SOVIET PHYSICS JETP VOLUME 17, NUMBER 2 AUGUST, 1963

QUANTUM STATISTICAL THEORY OF MAGNETIC RESONANCE IN SYSTEMS WITH STRONG EXCHANGE INTERACTION

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Submitted to JETP editor June 8, 1962

J. Exptl. Theoret. Phys. (U.S.S.R.) **44**, 514–521 (February, 1963)

From the quantum equation for the density matrix, equations are derived which characterize the behavior of magnetization of a system with strong exchange interaction in the high-temperature region ($T \gg T_C$, where T_C is the Curie temperature). These equations differ from the phenomenological equations previously derived [1-6] by some additional terms that take into account the variation of the mean exchange-interaction energy during saturation. It is shown that in accordance with the experimental data [7] the shape of the resonance absorption line is of the Lorentz type and that its width at saturation depends weakly on the temperature. The equations predict a shift of the center of the absorption line and a change in its width with variation of the constant magnetic field. Because of the condition $T \gg T_C$ the equations proposed can be employed only to describe magnetic resonance in systems with a sufficiently low Curie temperature (crystalline free radicals, some ferrites, antiferromagnetic substances, etc.).

AT the present time, phenomenological equations given by various authors [1-6] are used in the analysis of the behavior of the magnetization of ferromagnetic substances. In the majority of cases these equations are modifications of an equation first derived by Landau and Lifshitz, [1] which is in good agreement with the experimental data in the low-temperature region. All of these equations are equations only for the components of magnetization of the spin system. It has been subsequently shown, however, [7-9] that it is necessary to take into account the variation of the mean exchange energy during saturation for a correct interpretation of the experimental data in the region $T \gg T_C$. It is obvious that equations for the magnetization components only cannot account for this variation.

Our task is to obtain the magnetic resonance equations directly from the rigorous density matrix equation. In our case, this equation has the form

$$\hbar \partial\rho(t)/\partial t = -i\,[-\hbar\omega_0 \hat{S}_z + (\mu H_1/2s)\,(\hat{S}^1 e^{i\omega t} + \hat{S}^{-1} e^{-i\omega t}) + \hat{H}_{dip} + \hat{H}_{ex}, \rho(t)],$$

$$\hat{H}_{dip} = g^2\hbar^2 \sum_{i>k} \left(\frac{\hat{\mathbf{s}}_i \hat{\mathbf{s}}_k}{r_{ik}^3} - 3\,\frac{(\hat{\mathbf{s}}_i \mathbf{r}_{ik})(\hat{\mathbf{s}}_k \mathbf{r}_{ik})}{r_{ik}^5} \right),$$

$$\hat{H}_{ex} = -\frac{1}{2} \sum_{i>k} J_{ik} \hat{\mathbf{s}}_i \hat{\mathbf{s}}_k. \tag{1}$$

Here, $\hat{S}_x$, $\hat{S}_y$, $\hat{S}_z$ are the spin-component operators of the total spin, $\hat{S}^{\pm 1} = \hat{S}_x \pm i\hat{S}_y$, $\hat{\mathbf{s}}_i$ is the operator of a spin located on the i-th lattice site, $\mathbf{r}_{ik}$ is a vector connecting the i-th and k-th lattice sites, J_{ik} is the exchange integral, ω and H_1 are the frequency and amplitude of the alternating magnetic field, $\omega_0 = \mu H_0/\hbar s$, H_0 is the constant magnetic field, and μ is the magnetic moment of the particle. The z axis is taken parallel to the constant magnetic field. We shall consider the case in which $H_{ex} \gg H_{dip}, \hbar\omega_0 S_z$. In the solution of Eq. (1) we shall use the method developed by one of the authors. [10,11]

In order to clarify the nature of the processes which take place in the spin system under the influence of a high-frequency magnetic field, it will be convenient to transform to a rotating system of coordinates. This transition is accomplished by means of the transformation [12,13]

$$\rho(t) = \exp(i\omega \hat{S}_z t)\,\rho'(t)\exp(-i\omega \hat{S}_z t), \tag{2}$$

where $\rho'(t)$ is the density matrix in the rotating coordinate system. Substituting Eq. (2) into Eq. (1), we obtain

$$\partial\rho'(t)/\partial t = -i\hbar^{-1}[\hbar\Delta\hat{S}_z + \mu H_1 \hat{S}_x + \hat{H}_{ex} + \hat{V}(t), \rho'(t)],$$

$$\hat{V}(t) = \sum_{m=-2}^{+2} \hat{H}^m e^{im\omega t}. \tag{3}$$

Here, $\Delta = \omega - \omega_0$, $\hat{H}^m$ is the part of the dipole interaction that causes transitions between the levels of the system with a change of m units in the z component of the total spin.

