Simple model calculation of volume magnetostriction in rare-earth-Mn$_2$ cubic Laves phases at $T=0$

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Volume magnetostriction changes and transition pressures are calculated for the first-order phase transition for the cubic Laves phase rare-earth-Mn$_2$ intermetallic compounds at $T=0$ within the Blume-Capel model. The origin of the magnetovolume effect is the volume dependence of bandwidth and the instability of the Mn magnetic moments. Agreement with experiment is obtained for the magnetovolume strains, whereas the calculated transition pressures at $T=0$ are larger than the experimental values. The stability of Mn magnetic moments is found to depend on both the magnetic and elastic degrees of freedom. [S0163-1829(97)06538-7]

I. INTRODUCTION

The stability of magnetic moments of transition-metal atoms has been of interest in the theory of magnetism for more than 30 years. Mathematically, the question about the stability of the Hartree-Fock (HF) solution for the system of interacting fermions was raised in Refs. 1 and 2. Soon, an effective localized Hamiltonian was formulated[3] on the basis of the free-electron theory. This discussion was summarized by Friedel in 1969.[4] On the other hand, continuous efforts were undertaken to improve the HF solution itself. To achieve this aim, the functional integral formalism[5] and the Stratonovitch trick[6] were applied[7,8] to the well-known Hubbard model.[9] However, it was found difficult to generalize the approach and in most cases, the so-called static approximation was applied. Within this approximation, charge fluctuations were neglected and so was the noncommutativity of quantum operators in the formula for the partition function. At $T=0$, the HF solution was recovered. The current state of the art is reviewed in Ref. 10.

In a recent series of papers, the effective localized Hamiltonian was used[11–13] to discuss frustration effects in RMn$_2$ [R=rare earth (RE)] intermetallics. There, the expression for the interatomic exchange was the same as the one obtained earlier[14] with somewhat different assumptions; both expressions were derived from the nondegenerated Hubbard model within the static approximation. This expression is similar to the well-known Ruderman-Kittel-Kasuya-Yosida (RKKY) formula,[15] but the physical mechanism responsible for intersite coupling is the d-d correlation rather than the s-d exchange interaction. As was pointed out,[11–13] the stability of the magnetic moment of Mn atoms is governed mainly by a one-ion coupling constant. The effective Hamiltonian was shown to be the Blume-Capel Hamiltonian[16,17]

$$H = \Delta \sum_i s_i^2 + J \sum_{\langle ij \rangle} s_i s_j, \qquad (1)$$

where $s_i = 0$ or ± 1, and the second sum was over pairs of nearest neighbors (NN). The interaction constants: one ion (Δ) and two ions (J) were derived within a second-order perturbation calculation. Their expressions are

$$\Delta = \mu^2 \frac{U}{4} \left[1 + \frac{U}{N} \sum_{\mathbf{k,q}} \frac{f(\epsilon_{\mathbf{k}}) - f(\epsilon_{\mathbf{k+q}})}{\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k+q}}} \right], \qquad (2)$$

$$J_{ij} = \mu^2 \frac{U^2}{4N} \sum_{\mathbf{k,q}} e^{i\mathbf{k} \cdot \mathbf{R}_{ij}} \frac{f(\epsilon_{\mathbf{k}}) - f(\epsilon_{\mathbf{k+q}})}{\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k+q}}}, \qquad (3)$$

where μ is the magnetic moment of an atom, U is the Coulomb intra-atomic interaction, N is the number of atoms, $\mathbf{k}$ and $\mathbf{q}$ are wave vectors, $\epsilon_{\mathbf{k}}$ are one-electron energies, and $f_{\mathbf{k}}$ are average occupation numbers. On the basis of the Hamiltonian (1), mixed magnetic phases of a two-dimensional triangular lattice were investigated,[11–13] where some atoms possessed magnetic moments and some others did not. In this way, frustration was absent in the ground state, despite the antiferromagnetic Mn-Mn interaction.

