MAGNETIC PROPERTIES OF THIN METALLIC FILMS

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The topic treated is the change of the magnetic properties of a metallic film, placed in a magnetic field perpendicular to it, when the film thickness is so small that the quantization of the motion of the electrons in the direction of its smallest dimension becomes important. The magnetic properties of the film are then determined by five characteristic energies: the temperature, the Fermi energy, the distance between Landau levels, their spin splitting, and the distance between dimensional-quantization levels. Cases are considered in which these energies have various ratios. The entire investigation is carried out under the assumption of an isotropic quadratic electron spectrum.

THE magnetic properties of an electron gas in a metal change significantly if the specimen under consideration is a film whose thickness is so small that the motion of the electrons in the direction of its smallest dimension becomes quantized. We consider the case in which the magnetic field **H** is perpendicular to the surface of the film. (The case of a magnetic field lying in the plane of the film was considered by Kosevich and Lifshitz[1]. The general formula for the quasiclassical case was obtained by them in[2].) We further restrict ourselves to consideration of a quadratic isotropic electron spectrum. In this case, the energy of an electron has the form

$$\varepsilon_{nl} = (2n+1)\mu H \pm \mu_0 H + \frac{\pi^2\hbar^2 l^2}{2md^2} \tag{1}$$

(μ_0 is the Bohr magneton, $\mu = \mu_0 m_0/m$, m is the effective mass of the electron, n and l are integers, and the signs ± correspond to the two orientations of the spin).

The thermodynamic potential Ω can be written in the form

$$\Omega = -T\frac{eH}{2\pi\hbar c}\frac{V}{d}(\Omega^+ + \Omega^-), \tag{2}$$

where

$$\Omega^\pm = \sum_{l=1}^{\infty}\sum_{n=0}^{\infty}\ln\left[1+\exp\frac{1-n\eta-\eta^\pm-\lambda^2 l^2}{\tau}\right]. \tag{3}$$

The characteristic energies of the problem are: the distance $2\mu H$ between Landau levels, the energy $\mu_0 H$ of the spin in the magnetic field, the temperature T, and the energy $\epsilon_0 = \pi^2\hbar^2/2md^2$ that determines the distance between dimensional-quantization levels. These are here written in dimensionless form, in units of the chemical potential ζ:

$$\eta = 2\mu H/\zeta, \quad \eta_0 = 2\mu_0 H/\zeta, \quad \eta^\pm = {}^1\!/\!_2(\eta \pm \eta_0),$$
$$\tau = T/\zeta, \quad \lambda^2 = \pi^2\hbar^2/2m\zeta d^2.$$

The values of η, η_0, $\eta^\pm$, and τ will be supposed small. The following additional symbols will be used hereafter: $\zeta_0 = (\hbar^2/2m)(3\pi^2 n)^{2/3}$, the level of the chemical potential for a large specimen in the absence of a magnetic field, and $\lambda_0^2 = \pi^2\hbar^2/2m\zeta_0 d^2$.

On designating by $\Omega_l^\pm = \sum_{n=0}^{\infty}\Omega_{nl}^\pm$ the expression under the sign of summation over l in (3) and on applying the Poisson summation formula, we get

$$\Omega_l^\pm = \frac{1}{2}\ln\left(1+\exp\frac{1-\eta^\pm-\lambda^2 l^2}{\tau}\right) + I_0 + 2\sum_{n=1}^{\infty} I_n, \tag{4}$$

where

$$I_n = \int_0^\infty \ln\left(1+\exp\frac{1-x\eta-\eta^\pm-\lambda^2 l^2}{\tau}\right)\cos(2\pi nx)\,dx \quad (n=0,1,2\ldots).$$

For $n \neq 0$ we have[3]

$$I_n = \frac{\eta}{4\pi^2 n^2\tau}\left\{\left[1+\exp\frac{\lambda^2 l^2+\eta^\pm-1}{\tau}\right]^{-1}\right.$$
$$\left. - \operatorname{Re}\exp\left(2\pi in\frac{1-\eta^\pm-\lambda^2 l^2}{\eta}\right)\int_{(\lambda^2 l^2+\eta^\pm-1)/\tau}^{\infty}\frac{e^z e^{2\pi in\tau z/\eta}}{(e^z+1)^2}dz\right\}. \tag{5}$$