In order to determine the form of the density matrix, we now transform Eq. (3) into the interaction representation

$$\rho'(t) = \exp(-i\hat{H}t/\hbar)\,\rho''(t)\exp(i\hat{H}t/\hbar), \qquad (4)$$

where $\hat{H} = \hat{H}_{ex} + \mu\hat{\mathbf{S}}\cdot\mathbf{H}_{eff}/s$. Here $\mathbf{H}_{eff}$ is the effective magnetic field in the rotating coordinate system, the z component of which equals $-(H_0 - \hbar\omega/\mu)$, and the x component equals $-H_1$. (The y component is zero.) For $\rho''(t)$ we obtain

$$\begin{gathered}\partial\rho''(t)/dt = -i\hbar^{-1}[\hat{V}'(t), \rho''(t)],\\ \hat{V}'(t) = \exp(i\hat{H}t/\hbar)\,\hat{V}(t)\exp(-i\hat{H}t/\hbar).\end{gathered} \qquad (5)$$

We now make use of Zwanzig's method, [10,14] the essence of which is the application of the projection operator $\hat{P}$. We separate the density matrix into two parts:

$$\rho'' = \rho_1'' + \rho_2'', \qquad \rho_1'' = \hat{P}(t)\,\rho'', \qquad \rho_2'' = (1 - \hat{P}(t))\,\rho''.$$

Here $\hat{P}(t)$ is a projection operator that extracts from the density matrix ρ'' that part which is diagonal in the representation in which H_{ex} and the operator of the projection of the total spin on the instantaneous magnetization direction are simultaneously diagonal. Integrating Eq. (5) and then multiplying this equation on the left by $\hat{P}(t)$ and $1 - \hat{P}(t)$ respectively, we obtain

$$\rho_1''(t+\tau) - \rho_1''(t) = -\frac{i}{\hbar}\int\limits_t^{t+\tau} dt'\,\hat{P}(t)\,[\hat{V}'(t'), \rho_2''(t'')], \qquad (6)$$

$$\begin{aligned}\rho_2''(t) = &-\frac{i}{\hbar}\int\limits_0^t dt'\,[\hat{V}'(t'), \rho_1''(t')]\\ &-\frac{i}{\hbar}\int\limits_0^t dt'\,(1-\hat{P}(t))\,[\hat{V}'(t'), \rho_2''(t')].\end{aligned} \qquad (7)$$

In Eq. (6) we have eliminated a term containing $\hat{P}(t)[\hat{V}'(t'), \rho_1''(t')]$, which vanishes because the diagonal matrix elements of the operator $[\hat{V}'(t'), \rho_1''(t')]$ vanish. For the same reason a similar term has been eliminated from the right side of Eq. (7).

For the following discussion it is now convenient, having fixed the moment of time t, to direct the z axis along the instantaneous magnetization vector. In order to find a solution to the system (6) and (7), we assume that in the "instantaneous" coordinate system we have thus chosen the nondiagonal part of the density matrix is much less than the diagonal part, i.e., $\rho_2'' \ll \rho_1''$. In this case, neglecting the second term on the right-hand side of Eq. (7), we obtain for ρ_2''

$$\rho_2'' = -\frac{i}{\hbar}\int\limits_0^t dt'\,[\hat{V}'(t'), \rho_1''(t')]. \qquad (8)$$

We note that in this expression $\rho_1''(t')$ is a slowly varying function of t'.

The right-hand side of Eq. (8) is easily evaluated by direct calculation, just as was done in [11]. The result is $\rho_2'' \approx \rho_1'' H_{dip}/H_{ex}$, which confirms the validity of the assumption $\rho_2''(t) \ll \rho_1''(t)$ we made earlier.