This interesting idea raises some questions when applied to RMn$_2$ systems. The first question is about the actual role of frustration in a three-dimensional lattice. For the heavy R and Y, RMn$_2$ systems are known to crystallize in the C15 cubic Laves phase structure,[18] where all Mn atoms are equivalent. They form tetrahedra; each atom connects two of such tetrahedra and has therefore six NN. This is the same as in a triangular lattice: however, despite frustrations, magnetic energy of such a unit of seven atoms is the lowest when all of them are magnetic. For a tetrahedron, magnetic energy of configurations with 1, 2, 3, and 4 magnetic atoms, respectively, are 0, $-J$, $-J$, and $-2J$. To find a minimal density of frustration in the whole lattice is not a trivial task; however, it is obvious that the role of frustration in the Laves phases is not as substantial as in the triangular lattice.

Another question is about the role of a change of volume, which accompanies magnetism of Mn atoms. This effect was not included in the above model; however, a volume striction is known to be essential for the appearance of magnetic moments at Mn atoms. This dependence is known as the idea of a ''critical distance'' (d_c) between Mn atoms; below this distance, the Mn moment is unstable.[18] The value of d_c was evaluated to be 2.66 Å at room temperature.[19] The idea of a critical distance was criticized as an oversimplification;[20] in particular, it was pointed out that band filling influences its value. Nevertheless, the magnetoelastic energy associated to

the variation of the Mn magnetic moments cannot be ignored. Note that the largest ever observed forced volume magnetostriction ($\cong 1.7\%$) was found in TbMn$_2$.[21] Now, if we adopt the idea of a mixed phase, where magnetic and nonmagnetic atoms form a translationally invariant complex structure, we are forced to admit that a local volume strain follows to this structure in the same complex way, just because it is forced to do so by the magnetic interaction. As the elastic energy is usually larger than the magnetoelastic one, this picture seems to be controversial.

The aim of this paper is to include the volume magnetostriction energy into the Hamiltonian (1), and to discuss the three-dimensional Laves phase $C15$ structure within the Blume-Capel model. We assume that the magnetoelastic coupling is driven by the Heine's mechanism of the volume dependence of the bandwidth W,

$$\frac{\partial \ln W}{\partial \omega} = -\lambda, \qquad (4)$$

where $\omega = \Delta V / V$ is a homogeneous volume strain.[22,23] As the RKKY interaction (with those akin to it) is expected to depend on $\mathbf{k}_F a$, where a is the lattice constant and $\mathbf{k}_F$ is the Fermi wave vector, and as this latter product does not depend on volume, the Heine's coupling seems to be the only important one. This last assumption is by no means new; for references see Ref. 10. The constants Δ and J depend on volume because the energies $\epsilon_{\mathbf{k}}$ are proportional to the bandwidth W [see Eqs. (2) and (3)].

The tentative goal of this paper is to connect the above model with what is known about the magnetic phases of RMn$_2$ compounds from experimental data. Here, however, in many cases the situation is not clear. Although some local magnetic anisotropy is present for the Mn atoms, the orientation of their moments is noncollinear[24] and it is only an approximate picture that can be obtained within an Ising-like model. R^{3+} ions are magnetic (except indeed Y^{3+}) and the exchange interaction with the R^{3+} modifies the magnetism of Mn. Summarizing the experimental data collected[19–21,24–34] for the purposes of the discussion of the magnetovolume effect, three magnetic phases can be distinguished for the RMn$_2$ systems: (i) a phase where the Mn atoms have no magnetic moments (NMP); (ii) a phase where only 1/4 of the Mn atoms are magnetic (so-called nontransformed phase, NTP); (iii) a phase where all Mn atoms are magnetic (transformed phase, TP).

The above picture is based on the experimental magnetic phase diagrams.[32,34] NTP is possible, because some Mn atoms interact more strongly with the R^{3+} sublattice than the others. The problem is that the magnetic structure of NTP is not fully understood; the neutron-diffraction data do not give clear information. The relative orientations of Mn and R^{3+} magnetic moments are supposed to vary with the particular R compound, external magnetic field, pressure and temperature. Still, as it is argued below, a simple model which treats all $C15$ magnetic structures within the same scheme can provide some understanding of the low-temperature experimental data.

The plan of the paper is the following: in Sec. II, the model is described; in Sec. III the model results are compared with the experimental data on magnetovolume changes

at the phase magnetic transitions and with the transition pressures; in Sec. IV, the validity of these results are discussed and the main conclusions extracted.

