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FORMATION OF SPATIALLY INHOMOGENEOUS STATES IN 2D NON-HEISENBERG MAGNETICS

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INTRODUCTION

For decades the Heisenberg model has been a foundation, on which the theory of strong magnetism was built. However, it is possible to exceed the limits of the bilinear exchange interaction without breaking the system's isotropy. Magnetics, in the Hamiltonian of which the exchange interaction of high orders in spin is comparable with the bilinear Heisenberg interaction, are among the most interesting systems of such a class [1]. It is reasonable to suppose that the more complex systems should be characterized with the new properties, which considerably differ from the properties of the Heisenberg magnetics. And indeed, the experimental investigations of the magnets with complex exchange interaction reveal magnetic structures, which are in principle impossible in the Heisenberg model [1]. The study of such systems has showed that in the magnetics with considerable exchange interaction of high orders in spin a quantum quadrupolar order can be realized, when all average projections of the spins equal to zero in the ground state of a crystal, and the cooperative ordering takes place not on the magnetic moment, but on the quadropolar one [2-7]. The examples of such systems are $TmAu_2$, UPd_3, UCu_2Sn and others [8-12]. The properties of such systems resemble much the properties of the magnetics without any ferromagnetic ordering due to the strong influence of the one-ion anisotropy crystal field. Incidentally, in certain cases, the influence of a crystal field of the one-ion anisotropy can be formally reduced to the exchange of high order in spin [1].

The quadrupolar order is not observed for all systems exhibiting exchange interaction of high orders in spin. It is supposed in [13] that exchange interaction of high orders in spin is quite common in magnetic materials and can be found by deviations of the temperature curve from Bloch's $T^{3/2}$ law. Such deviations are observed, for example, in EuTe, GdAg, EuS, UO_2 and even in iron [13-14]. Thus, the constant of biquadratic exchange interaction founded in these experiments is sometimes comparable or even larger than the constant of the bilinear exchange interaction. However, the phases with the quadrupolar order are not implemented in these systems. This is connected with the fact that the orbital moment is quenched in such systems, and their magnetism has pure spin origin.

The purpose of the present work is to investigate the phase states of the 2D ferromagnets with the biquadratic exchange interaction, so-called non-Heisenberg ferromagnets, in which the quadrupolar phases are observed. The fact is that the origin of magnetism in 2D-systems has a fundamental interest, because it differs in principle from the magnetism of 3D-systems. It is known [15,16] that there is no long-range magnetic order in an isotropic 2D-magnet at any finite temperature. However, the account of a magnetodipolar interaction leads to the stabilization of the long-range magnetic order in 2D-systems [17]. The same effect has also the influence of a magnetoelastic interaction [18-20]. However, these mechanisms of long-range

On the basis of the eigenfunctions of the one-site Hamiltonian we build the Hubbard operators, $X_n^{M'M} \equiv |\Psi_n(M')\rangle\langle\Psi_n(M)|$, in the basis of which the one-site Hamiltonian is diagonal [31,32]. These operators are related to the spin operators as follows

$$S_n^z = \cos 2\theta \left(H_n^1 - H_n^{-1}\right) - \sin 2\theta \left(X_n^{1-1} + X_n^{-11}\right),$$
$$S_n^+ = \sqrt{2}\cos\theta \left(X_n^{10} + X_n^{0-1}\right) + \sqrt{2}\sin\theta \left(X_n^{01} - X_n^{-10}\right), \quad S_n^- = \left(S_n^+\right)^+, \tag{5}$$

where $H_n^M \equiv X_n^{MM}$ are the diagonal Hubbard operators.

The components of the deformation tensor can be presented as a sum of two terms: $u_{ij} = u_{ij}^{(0)} + u_{ij}^{(1)}$. The first term in this formula corresponds to the spontaneous deformations; the second one is the dynamic part of the deformation tensor and is determined by the vibrations of the crystal lattice. After the quantization of the crystal lattice vibrations according to the standard scheme [33], we separate in the one-site Hamiltonian the terms, which contain both, the phonon operators, and the Hubbard ones. Let us denote this part of the Hamiltonian as the Hamiltonian of transformations, because it describes the processes of transformation of phonons to magnons and vice versa. The Hamiltonian of transformations can be written as