When $\tau \gg \eta$, the integral is equal to zero because of the rapid oscillations of the integrand. When $\eta \gg \tau$, we get

$$\int_{(\lambda^2 l^2+\eta^\pm-1)/\tau}^{\infty}\frac{e^z e^{2\pi in\tau z/\eta}}{(e^z+1)^2}dz = \begin{cases}1; & \eta^\pm+\lambda^2 l^2 < 1\\ 0; & \eta^\pm+\lambda^2 l^2 > 1\end{cases}.$$

Various ratios are possible between the characteristic energies mentioned above. We shall consider three cases:

$$\lambda_0^2 < \eta, \quad \eta < \lambda_0^2 < 1, \quad \lambda_0 > 1.$$

In each of these, consideration will be given to the high-temperature ($\tau \gg \eta$) and low-temperature ($\eta \gg \tau$) regions.

1. THE CASE $\lambda_0^2 < \eta$

In this case $\lambda_0 \ll 1$, and we shall, first, neglect the dependence of ζ on H and d and set $\lambda = \lambda_0$; and, second, expand all the expressions obtained in powers of λ_0, keeping only terms of the two lowest orders. We shall hereafter denote by $l_0^\pm$ the value of l for which $\eta^\pm + \lambda_0^2 l^2 = 1$. Since $l_0^\pm \gg 1$, we shall not commit a great error by treating it as an integer.

The first term in $\Omega^\pm$ we calculate as follows:

$$\frac{1}{2}\sum_{l=1}^{\infty}\ln\left(1+\exp\frac{1-\eta^\pm-\lambda_0^2 l^2}{\tau}\right) \approx \frac{1}{2}\left\{\sum_{l=1}^{l_0^\pm}\left(\frac{1-\eta^\pm-\lambda_0^2 l^2}{\tau}\right.\right.$$
$$\left.\left.+\exp\frac{\lambda_0^2 l^2+\eta^\pm-1}{\tau}\right) + \sum_{l=l_0^\pm}^{\infty}\exp\frac{1-\eta^\pm-\lambda_0^2 l^2}{\tau}\right\}. \tag{6}$$

The sums of exponentials can be neglected; for the largest terms in them, corresponding to $l = l_0^\pm$, are equal to unity and the terms decrease rapidly with separation of l from $l_0^\pm$, whereas the first term, containing a power of λ_0, has the order $1/\tau \gg 1$. The expression (6) then becomes equal to

$$\frac{1}{3\tau\lambda_0}\left(1-\frac{3}{2}\eta^\pm\right)-\frac{1}{4\tau}(1-\eta^\pm).$$

Here and later (with the exception of the oscillating terms), we shall keep only the lowest powers of η, η_0, and $\eta^\pm$; these lead in Ω to terms of not higher than the second order in H.

We have further

$$\sum_{l=1}^{\infty} I_0 \approx \sum_{i=1}^{l_0^\pm}\frac{(1-\eta^\pm-\lambda_0^2 l^2)^2}{2\tau\eta} \approx \tag{7}$$

$$\approx \frac{1}{2\tau\eta}\left\{\frac{1}{\lambda_0}\left(\frac{8}{15}-\frac{4}{3}\eta^\pm+\eta^{\pm 2}\right)-\frac{1}{2}(1-\eta^\pm)^2\right\}.$$

It can be shown that to the accuracy with which we have calculated (6), we may, in summing the first terms in (5) over l, neglect the exponentials in the denominators for $l < l_0^\pm$. Then the contribution to $\Omega^\pm$ from the nonoscillatory parts of I_n is equal to

$$\frac{\eta}{2\pi^2\tau} l_0^\pm \sum_{n=1}^{\infty}\frac{1}{n^2} \approx \frac{\eta}{12\tau\lambda_0}.$$

We have finally for the case $\tau \gg \eta$

$$\Omega = -T\frac{eH}{2\pi\hbar c}\frac{V}{d}\left\{\frac{1}{\tau\lambda_0}\left[\frac{8}{15}\frac{1}{\eta}+\frac{1}{4}\eta\left(\frac{\eta_0^2}{\eta^2}-\frac{1}{3}\right)\right]\right.$$

$$\left.-\frac{1}{\tau}\left[\frac{1}{2\eta}+\frac{1}{8}\eta\left(\frac{\eta_0^2}{\eta^2}-1\right)\right]\right\}. \tag{8}$$