Regarding the diagonal part of the density matrix $\rho_1''(t)$, it should obviously have the following property because of the homogeneity of the macroscopic system under consideration:

$$\rho_1''(t) = \rho_{11}''(t)\,\rho_{12}''(t),$$

where $\rho_{11}''(t)$ and $\rho_{12}''(t)$ are the density matrices of any two arbitrary spin-systems resulting from the division of the system into two arbitrary macroscopic parts. But the most general form for the diagonal density matrix that possesses this property is [15]

$$\rho_1''(t) = C\exp[\sigma(t)\hat{S}_z + \delta(t)\hat{H}_{ex}], \qquad (9)$$

where C is a normalization constant.

This form for the principal part of the density matrix is also easily understood on the basis of general physical principles. In fact, since the exchange interaction in our problem far exceeds all other interactions existing in the spin system considered, it is natural to assume that thermodynamic equilibrium will prevail at any instant in our system during any relaxation process effected by the weak dipole interaction, owing to the strong exchange interaction, and the density matrix will in the general case have the form (9) accurately to terms of order H_{dip}/H_{ex}. This situation is completely analogous to that which arises, for example, in the analysis of a vessel of gas that interacts weakly with the surrounding medium, resulting in its behavior being described at each moment of time by an equilibrium distribution function of the Boltzman type.

If we now direct the z axis along the direction of the constant magnetic field, then the principal part of the density matrix will obviously take the form

$$\rho_1(t) = C\exp[\alpha(t)\hat{S}_z + \beta(t)\hat{S}_x + \gamma(t)\hat{S}_y + \delta(t)\hat{H}_{ex}]. \qquad (10)$$

We return now to the rotating system of coordinates, using the transformation (4). Applying this transformation to Eq. (8), we obtain

$$\rho_2'(t) = -\frac{i}{\hbar}\int\limits_0^t e^{-i\hat{H}(t-t')/\hbar}\,[\hat{V}(t'), \rho_1'(t')]\,e^{i\hat{H}(t-t')/\hbar}\,dt'. \qquad (11)$$

It is not difficult to see that applying the transformation (4) to $\rho_1''(t)$ does not alter its basic form and leads only to a change in the coefficients standing in front of the operators S_x, S_y, and S_z. Therefore, in order not to introduce new symbols we shall continue to write the principal part of the density matrix in the rotating system in the same form as $\rho_1''(t)$:

$$\rho_1'(t) = C \exp\,[\alpha(t)\hat{S}_z + \beta(t)\hat{S}_x + \gamma(t)\hat{S}_y + \delta(t)\hat{H}_{ex}]. \tag{12}$$

Considering that $\mu \mathbf{S}\cdot\mathbf{H}_{\mathrm{eff}} \ll H_{\mathrm{ex}}$, we can replace $\hat{H}$ by $\hat{H}_{\mathrm{ex}}$ in the exponents in Eq. (11). Keeping in mind that $\rho_1'(t)$ commutes with $\hat{H}_{\mathrm{ex}}$, we rewrite Eq. (11) in the form

$$\rho_2'(t) = -\frac{i}{\hbar}\int_0^t dt'\,[\hat{V}''(t,t'), \rho_1'(t')],$$

$$\hat{V}''(t,t') = \exp(-i\hat{H}_{ex}(t-t')/\hbar)\,\hat{V}(t')\exp(i\hat{H}_{ex}(t-t')/\hbar). \tag{13}$$

We now calculate $\partial H_{\mathrm{ex}}/\partial t$ and $\partial S_{x,y,z}/\partial t$ in the rotating coordinate system, using Eq. (3), (12), and (13). Consider, for example, $\partial H_{\mathrm{ex}}/\partial t$. It is easy to see that the expression for $\partial H_{\mathrm{ex}}/\partial t$ can be represented in the form

$$\frac{\partial H_{ex}(t)}{\partial t} = -\frac{1}{\hbar^2}\int_0^t dt'\,\mathrm{Sp}\,\hat{H}_{ex}[\hat{V}(t)\,[\hat{V}''(t,t'), \rho_1'(t')]]. \tag{14}$$