II. THE MODEL

Our starting point is the Hamiltonian (1), with the constants Δ and J which depend on volume strain, ω, as follows:

$$\Delta(\omega) = \mu^2 \frac{U}{4} + \frac{W(0)}{W(\omega)} \left[\Delta(0) - \mu^2 \frac{U}{4} \right], \qquad (5)$$

$$J(\omega) = \frac{W(0)}{W(\omega)} J(0). \qquad (6)$$

These expressions can be easily derived from Eqs. (2), (3), if we substitute $\epsilon_{\mathbf{k}}(\omega)/\epsilon_{\mathbf{k}}(0) = W(\omega)/W(0)$, and we neglect the volume dependence of the Fermi level. Explicitly, for $\Delta(\omega)$ we get

$$\Delta(\omega) = \mu^2 \frac{U}{4} \left[1 + \frac{U}{N} \frac{W(0)}{W(\omega)} \sum_{\mathbf{k},\mathbf{q}} \frac{f(\epsilon_{\mathbf{k}}) - f(\epsilon_{\mathbf{k+q}})}{\epsilon_{\mathbf{k}}(0) - \epsilon_{\mathbf{k+q}}(0)} \right], \qquad (5a)$$

and when the sum over wave vectors $\mathbf{k},\mathbf{q}$ is separated out from Eq. (5a), we obtain Eq. (5). A similar procedure gives Eq. (6). From now on, we assume $\mu^2 = 1$. Substituting Eq. (4) into Eqs. (5), (6), we obtain

$$\left(\frac{\partial \Delta(\omega)}{\partial \omega} \right)_{\omega=0} = \lambda \left[\Delta(0) - \frac{U}{4} \right] \qquad (7)$$

and

$$\left(\frac{\partial J(\omega)}{\partial \omega} \right)_{\omega=0} = \lambda J(0). \qquad (8)$$

For simplicity, we will denote $\Delta(0) \equiv D$, $J(0) \equiv K$ from here on. Equations (7), (8) are the linear approximations of the volume dependence of $\Delta(\omega)$ and $J(\omega)$, defined by Eqs. (4)–(6). The total Hamiltonian is

$$H(\omega) = \left\{ D + \lambda \omega \left[D - \frac{U}{4} \right] \right\} \sum_i s_i^2 + K\{1 + \lambda \omega\} \sum_{\langle ij \rangle} s_i s_j$$

$$- \sum_j h_{\mathrm{eff}}(i) s_i. \qquad (9)$$

The third term is the Zeeman effective interaction, which comprises an interaction with the R^{3+} sublattice. The Gibbs free-energy density becomes

$$G = \frac{c}{2} \omega^2 + \langle H(\omega) \rangle + p\omega, \qquad (10)$$

where p is the external applied pressure, c is the relevant elastic constant, and $\langle \cdots \rangle$ is the thermodynamical average. In fact, it is the average over the ground state of magnetic variables s_i. Minimizing G with respect to the volume strain ω, we get the equilibrium values of the volume strains ω for the three phases: NMP, NTP, and TP. Substituting such ω equilibrium values in Eq. (10) gives the values of G for the three phases. Comparing these G values, we find the ground-state

(GS) phase for a given pressure, and the transition pressure p_c where the equilibrium transition takes place at $T=0$. The GS depends also on the magnetic interaction of the Mn sublattice with the R^{3+} sublattice. Finally we calculate the values $\Delta\omega$ of the change in volume strain at $p=p_c$ and $T=0$, the main aim of the present work. As we will comment below these $\Delta\omega$ values are close to the ω "jumps" at the magnetic phase transition temperatures at atmospheric pressure.

For the NMP, the averages $\langle s_i \rangle$, $\langle s_i^2 \rangle$ and the spin-correlation function $\langle s_i s_j \rangle$ are equal to zero. For the TP we have $\langle s_i \rangle = 0$, $\langle s_i^2 \rangle = 1$ and $\langle s_i s_j \rangle = -1/3$; a half of the spins is "up" and a half is "down." For the NTP $\langle s_i \rangle = -1/4$, $\langle s_i^2 \rangle = 1/4$, and $\langle s_i s_j \rangle = 0$; the latter is true because in NTP there are no Mn-Mn nearest-neighbor pairs. Some net magnetic moment of the Mn sublattice is present in this phase, and it is antiferromagnetically coupled to the R^{3+} sublattice. The mutual R^{3+} magnetic moments interaction is ferromagnetic. The R^{3+} magnetic moment is larger than this one of Mn; that is why the magnetic moment of the Mn sublattice is antiparallel to an external magnetic field. Note that in the NTP, the Mn atoms are not equivalent.