On going over to ordinary notation and differentiating with respect to H, we get the magnetic susceptibility

$$\chi = \frac{\sqrt{2}}{\pi^2}\frac{m^{3/2}\mu^2\zeta_0^{1/2}}{\hbar^3}\left(\frac{m^2}{m_0^2}-\frac{1}{3}\right)-\frac{1}{2\pi}\frac{m\mu^2}{\hbar^2 d}\left(\frac{m^2}{m_0^2}-1\right). \tag{9}$$

In the case $\eta \gg \tau$, it is necessary also to sum the oscillatory parts of I_n:

$$\Omega_{\rm osc}^\pm = 2\sum_{n=1}^{\infty}(\Omega_{\rm osc}^\pm)_n, \tag{10}$$

where

$$(\Omega_{\rm osc}^\pm)_n = -\frac{\eta}{4\pi^2 n^2\tau}\,{\rm Re}\left\{\exp\left[\mp\frac{2\pi i n}{\eta}(1-\eta^\pm)\right]\right.$$

$$\left.\times\sum_{l=1}^{l_0^\pm}\exp\left[-\frac{2\pi i n\lambda_0^2 l^2}{\eta}\right]\right\} \tag{11}$$

As a preliminary step, we shall calculate $\sum_{k=1}^{S} e^{-\alpha k^2}$, where $|\alpha| < 1$. In the calculation, we shall keep terms of the two lowest orders in α:

$$\sum_{k=1}^{s} e^{-\alpha k^2} = s-\alpha\sum_{k=1}^{s}k^2+\frac{\alpha^2}{2!}\sum_{k=1}^{s}k^4-\ldots \tag{12}$$

In each sum, we shall keep the two highest powers of s:

$$\sum_{k=1}^{s}k^m \approx \frac{s^{m+1}}{m+1}+\frac{s^m}{2}. \tag{13}$$

Retention of additional terms leads, as can be shown, to terms of higher order in α in the answer. On substituting (13) into (12), we get

$$\sum_{k=1}^{s} e^{-\alpha k^2} \approx \sum_{m=1}^{\infty}\frac{s^{2m-1}(-1)^{m-1}}{(2m-1)(m-1)!}\alpha^{m-1}$$

$$+\frac{1}{2}\sum_{m=1}^{\infty}(-1)^m\frac{s^{2m}\alpha^m}{m!} = \frac{1}{2}\sqrt{\frac{\pi}{\alpha}}\,\Phi(s\sqrt{\alpha})+\frac{1}{2}(e^{-\alpha s^2}-1), \tag{14}$$

where $\Phi(x)$ is the probability integral.

By using (14), we get

$$\sum_{i=1}^{l_0^\pm}\exp\left[-\frac{2\pi i n\lambda_0^2 l^2}{\eta}\right] = \frac{1}{2}\sqrt{\frac{\eta}{2in}}\frac{1}{\lambda_0}\Phi\left(\left[\frac{2\pi i n}{\eta}(1-\eta^\pm)\right]^{1/2}\right)$$

$$+\frac{1}{2}\left(\exp\left[-\frac{2\pi i n}{\eta}(1-\eta^\pm)\right]-1\right). \tag{15}$$

Since $\eta \ll 1$, we may set $\Phi(\ldots) = 1$. As a result we get

$$(\Omega_{\rm osc}^\pm)_n = -\frac{\eta^{3/2}}{8\sqrt{2}\,\pi^2 n^{5/2}\tau\lambda_0}\,{\rm Re}\exp\left\{\frac{2\pi i n}{\eta}(1-\eta^\pm)-\frac{\pi i}{4}\right\}$$

$$-\frac{\eta}{8\pi^2 n^2\tau}{\rm Re}\left(1-\exp\left[\frac{2\pi i n}{\eta}(1-\eta^\pm)\right]\right). \tag{16}$$