In obtaining this relation we used the fact that $[\hat{H}_{\mathrm{ex}}, \rho_1'(t)] = 0$ and $\mathrm{Sp}\,\hat{A}[\hat{B}\hat{C}] = \mathrm{Sp}\,\hat{B}[\hat{C}\hat{A}]$. For the case $T \gg T_C$, we can replace the exponential in Eq. (12) by $1 + \alpha(t)\hat{S}_z + \beta(t)\hat{S}_x + \gamma(t)\hat{S}_y + \delta(t)\hat{H}_{\mathrm{ex}}$. Thus, the calculation of $\partial H_{\mathrm{ex}}/\partial t$ comes down to calculating integrals of the type

$$\int_0^t dt'\,\mathrm{Sp}\,\hat{H}_{ex}[\hat{V}(t)\,[\hat{V}''(t,t'), \hat{S}_z]].$$

The calculation is accomplished in the same manner as in [10,11]. We need only stop to consider the calculation of integrals of the type

$$\int_0^t dt'\,\mathrm{Sp}\,\hat{H}_{ex}[\hat{V}(t)\,[\hat{V}''(t,t'), \hat{S}_{x,y}]].$$

A direct calculation shows that such terms are proportional to $e^{im\omega t}$, i.e., they are oscillating functions of the time. The characteristic time for a dipole relaxation process is of the order $\hbar/H_{\mathrm{dip}}$. The frequency of the alternating magnetic field $\omega \sim \mu H_0/\hbar$, and since in the usual experiment $H_0 \gg H_{\mathrm{dip}}$, we can neglect such rapidly oscillating terms.

Finally, we obtain the following system of equations in the rotating frame, which describes the variation of H_{ex} and $S_{x,y,z}$ in the dipolar-dipolar relaxation process

$$\begin{aligned}
\partial S_x/\partial t &= \gamma\,[\mathbf{SH}_{\mathrm{eff}}]_x - S_x/T_2 - S_y/T_3,\\
\partial S_y/\partial t &= \gamma\,[\mathbf{SH}_{\mathrm{eff}}]_y - S_y/T_2 + S_x/T_3,\\
\partial S_z/\partial t &= \gamma\,[\mathbf{SH}_{\mathrm{eff}}]_z - (S_z + \mathscr{H})/T_1,\\
\partial \mathscr{H}/\partial t &= -\varkappa\,(S_z + \mathscr{H})/T_1.
\end{aligned} \tag{15}$$*

Here

$$\mathscr{H} = \hbar\omega\,\mathrm{Sp}\,\hat{S}_z^2\delta(t), \quad \gamma = \mu/\hbar, \quad \varkappa = \hbar^2\omega^2\,\mathrm{Sp}\,\hat{S}_z^2/\mathrm{Sp}\,\hat{H}_{ex}^2,$$

$$T_1^{-1} = 2\pi\sum_{m=1,2} m^2\,\mathrm{Sp}'\,\hat{H}_{m\omega}^{m}\hat{H}_{-m\omega}^{-m}/\hbar^2\,\mathrm{Sp}\,\hat{S}_z^2,$$

$$T_2^{-1} = \pi\sum_{m=-2}^{m=+2}\mathrm{Sp}'\,\hat{S}^{-1}\,[\hat{H}_{m\omega}^{m}[\hat{H}_{-m\omega}^{-m}, \hat{S}^{+1}]]/\hbar^2\,\mathrm{Sp}\,\hat{S}_z^2,$$

$$T_3^{-1} = \sum_{m=-2}^{+2}\mathrm{P}\int_{-\infty}^{+\infty}\frac{d\omega'}{m\omega-\omega'}\,\mathrm{Sp}'\,\hat{S}^{-1}\,[\hat{H}_{\omega'}^{m}[\hat{H}_{-\omega'}^{-m}, \hat{S}^{+1}]]/\hbar^2\,\mathrm{Sp}\,\hat{S}_z^2.$$

(The symbol P before the integral over ω' signifies principal value.) The relation defining $\hat{H}_\omega^m$ is

$$\hat{H}_\omega^m = \frac{1}{2\pi}\int_{-\infty}^{\infty} dt\,e^{-i\omega t}\exp\left(\frac{i\hat{H}_{ex}t}{\hbar}\right)\hat{H}^m\exp\left(-\frac{i\hat{H}_{ex}t}{\hbar}\right).$$