$$\mathsf{H}_{tr} = \sum_n \left\{ \sum_M P_M H_n^M + \sum_\alpha P_\alpha X_n^\alpha \right\},$$

where $P_{M(\alpha)} = \dfrac{1}{\sqrt{N}} \sum_{q,\nu} \left(b_{q,\nu} + b_{-q,\nu}^+\right) \cdot T_n^{M(\alpha)}(q,\nu)$; α are the root vectors determined by the Hubbard operators algebra [31,32]; $T_n^{M(\alpha)}(q,\nu)$ are the amplitudes of transformations; $b_{q,\nu}^+$ and $b_{q,\nu}$ are the operators of creation and annihilation of phonons.

Consider the case when the wave-vector $\vec{k}$ is oriented along the X-axis. Such assumption does not lead to the loss of generality, but simplifies computations. In such geometry, the non-zero components of the polarization vector are e_l^x and e_t^z, and the non-zero non-diagonal amplitudes of transformations look like

$$T^{1-1}(k,l) = T^{-11}(k,l) = \frac{i\lambda k}{2} T^0(k,l)\cos 2\theta,$$
$$T^{01}(k,t) = T^{10}(k,t) = \frac{i\lambda k}{2\sqrt{2}} T^0(k,t)(\cos\theta - \sin\theta),$$
$$T^{0-1}(k,t) = T^{-10}(k,t) = -\frac{i\lambda k}{2\sqrt{2}} T^0(k,t)(\cos\theta + \sin\theta),$$

where $T^0(k,\nu) = \dfrac{e^{-i\vec{k}\vec{n}}}{\sqrt{2m\omega_\nu(k)}}$.

The total Hamiltonian of the system can be written as

$$\mathsf{H} = \mathsf{H}_0 + \mathsf{H}_{tr} + \mathsf{H}_{int}^{\perp}. \tag{6}$$

where $\mathsf{H}_{int}^{\perp}$ is the transverse part of the exchange Hamiltonian, which can be expressed through the Hubbard operators as follows

$$\mathsf{H}_{int}^{\perp} = -\frac{1}{2}\sum_{\substack{n,m\\ \alpha,\beta}} B_n(\alpha) A_{nm} B_m^{+}(\beta) X_n^{\alpha} X_m^{\beta}. \tag{7}$$

Here

$B_n(\alpha) = \left[\gamma_1^{\parallel}(\alpha) \quad \gamma_1^{\perp}(\alpha) \quad \gamma_1^{\perp *}(-\alpha) \quad \gamma_2^{\parallel}(\alpha) \quad \gamma_2^{\perp}(\alpha) \quad \gamma_2^{\perp *}(-\alpha) \quad \gamma_3^{\perp}(\alpha) \quad \gamma_3^{\perp *}(-\alpha)\right]$ is the eight-dimensional vector, and $\gamma(\alpha)$ are determined by the connection (5) between the spin and the Hubbard operators.

The A_{nm} matrix in Eq. (7) can be factorized into the Cartesian sum of two matrices, $\hat{A}_{nm} = \hat{A}_{nm}^{(3)} \oplus \hat{A}_{nm}^{(5)}$, where

$$A_{nm}^{(3)} = \begin{pmatrix} J_{nm} - \frac{K_{nm}}{2} + I_{nm}^{zz} & 0 & 0 \\ 0 & \frac{I_{nm}^{xx} - I_{nm}^{yy}}{4} & \frac{2J_{nm} - K_{nm} + I_{nm}^{xx} + I_{nm}^{yy}}{4} \\ 0 & \frac{2J_{nm} - K_{nm} + I_{nm}^{xx} + I_{nm}^{yy}}{4} & \frac{I_{nm}^{xx} - I_{nm}^{yy}}{4} \end{pmatrix};$$

$$A_{nm}^{(5)} = \frac{K_{nm}}{2}\begin{pmatrix} 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1/2 & 0 & 0 \\ 0 & 1/2 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1/2 \\ 0 & 0 & 0 & 1/2 & 0 \end{pmatrix}.$$