We have finally for $\eta \gg \tau$

$$\Omega = -\frac{eH}{\pi\hbar c}\frac{V}{d}\left\{\frac{\zeta_0^{3/2}d\sqrt{2}\,m^{1/2}}{2\pi\hbar}\left[\frac{4}{15}\frac{\zeta_0}{\mu H}+\frac{\mu H}{2\zeta_0}\left(\frac{m^2}{m_0^2}-\frac{1}{3}\right)\right]\right.$$

$$-\zeta_0\left[\frac{\zeta_0}{\mu H}+\frac{1}{4}\frac{\mu H}{\zeta_0}\left(\frac{m^2}{m_0^2}-\frac{1}{3}\right)\right]-\frac{\mu^{3/2}H^{3/2}m^{1/2}d}{\sqrt{2}\,\pi^3\hbar}$$

$$\times\sum_{n=1}^{\infty}n^{-5/2}\cos\left(\frac{\pi n\zeta_0}{\mu H}-\pi n-\frac{\pi}{4}\right)\cos\left(\pi n\frac{m}{m_0}\right)$$

$$\left.+\frac{\mu H}{2\pi^2}\sum_{n=1}^{\infty}n^{-2}\cos\left(\frac{\pi n\zeta_0}{\mu H}-\pi n\right)\cos\left(\pi n\frac{m}{m_0}\right)\right\}, \tag{17}$$

$$M = \left\{\frac{\sqrt{2}}{\pi^2}\frac{m^{3/2}\mu^2\zeta_0^{1/2}}{\hbar^3}\left(\frac{m^2}{m_0^2}-\frac{1}{3}\right)\left(1-\frac{\pi\hbar}{\sqrt{2m\zeta_0}d}\right)\right\}H$$

$$-\frac{\sqrt{2}}{\pi^3}\frac{\mu^{3/2}H^{1/2}m^{3/2}\zeta_0}{\hbar^3}\left\{\sum_{n=1}^{\infty}n^{-3/2}\sin\left(\frac{\pi n\zeta_0}{\mu H}-\pi n-\frac{\pi}{4}\right)\cos\left(\pi n\frac{m}{m_0}\right)\right.$$

$$\left.-\frac{\pi}{2\sqrt{2}}\frac{\hbar}{(m\mu H)^{1/2}d}\sum_{n=1}^{\infty}n^{-1}\sin\left(\frac{\pi n\zeta_0}{\mu H}-\pi n\right)\cos\left(\pi n\frac{m}{m_0}\right)\right\}. \tag{18}$$

From this expression, as also from (9), it is seen that the result in the usual expression for the magnetization of an electron gas (see, for example,[3]), with additional terms due to the dimensional quantization. The correction to the constant part of the magnetization has the order $\hbar/(m\zeta_0)^{1/2}d$. For $\lambda_0^2 < \eta$, this quantity is very small. Considerably more interesting is the change of the oscillatory part. It has the form of additional oscillations of the same frequencies, but shifted in phase by $\frac{3}{4}\pi$ and having a relative amplitude of order $\hbar/(m\mu H)^{1/2}d$.

2. THE CASE $\eta < \lambda_0^2 < 1$

By virtue of the right-hand inequality, we may first set $\lambda = \lambda_0$. In the present case the high-temperature limit is not of interest, since it leads to results obtained in the preceding section. This is due to the fact that their derivation was based only on the suppositions that $\tau \gg \eta$ and that $\lambda = \lambda_0$ and did not depend on the ratio between η and λ_0.

Therefore we deal only with the oscillatory part of the thermodynamic potential for $\eta \gg \tau$. We have

$$\Omega = \frac{e\mu H^2 V}{\pi^3\hbar c d}\sum_{l=1}^{l_0}\sum_{n=1}^{\infty}\frac{1}{n^2}\cos\left(2\pi n\cdot\frac{1-\lambda_0^2 l^2-\eta/2}{\eta}\right)\cos\left(\pi n\frac{\eta_0}{\eta}\right), \tag{19}$$

Here

$$l_0 = [\sqrt{2m\zeta_0}d/\pi\hbar], \tag{20}$$

the square brackets denote the integer part of the number.