The primes on the Sp symbols indicate that in the summation of the diagonal elements, the singular factors $\delta(0)$ contained in the diagonal matrix elements of $\hat{H}_\omega^m H_{-\omega}^{-m}$ (by virtue of the definition of $\hat{H}_\omega^m$) are omitted. [10]

Physically, it is clear that in the absence of dipole-dipole interaction the magnetization of the system in the rotating frame should precess freely around the effective field $\mathbf{H}_{\mathrm{eff}}$. In our equations, a transition to the limit $T_1 = T_2 = T_3 = \infty$ corresponds to "turning off" the dipole-dipole interaction. Then the system (15) takes the form

$$\partial \mathbf{S}/\partial t = \gamma\,[\mathbf{SH}_{\mathrm{eff}}].$$

But the solution of such a system of equations, as is well known, is a precession of the vector $\mathbf{S}$ about $\mathbf{H}_{\mathrm{eff}}$, indicating that our system of equations (15) correctly contains the limiting case in which the dipole-dipole interaction may be neglected.

However, besides this natural result, the system (15) leads to the following physically interesting fact. The equations can easily be rewritten in the form

$$\begin{aligned}
\partial S_{x,y}/\partial t &= \gamma\,[\mathbf{S}, \mathbf{H}_{\mathrm{eff}} + \Delta\mathbf{H}]_{x,y} - S_{x,y}/T_2,\\
\partial S_z/\partial t &= \gamma\,[\mathbf{S}, \mathbf{H}_{\mathrm{eff}} + \Delta\mathbf{H}]_z - (S_z + \mathscr{H})/T_1,\\
\partial \mathscr{H}/\partial t &= -\varkappa\,(S_z + \mathscr{H})/T_1.
\end{aligned} \tag{16}$$

Here $\Delta\mathbf{H}$ is some increment to the effective field, more precisely an increment to the z component of this field, since $\Delta\mathbf{H} = \Delta H_z = 1/\gamma T_3$. Thus we

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*$[\mathbf{SH}] = \mathbf{S}\times\mathbf{H}$.

see that the dipole-dipole interaction produces an apparent change in the magnitude of the constant external field or, in other words, a change in the value of the resonance frequency.

To obtain the magnetic resonance equations it is necessary to add terms connected with spin-lattice interactions to the equations (15). Without these terms, Eqs. (15), as might be expected, yield zero solutions in the steady state, which means, physically, the heating of the system to an infinite temperature on account of the absence of a dissipative process by which the energy of the external field can be transferred to the lattice.

We take the spin-lattice relaxation into account in the usual manner, [3] i.e., we assume that the variation in $S_{x,y,z}$ and $\mathcal{H}$ due to this interaction can be written in the form

$$(\partial S_z/\partial t)_{\text{s-l}} = (S_z^0 - S_z(t)) / \tau_1,$$

$$(\partial S_{x,y}/\partial t)_{\text{s-l}} = - S_{x.y}(t) / \tau_2,$$

$$(\partial \mathcal{H}/\partial t)_{\text{s-l}} = (\mathcal{H}^0 - \mathcal{H}(t)) / \tau_0.$$

Here S_z^0 and $\mathcal{H}^0$ are the initial values of $S_z(t)$ and $\mathcal{H}(t)$; τ_0, τ_1, and τ_2 are the corresponding spin-lattice relaxation times.

After the addition of the above terms to the right sides of Eqs. (15), we have

$$\begin{aligned} \partial S_{x,y}/\partial t &= \gamma\, [\mathbf{S}\mathbf{H}'_{\text{eff}}]_{x,y} - (T_2^{-1} + \tau_2^{-1})\, S_{x,y}, \\ \partial S_z/\partial t &= \gamma\, [\mathbf{S}\mathbf{H}'_{\text{eff}}]_z - (S_z + \mathcal{H}) / T_1 + (S_z^0 - S_z) / \tau_1, \\ \partial \mathcal{H}/\partial t &= -\varkappa\, (S_z + \mathcal{H}) / T_1 + (\mathcal{H}^0 - \mathcal{H}) / \tau_0. \end{aligned} \tag{17}$$

The prime on $\mathbf{H}_{\text{eff}}$ means that the z component of the effective field contains the aforementioned increment $\Delta H_{\text{eff},z} = -1/\gamma T_3$.