The account of the magnetoelastic interaction results in the hybridization of the elastic and magnetic excitations, which leads to the appearance of the so-called magnetoelastic wave [34] in the magnetically ordered crystal. The spectrum of these magnetoelastic excitations is determined by the poles of the Green function, which is defined by the following expression

$$G^{\alpha\alpha'}(n,\tau;n',\tau') = -\left\langle T\tilde{X}_n^{\alpha}(\tau)\tilde{X}_{n'}^{\alpha'}(\tau')\right\rangle, \tag{8}$$

where T is Wick's operator; $\tilde{X}_n^{\alpha}(\tau) = e^{i\mathsf{H}\tau} X_n^{\alpha} e^{-i\mathsf{H}\tau}$ is the Hubbard operator in the representation of interaction, and the averaging is carried out with the total Hamiltonian of the system (6).

The dispersion equation determining the spectra of coupled magnetoelastic waves is of Dyson's type [35], and looks like

$$\det\left\|x_{ij}\right\| = 0, \tag{9}$$

where

$$x_{ij} = \delta_{ij} + G_0^{\alpha}(\omega) b(\alpha) B_i^{+}(\alpha) A_{pj} B_p(-\alpha) +$$

$$+ \Phi^{0}(k, \nu, \nu') T^{-\alpha}(k, \nu) G_o^{\alpha}(\omega) b(\alpha) T^{\beta}(-k, \nu') G_0^{\beta}(\omega) b(\beta) B_i^{+}(\alpha) A_{pj} B_p(-\beta);$$

$$\Phi^{0}(k, \nu, \nu') = \frac{D_{\nu}(k, \omega)}{1 - Q_{\nu\nu'} D_{\nu}(k, \omega)}; \qquad Q_{\nu\nu'} = T^{\alpha}(-k, \nu) G_0^{\alpha}(\omega) T^{-\alpha}(k, \nu'); \qquad b(\alpha) = \left\langle \vec{\alpha} \vec{H} \right\rangle_0;$$

$D_{\nu}(k, \omega) = \dfrac{2\omega_{\nu}(k)}{\omega^2 - \omega_{\nu}^2(k)}$ is the Green function of a free ν-polarized phonon;

$G_0^{\alpha}(\omega) = \left[\omega + \left(\vec{\alpha} \vec{E}\right)\right]^{-1}$ is the zero Green function.

It should be noted that due to the exact account of the one-site correlators, the dispersion equations are valid at any relation between the constants of exchange interactions. However, we want to investigate the spectral properties of the magnet in the vicinity of the phase transitions. Therefore, we restrict ourselves to the analysis of the dispersion equation solutions in the corresponding phases, near the points of the phase transitions.

LARGE HEISENBERG EXCHANGE INTERACTION

Consider the solutions of the dispersion equation (9) for the case of prevailing Heisenberg exchange interaction ($J_0 > K_0$).

In this case, due to the influence of the magnetodipolar and the magnetoelastic interactions, a long-range ferromagnetic (FM) order is stabilized [17,18]. Thus, the order parameters of the system have the following form

$$\left\langle S^z \right\rangle = 1, \; q_2^0 = 1, \; q_2^2 = 0.$$

In this phase the spontaneous deformations (at $T << T_C$) look like

$$u_{xx}^{(0)} = -\frac{\lambda}{2E}(1 - 2\sigma), \; u_{zz}^{(0)} = -\frac{\lambda}{2E}(2 - \sigma), \; u_{xz}^{(0)} = 0.$$

Reckoning this, the spectra of quasimagnons can be presented as

$$\varepsilon_{\parallel}(k) = 2\left(J_0 - K_0 + \frac{A_0}{3}\right) + \gamma k^2;$$

$$\varepsilon_{\perp}(k) = \sqrt{\left(A_0 + a_1 - \Omega_0 k + \alpha k^2\right)\left(a_2 + \Omega_0 k + \alpha k^2\right)}, \tag{10}$$

where $a_1 = \dfrac{\lambda^2(5 + 2\sigma)}{8E}$; $a_2 = \dfrac{\lambda^2(7 - 2\sigma)}{8E}$; $\alpha = J_0 R_0^2$; $\gamma = K_0 \tilde{R}_0^2$; R_0 and $\tilde{R}_0$ are the radii of the bilinear and the biquadratic exchanges, accordingly.