The magnetization can be written in the form

$$M = -\frac{2m\mu}{\pi^2\hbar^2 d}\sum_{l=1}^{l_0}\left\{\left(\zeta_0 - \frac{\pi^2\hbar^2}{2md^2}l^2\right)\cdot\right.$$

$$\left.\times\sum_{n=1}^{\infty}\frac{1}{n}\sin 2\pi n\left(\frac{\zeta_0 - \pi^2\hbar^2 l^2/2md^2}{2\mu H} - \frac{1}{2}\right)\cos\left(\pi n\frac{m}{m_0}\right)\right\}. \tag{21}$$

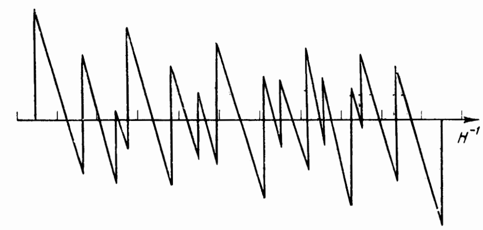
For semimetals—where, as a rule, $m \ll m_0$—, the cosine can be replaced by unity, and the series in n is easily summed:

$$M = -\frac{m\mu}{\pi\hbar^2 d}\sum_{l=1}^{l_0}(\zeta_0 - \pi^2\hbar^2 l^2/2md^2)\cdot$$

$$\times\left\{1 - 2\,\mathrm{Rem}\left(\frac{\zeta_0 - \pi^2\hbar^2 l^2/2md^2}{2\mu H} - \frac{1}{2}\right)\right\}, \tag{22}$$

where Rem $x = x - [x]$. It is seen that the oscillatory part of M is formed by superposition of l_0 functions of saw-tooth form, periodic in $1/H$ with frequencies $(\zeta_0 - \pi^2\hbar^2 l^2/2md^2)/2\mu$ ($l = 1, 2, \ldots, l_0$). The picture of the oscillations for $l_0 = 3$ is shown in the figure.

If the inequality $m \ll m_0$ does not hold, then by transforming the expression under the sign of summation over n, in (21), to a sum of sines, we see that there occurs an additional splitting of the oscillations, connected with the spin of the electron.



One division of the axis corresponds to a period of the oscillation in a bulk specimen.

3. THE CASE $\lambda_0 > 1$

In the circumstance it is necessary to take into account the dependence of ζ on H and d. At not too high temperatures ($T < \epsilon_0$), the energy levels described by quantum numbers $l \geq 2$ are practically empty. Therefore we omit the summation over l in the formula for Ω. For $\tau \gg \eta$ we get

$$\Omega = -T\frac{eH}{2\pi\hbar c}\frac{V}{d}\left(\frac{T}{2\mu H}\left\{\mathcal{F}_1\left(\frac{\zeta - \varepsilon_0 - \mu H - \mu_0 H}{T}\right)\right.\right.$$

$$\left.+\mathcal{F}_1\left(\frac{\zeta - \varepsilon_0 - \mu H + \mu_0 H}{T}\right)\right\} + \frac{1}{2}\left\{\ln\left(1 + \exp\frac{\zeta - \varepsilon_0 - \mu H - \mu_0 H}{T}\right)\right.$$

$$\left.\left.+\ln\left(1 + \exp\frac{\zeta - \varepsilon_0 - \mu H + \mu_0 H}{T}\right)\right\}\right) \tag{23}$$

Here

$$\mathcal{F}_1(x) = \int_0^\infty \frac{y\,dy}{1 + \exp(x - y)}$$

is the Fermi integral. In the lowest order with respect to $\mu H/T$ and $\mu_0 H/T$, we get

$$\chi = \frac{e\mu^2}{2\pi\hbar c d}\left(\frac{m^2}{m_0^2} - \frac{1}{3}\right)\left(1 + \exp\frac{\varepsilon_0 + \mu H - \zeta}{T}\right)^{-1}. \tag{24}$$

This expression contains the unknown quantity $\zeta(H, d)$. To eliminate it, we use the equation $n = -V^{-1}(\partial\Omega/\partial\zeta)_{T,H}$ (n = electron concentration). In the lowest order in $\mu H/T$, we get

$$n = T\frac{e}{2\pi\hbar c\mu d}\ln\left(1 + \exp\frac{\zeta - \varepsilon_0 - \mu H}{T}\right). \tag{25}$$

On comparing (24) and (25), we have finally

$$\chi = \frac{m\mu^2}{\pi\hbar^2 d}\left(\frac{m^2}{m_0^2} - \frac{1}{3}\right)\left\{1 - \exp\left(-\frac{\pi\hbar^2 nd}{mT}\right)\right\}. \tag{26}$$

If in this formula we let $T \to \infty$, we get the known expression for the susceptibility of a nondegenerate electron gas.