The system (17) can be simplified, if one considers that $\tau_{1,2} \gg T_{1,2}$, as was shown by Anderson [16] and by Skrotskiĭ and Shmatov. [6] After this simplification the stationary solution of Eqs. (17) has the form

$$S_{x,\text{st}} = \gamma H_1 (\omega - \omega_{0,\text{eff}})\, T_2^2 S_z^0/D,$$

$$S_{y,\text{st}} = -\gamma H_1 T_2 S_z^0/D,$$

$$S_{z,\text{st}} = [1 + (\omega - \omega_{0,\text{eff}})^2 T_2^2]\, S_z^0/D,$$

$$\mathcal{H}_{\text{st}} = \mathcal{H}^0\, [1 - \varkappa\gamma^2 H_1^2 \tau_0 T_2/D],$$

$$D = 1 + (\omega - \omega_{0,\text{eff}})^2\, T_2^2 + \gamma^2 H_1^2 T_1 T_2\, (1 + \varkappa\tau_0/T_1). \tag{18}$$

Here $\omega_{0,\text{eff}}$ is the effective resonance frequency, and $\omega_{0,\text{eff}} = \omega_0 + \Delta\omega_0$, where $\Delta\omega_0 = \mu\Delta H_z/\hbar = -1/T_3$.

For a comparison of these results with experimental data we turn to the work of Bloembergen and Wang, [7] in which the saturation of DPPH (diphenylpicrylhydrazyl) was measured. The measurements were made at 10^4 Mc/sec ($H_0 = 3600$ Oe) at 77 and 300° K. The Curie temperature of crystalline DPPH lies between 0.5 and 50° K, depending on the method of its preparation. The experimental data indicate that the shape of the absorption curve is Lorentzian, just as follows from our solution (18)

In [7] it was also reported that the results of the measurement were independent of temperature. It is not difficult to verify that our solution under the conditions of this experiment leads to the very same result. In fact, the quantity S_z/S_z^0 during resonance was measured in the experiment. From our solution (18) we have

$$(S_z/S_z^0)_{\text{res}} = [1 + \gamma^2 H_1^2 T_1 T_2 (1 + \varkappa\tau_0/T_1)]^{-1}. \tag{19}$$

In this expression only the factor $(1 + \kappa\tau_0/T_1)$ depends on temperature. By means of a direct calculation of the second moment T_1 is found to have a value of the order 10^{-8} sec, which also agrees with the data of Bloembergen and Wang. [7] (Calculation of the second and fourth moments shows that $T_1^{-1}(\omega)$ and $T_2^{-1}(\omega)$ can be represented by Gaussian functions with good accuracy. [17,18]) Values of τ_0 for these experimental conditions have been obtained theoretically by Griffiths, [8] who in particular showed that for $T_1 \approx 5 \times 10^{-8}$ sec the quantity $\kappa\tau_0/T_1$ is much less than unity under the conditions of Bloembergen and Wang's experiment. [7] Consequently, our expression (19) for S_z/S_z^0 should not depend on temperature in this case, as indeed was observed experimentally.

As we have already mentioned, our equations predict a shift in the resonance frequency ω_0; this was not observed in [7]. However, an estimate of $\Delta\omega_0$ shows that under the conditions of this experiment the shift in resonance frequency is very small. In fact, it is easy to show, using our expressions for $T_3(\omega)$, that $\Delta\omega_0 \approx \overline{\Delta\omega}\mu H_0/H_{\text{ex}}$, where $\overline{\Delta\omega}$ is the line width. Since $H_0 \approx 4 \times 10^3$ Oe and $H_{\text{ex}}/\mu \approx 10^5$ Oe, then $\Delta\omega_0/\overline{\Delta\omega} \ll 1$, which shows that the constant field used in the experiment was much too weak to produce any noticeable shift in resonance frequency.

As we have indicated, our results are valid only in the region $T \gg T_C$. In the region of lower temperatures $T \gtrsim T_C$ the appropriate equations for $S_{x,y,z}$ and H_{ex} have the same form as Eq. (14). However, increasing mathematical difficulties significantly complicate the solution, even though no new fundamental difficulties arise.

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Translated by L. M. Matarrese
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