The quasiphonon spectra in the FM phase look like

magnetic order stabilization are different. In the first case, the magnetodipolar interaction changes the dependence of magnons dispersion law on the wave-vector from the square law to the root one, which results in the convergence of the fluctuation integral on the lower limit; in the second case, the long-range magnetic order is stabilized due to the appearance of a magnetoelastic gap in the quasimagnon spectrum, which also leads to the convergence of the fluctuation integral. Besides, the magnetodipolar interaction plays an essential role in the formation of the phases with spatially inhomogeneous distribution of the magnetic moment (i.e. in the formation of the magnetic domains) in 2D-systems [21-23].

For example, such domain phases were experimentally observed in ultrathin ferromagnetic films [24-26], and the models describing these phenomena were suggested in [24-29]. As was shown in [30], the magnetoelastic interaction has a crucial influence on the formation of such phases, which is exhibited in broadening of the interval of domain structure existence and in a set of other interesting effects. Besides, the magnetic phase transitions in the materials with the quadrupolar order are accompanied by a crystal lattice distortion as, for example, in TbP and DySb [8 and Ref. herein]. That is why, it is essential to take into account the magnetoelastic coupling at consideration of the phase transitions in non-Heisenberg magnetics.

However, to our knowledge, nobody has either taken into account both, the magnetodipolar interaction and the magnetoelastic coupling in 2D non-Heisenberg magnets, nor has investigated the possibility of the formation of the phases with inhomogeneous distribution of the quadrupolar order parameters in such magnetics.

MODEL

Consider a 2D non-Heisenberg ferromagnetic taking into account the magnetodipolar and the magnetoelastic interactions. The Hamiltonian of such magnetic reads

$$\mathsf{H} = -\frac{1}{2}\sum_{n,m}\left[J(n-m)\delta_{ij} + V_{ij}(n-m)\right]S_n^i S_m^j - \frac{1}{2}\sum_{n,m} K(n-m)\left(\vec{S}_n\vec{S}_m\right)^2 + \\ + \lambda\sum_n u_{ij}(n)S_n^i S_m^j + \frac{E}{2\left(1-\sigma^2\right)}\int\left[u_{xx}^2 + u_{zz}^2 + 2\sigma u_{xx}u_{zz} + (1-\sigma)u_{xz}^2\right]dr, \tag{1}$$

where $J, K > 0$ are the constants of the Heisenberg and the biquadratic interactions, accordingly; S_n^i is the i-th component of the spin operator at the lattice site n; V_{ij} are the components of the tensor of magnetodipolar interaction; u_{ij} are the components of the deformation tensor; λ is the constant of the magnetoelastic coupling; E is Young's modulus; σ is Poisson's ratio. The film plane is XOZ. We suppose that the effective spin of a magnetic ion is unity. The further consideration will be carried out for the low-temperature case ($T << T_C$, T_C is the Curie temperature).

If XOZ is the film plane, the components of the tensor of magnetodipolar interaction for a 2D system have the form [17]:

$$V_k^{xx} = \frac{A_0}{3} - \Omega_0 k \cos^2\varphi; \quad V_k^{yy} = -\frac{2}{3}A_0 + \Omega_0 k;$$
$$V_k^{zz} = \frac{A_0}{3} - \Omega_0 k \sin^2\varphi; \quad V_k^{xz} = -\frac{\Omega_0 k}{2}\sin 2\varphi; \quad V_k^{xy} = V_k^{yz} = 0; \qquad (2)$$
$$A_0 = \frac{3}{2}(g\mu_B)^2 \sum_{R=0} R^{-3}; \quad \Omega_0 = \frac{2\pi(g\mu_B)^2}{a^2},$$

where a^2 is the "volume" of a flat elementary cell; g is the Lande factor; μ_B is the Bohr magneton; φ is the angle between the direction of a wave-vector $\vec{k}$ and the X-axis.