For the transition between NMP and TP we get the transition pressure

$$p_c = \frac{c(K-D)}{\lambda(U/4+K-D)} + \frac{\lambda}{2}(U/4+K-D) \qquad (11)$$

and the change in volume strain between both phases,

$$\Delta\omega = \frac{\lambda}{c}(U/4+K-D). \qquad (12)$$

For the transition between NMP and NTP we get

$$p_c = \frac{\lambda}{8}(U/4-D) + \frac{c(\Gamma-D)}{\lambda(U/4-D)} \qquad (13)$$

and similarly

$$\Delta\omega = \frac{\lambda}{4c}(U/4-D), \qquad (14)$$

where Γ is the effective magnetic field created at the Mn sites by the R^{3+} sublattice. Finally, for the transition between NTP and TP,

$$p_c = \frac{1}{3U/4+4K-3D} \left\{ \frac{c}{\lambda}(4K-\Gamma-3D) \right.$$
$$\left. + 2\lambda[(U/4+K-D)^2 - (U/4-D)^2/16] \right\} \qquad (15)$$

and

$$\Delta\omega = \frac{\lambda}{4c}(3U/4+4K-3D). \qquad (16)$$

III. COMPARISON WITH EXPERIMENT

We remind the reader that we have to distinguish between ω, which is the change of volume with respect to the situation of the nonmagnetized sample without applied pressure, and $\Delta\omega$, which is the change of volume strain between two

TABLE I. Model calculation parameters for various RMn$_2$ compounds, in eV/atom. Third column values (elastic constants) are taken from Ref. 19. The parameters D and K are defined in the text.

Rare earth	D	K	c
Dy	0.0	0.0087	
Tb	0.0025	0.0094	3.1
Y	0.0056	0.01	2.0
Gd	0.0056	0.01	3.6

given magnetic phases. Experimentally we will take for $\Delta\omega$ the volume distortion "jump" at the magnetic phase transition temperatures; this value should be practically equal to the calculated one at $p=p_c$ and $T=0$, inasmuch as it is the volume strain variation between the two magnetic phases. Moreover, experimentally the thermal variations of ω between 0 K and the magnetic phase transition temperatures are negligible compared with $\Delta\omega$. Therefore it is $\Delta\omega$ at $T=0$ K and $p=p_c$ which is compared with the experimental data, as we will show later on.

Some parameters of the calculations can be fixed easily. The compressibility $\kappa=1/c$ for GdMn$_2$ at room temperature was determined[30] to be 35×10^{-3} GPa^{-1}; for YMn$_2$ and TbMn$_2$ we adopt the values of 62×10^{-3} and 40×10^{-3} GPa^{-1}, respectively.[19] For 24 Mn and R^{3+} atoms per a cubic unit cell with a lattice constant of 7.75 Å,[35] we obtain for the elastic constant $c=3.571$, 2.016, and 3.125 eV/atom for GdMn$_2$, YMn$_2$, and TbMn$_2$, respectively. The Néel temperature does not depend strongly on the R partner[26] and is about 100 K, except for the case of TbMn$_2$, where it is about 40 K. This exception could be due to some reorientations of the $3d$ Mn or Tb^{3+} moments near T_N; here we are interested in the energy scale and not in thermal properties, and we will neglect it. The dependence of J and Δ on the particular RMn$_2$ compound are to be evaluated from their linearized volume strain dependence, i.e., Eqs. (7), (8). The initial condition for Eq. (7) is chosen to be $\Delta(0)=0$ for DyMn$_2$, because the spontaneous jump of the Mn moment is observed near this compound.[18] To evaluate $\Delta(0)\equiv D$ for the other compounds, the volume dependence of Δ is used as in Eq. (7), from where we immediately get $\Delta_R(0)=\Delta_{\text{Dy}}(0)+\lambda(D-U/4)\omega_R$, where $\omega_R=(V_R-V_{\text{Dy}})/V_{\text{Dy}}$, V_R being the lattice cell volume for the RMn$_2$ compound with $R=$Y, Gd, and Tb. The range of the change of ω_R is from DyMn$_2$ ($a=7.53$ Å) to GdMn$_2$ ($a=7.73$ Å),[18] i.e., about 0.08. The values of J for the particular compounds are obtained in a similar way from Eq. (8). For Eq. (8) we choose $J=0.01$ eV, i.e., the value for YMn$_2$, because Y is not magnetic and does not change the picture; however, as J is far from zero and its derivative is small, the $J(\omega)$ dependence is of minor importance. The values of the obtained $\Delta(0)\equiv D$ and $J(0)\equiv K$ parameters are collected in Table I. For the constant λ, the value 5/3 is adopted,[22] although band calculations[23] suggest some variations for this parameter. We are left with only one free parameter, i.e., U. As its very origin is an intra-atomic Coulomb interaction, we do not expect it to vary strongly with volume; we note, however, that the difference between intra-atomic and interatomic interaction is not well-defined in the limit of free electrons. Let us also mention that, as it was suggested by Friedel,[4] various

