarily small value of the hopping probability. At temperatures not very close to $T_c$, the superconducting state with pairs having nonzero spin is not destroyed by an arbitrarily large field. In the quasi-one-dimensional case the direction of the field may be arbitrary, but in the quasi-two-dimensional case it must be parallel to the conducting planes.

The behavior of layered superconductors of $TaS_2$-(pyridine) was investigated in experiment [1] in magnetic fields up to 150 kOe. Such a field, parallel to the layers, did not destroy the superconductivity. The ordinary pairing would have a paramagnetic limit equal to 60 kOe. By analyzing the experiments of [4,5], Bulaevskiĭ showed that the coherence length and the total mean free path are close to one another. Therefore, such stability with regard to the disappearance of superconductivity cannot be a consequence of spin-orbital scattering. The existence in $TaS_2$-(pyridine) of a pairing of electrons located in different planes serves as a possible explanation. Further experimental investigation is required for a final answer to this question.

[1)] Note added in proof (20 November 1974). As noted by B. I. Gal'perin, to whom the authors express their gratitude, the fluctuations of the spin are also one-dimensional.


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