Separating in the exchange part of the Hamiltonian (1) the mean field proportional to $\langle S_n^z \rangle$ and connected with the ordering of the magnetic moments, and the auxiliary fields proportional to $\langle O_{2n}^p \rangle \equiv q_2^p$ (p = 0, 2), which define the average quadrupolar moments, we obtain the equation for the one-site Hamiltonian $\mathsf{H}_0(n)$:

$$\mathsf{H}_0(n) = -\bar{H}S_n^z - B_2^0 O_{2n}^0 - B_2^2 O_{2n}^2 + \lambda u_{ij} S_n^i S_n^j$$

where $\bar{H} = \left(J_0 - \frac{K_0}{2} + \frac{A_0}{3}\right)\langle S^z \rangle$; $B_2^0 = \frac{K_0}{6}q_2^0$; $B_2^2 = \frac{K_0}{2}q_2^2$; $O_{2n}^0 = 3(S_n^z)^2 - S(S+1)$; $O_{2n}^2 = \frac{1}{2}\left[(S_n^+)^2 + (S_n^-)^2\right]$. Solving the one-site problem with the Hamiltonian $\mathsf{H}_0(n)$, we obtain the energy levels of a magnetic ion

$$E_{\pm 1} = -B_2^0 + \frac{\lambda}{2}\left(u_{xx}^{(0)} + 2u_{zz}^{(0)}\right) \mp \kappa; \quad E_0 = 2B_2^0 + \lambda u_{xx}^{(0)};$$
$$\kappa = \sqrt{\bar{H}^2 + \left(B_2^2 - \frac{\lambda}{2}u_{xx}^{(0)}\right)^2}, \qquad (3)$$

and the eigenfunctions of the one-site Hamiltonian, which are represented as a linear combination of the eigenvectors $|M\rangle$ ($M = -1, 0, 1$) of the S^z operator:

$$\Psi_n(1) = \cos\theta|1\rangle + \sin\theta|-1\rangle, \quad \Psi_n(0) = |0\rangle, \quad \Psi_n(-1) = -\sin\theta|1\rangle + \cos\theta|-1\rangle,$$

where $\cos\theta = \sqrt{\frac{\kappa + \bar{H}}{2\kappa}}$; $u_{ij}^{(0)}$ are the spontaneous deformations, which explicit solutions is determined by the condition of the free energy density minimum:

$$F = F_{el} - T\ln Z, \qquad (4)$$

where $Z = \sum_M \exp\left(-\frac{E_M}{T}\right)$ is the partition function.

$$\omega_1^2(k)=\omega_t^2(k)\frac{2\left(J_0-K_0+\frac{A_0}{3}-2b_1\right)+\gamma k^2}{2\left(J_0-K_0+\frac{A_0}{3}\right)+\gamma k^2};\ \omega_2^2(k)=\omega_t^2(k)\frac{b_2+\Omega_0 k+\alpha k^2}{a_2+\Omega_0 k+\alpha k^2}, \quad (11)$$

where $b_1=\frac{\lambda^2}{8E}$; $b_2=\frac{3\lambda^2(1-2\sigma)}{8E}$.

It follows from Eqs. (10)-(11) that the longitudinally polarized sound mode is a soft quasiphonon mode. The phase transition from the FM ordered state to the quadrupolar phase takes place at decreasing of the constant of the bilinear exchange interaction. The critical relation between the material constants corresponding to this phase transition is obtained from the condition of softening of the spectrum of longitudinally polarized quasiphonons, and yields

$$J_0^{FM}=K_0-\frac{A_0}{3}+\frac{\lambda^2}{4E}. \quad (12)$$

Eq. (12) determines the stable point of the corresponding phases, and this phase transition is of the first-order.

LARGE BIQUADRATIC EXCHANGE INTERACTION

Now consider the opposite case when the constant of the biquadratic exchange interaction is much larger than the constant of the bilinear exchange interaction ($K_0 > J_0$). The long-range magnetic order is also stabilized in this case, but it is not of the ferromagnetic type, but of the quadrupolar one [1,2], i.e. the homogeneous quadrupolar (QU) phase is realized. The order parameters in this phase look like

$$\langle S^z\rangle=0,\ q_2^0=q_2^2=1.$$

In this phase the non-diagonal spontaneous deformations vanish, and the diagonal spontaneous deformations are equal and read as

$$u_{xx}^{(0)}=u_{zz}^{(0)}=-\frac{\lambda}{E}(1-\sigma),\ u_{xz}^{(0)}=0.$$

The equality of the spontaneous deformations $\left(u_{xx}^{(0)}=u_{zz}^{(0)}\right)$ results in the degeneracy of the excited energy levels E_0 and E_{-1} in the QU phase.