In the low-temperature limit, when T is less than the distance between any energy levels of the spectrum, the thermodynamic characteristics of the system are most simply derived by a simple calculation of the energy of the Ground state.

Each energy level is degenerate, with multiplicity

$$\frac{eH}{2\pi\hbar c}\frac{V}{d} = pH.$$

The number of completely filled levels will be

$$s = \left[\frac{N}{pH}\right] = \left[\frac{2\pi\hbar cdn}{eH}\right] \tag{27}$$

The energy of the system is

$$E = \begin{cases} N\varepsilon_0 + \mu H\left\{N(s+1) - pH\frac{s^2}{2} - pHs\right\} - \mu_0 H(N - pHs), & s - \text{even}, \\ N\varepsilon_0 + \mu H\left\{Ns - pH\frac{s^2}{2} + pH\frac{s}{2}\right\} - \mu_0 H\{(s+1)pH - N\}, & s - \text{odd} \end{cases} \tag{28}$$

Since we are considering the case $T \to 0$, we may set the free energy F equal to E. We have

$$M = -\frac{1}{V}\left(\frac{\partial F}{\partial H}\right)_N = \begin{cases} -\mu\{n(s+1) - p'H(s+1)^2 + p'H\} - \mu_0(2p'Hs - n), & s - \text{even} \\ -\mu\{ns - p'Hs^2 + p'H\} + \mu_0\{2(s+1)p'H - n\}, & s - \text{odd} \end{cases} \tag{29}$$

Here $p' = p/V$.

It is seen that the expression for M, along with terms proportional to H, contains terms independent of H (of course only in the range $\mu H > T$), proportional to the electron concentration. The value of M undergoes a jump corresponding to a change of s by unity, every time that $n/p'H = 2\pi\hbar cdn/eH$ assumes an integral value. These jumps cease when $H \geq H_{max} = 2\pi\hbar cdn/e$, after which $M = (\mu_0 - \mu)n$. For Bi with $d \sim 10^{-6}$ cm and $n \sim 2 \times 10^{17}$ cm^{-3}, this value is $H_{max} \sim 7 \times 10^4$ Oe.

In the case $\mu_0 H < T < \mu H$, which can occur for semimetals, the last terms in (29), connected with electron spin, disappear.

4. OTHER CASES

Besides the cases considered above, in semimetals there can arise a situation in which the energy of the Landau quantization exceeds the Fermi energy, calculated in the absence of a magnetic field.

At the lowest temperature, less than all the characteristic energies, two cases must be distinguished: $\epsilon_0 < \mu_0 H$ and $\epsilon_0 > \mu_0 H$. In the first of these, the energy of the system is

$$E = N(\mu - \mu_0)H + pH\frac{s(s+1)(2s+1)}{6}\varepsilon_0 + (N - pHs)(s+1)^2\varepsilon_0 \tag{30}$$

and therefore the magnetic moment is

$$M = (\mu_0 - \mu)n + p'(2/3 s^3 + 3/2 s^2 + 5/6 s). \tag{31}$$

It is seen that M is independent of H and undergoes a jump upon change of s by unity; this is similar to the case described by the expression (29).

When $\epsilon_0 > \mu_0 H$, the spin sublevels may be considered degenerate, the multiplicity p of the degeneracy is doubled, and in (31) the term $\mu_0 n$ must be dropped. If $\mu_0 H < T < \epsilon_0$, then we get the same situation. Finally, if $\epsilon_0 < T < \mu H$, then the dimensional quantization ceases to have an effect, and the magnetization is described by the usual formulas, with allowance for the dependence of ζ on H (see, for example,[4]).

Until now it has always been assumed that $T \ll \zeta$; but for semimetals the degeneracy temperature is very low, and the nondegenerate state is also of interest. It is easy to show, however, that in a film perpendicular to the magnetic field, the dimensional quantization will not change the magnetic properties of a nondegenerate electron gas. In fact, in the classical statistics the free energy consists of two terms, of which one is determined by the energy of the electrons on the Landau levels, and the second by the translational motion along the field. The dimensional quantization changes only the second term; but it is independent of H, and upon differentiation it drops out.

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