The quasiphonon spectra in the QU phase are linear in the wave-vector: $\omega_1(k)=c_t k$, $\omega_2(k)=c_t k$, and the spectra of quasimagnons are statically renormalized by the magnetoelastic coupling:

$$\varepsilon_{\parallel}(k)=\sqrt{\left(c_0+\gamma k^2\right)\left[2\left(K_0-J_0-\frac{A_0}{3}\right)+c_0\right]};$$
$$\varepsilon_{\perp}(k)=\sqrt{\left(c_0+\gamma k^2\right)\left[2\left(K_0-J_0-\frac{A_0}{3}\right)+c_0+2\Omega_0 k-\gamma k^2+2\alpha k^2\right]}. \quad (13)$$

where $c_0=\frac{\lambda^2(1-\sigma)}{E}$. The quasimagnon spectra (13) coincide in the absence of the magnetodipolar interaction ($A_0 = \Omega_0 = 0$), $\varepsilon_{\parallel}(k)=\varepsilon_{\perp}(k)$, which corresponds to the results of Ref. [36].

The critical values of the material constants, at which the phase transition from the QU phase to the FM phase takes place, are defined from the condition of vanishing of the gap in the spectrum of "low-frequency" quasimagnons $\varepsilon_{\perp}(k)$. However, due to the presence of the terms linear in the wave-vector k in $\varepsilon_{\perp}(k)$, the minimal value of the wave-vector corresponding to vanishing of the gap is not zero anymore, but $k^*=\frac{\Omega_0}{\gamma-2\alpha}$. This leads to the renormalization of the gap in $\varepsilon_{\perp}(k)$, and consequently to the change in the critical value of the constant of the bilinear exchange interaction:

$$J_0^{QU}=K_0-\frac{A_0}{3}+\frac{\lambda^2(1-\sigma)}{2E}+\frac{\Omega_0^2}{\gamma-2\alpha}. \quad (14)$$

The last term in Eq. (14) is associated with the finitude of the wave-vector.

Eq. (14) determines the stable point at the phase transition from the spatially inhomogeneous state to the FM phase. The inhomogeneity in this spatially modulated phase is not connected with the spatial distribution of the magnetization $\left(\langle S^z\rangle=0\right)$, but rather with the change of the quadrupolar order parameters, which are related with the orientation of the main axes of the tensor of quadrupolar moments. The period of this spatial inhomogeneity is determined by the reciprocal value of the critical wave-vector k^*. Its estimations for the characteristic values of the material constants ($K_0 \sim 1000$ kOe, $J_0 \sim 250$ kOe, $\Omega_0 \sim 14$ kOe [28,37]) yields $\frac{1}{k^*}\sim 3\cdot 10^{-7}$ cm.

As follows from Eqs. (13) and (15), $J_0^{QU}>J_0^{FM}$, i.e. the phase transition considered is the first-order phase transition with hysteresis. Thus, the region of the inhomogeneous phase existence is

$$\Delta J=\frac{\lambda^2}{4E}(1-2\sigma)+\frac{\Omega_0^2}{\gamma-2\alpha}.$$

The quantitative estimations of ΔJ give about 100 Oe, so this interval can be found experimentally.

CONCLUSION

Due to the account of the magnetodipolar interaction, in a 2D non-Heisenberg magnetic not only spatially homogeneous phases can be realized (ferromagnetic and quadrupolar), but also the spatially inhomogeneous state. Thus, in the last state the inhomogeneity is connected with the distribution of the tensor of quadrupolar moments, or more accurately – with the orientation of its main axes.

Apparently, the absence of the inhomogeneous FM phase (magnetic domains) in the system considered is connected with the fact that the magnetodipolar interaction creates effective easy-plane anisotropy in the film plane, and the bilinear exchange interaction, which is an isotropic one, cannot compete with this anisotropy and ranges the elementary magnetic moments in the film plane. The biquadratic exchange interaction can be considered as a quasiantiferromagnetic one, which results in origin of an effective anisotropy in the plane perpendicular to the film plane. This leads to the competence between the biquadratic exchange interaction and the magnitodipolar one, which is exhibited in the possibility of inhomogeneous phase formation.

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