correlations reduce the effective value of U to be less than 1 eV/atom. We have found that the best accordance with the experimental results on the changes of volume strain between the different magnetic phases for the $R\mathrm{Mn}_2$ intermetallics can be obtained with the value $U = 0.17$ eV/atom.

A. TP-NMP transition: YMn$_2$

The case of TP-NMP transition in the absence of RE magnetic moments [Eq. (12)] can be assigned to YMn$_2$. The experimental data[18] give the value $\Delta\omega = 0.05$; from Eq. (12), we get $\Delta\omega = 0.039$. Note that the value of $D \equiv \Delta(0)$ for YMn$_2$ was evaluated from Eq. (7). Forced volume magnetostriction for this compound is close to zero. This is in accordance to our assumption that at TP, an external magnetic field acts mainly through the R sublattice. Note that for a small amount of Ni (substitution of 0.01 at. % of Mn), $\Delta\omega$ is reduced twice.[25] This fact confirms the correctness of the model; the phase diagram was found to be sensitive to band filling.[36] The transition pressure p_c calculated from Eq. (11) is 12 Kbars. This is about three times larger than the experimental value of about 4 Kbars, needed to reduce the Néel temperature for YMn$_2$ down to zero.[19]

B. NTP-NMP transition: Dy$_{1-x}$Y$_x$Mn$_2$ series

Another case is the change of volume from NTP to NMP, which can be observed for Dy$_{0.7}$Y$_{0.3}$Mn$_2$. The value of $D \equiv \Delta(0)$ is adjusted with the above assumed linear dependence on the R to be 0.01 U. Unfortunately, we have no data on compressibility; adopting the value for YMn$_2$, we get $\Delta\omega = 8.4 \times 10^{-3}$ from Eq. (14), whereas experimentally $\Delta\omega = 7 \times 10^{-3}$.[29] For DyMn$_2$ the experiment gives $\Delta\omega = 5 \times 10^{-4}$,[29] and this effect is assigned to the Dy sublattice. As Mn carries no magnetic moment, model calculation gives $\Delta\omega = 0$ for this particular case. For $x = 0.8$, the experimental value of $\Delta\omega$ in a field of 12 T shows large thermal hysteresis for $\Delta\omega$, between 10^{-2} and 2×10^{-2}. In this latter case, the NTP-NMP transition seems to be mixed with TP-NMP or TP-NTP transitions. Therefore it would be a speculation to apply any concrete expression for $\Delta\omega$. Also we have no experimental data on the transition pressure.

C. TP-NTP transition: TbMn$_2$ and GdMn$_2$

The transition from TP to NTP in the presence of RE magnetism can be studied in TbMn$_2$, where experiment gives $\Delta\omega = 1.7 \times 10^{-2}$.[21] From Eq. (16) we get $\Delta\omega = 2.1 \times 10^{-2}$. The experimental value[32] is approximately constant when Tb is substituted by Y up to the concentration Tb$_{0.5}$Y$_{0.5}$Mn$_2$, where we obtain $\Delta\omega = 2.5 \times 10^{-2}$ from Eq. (16).

The same transition from TP to NTP can be expected in GdMn$_2$, where the experimental value for $\Delta\omega$ was found to be about 0.6×10^{-2} at T_N (Ref. 30) or $0.6 - 1.2 \times 10^{-2}$.[18] Our calculation for this case gives 1.7×10^{-2}. The D and J parameters for GdMn$_2$ are the same as for YMn$_2$, because ω_R for the two compounds are very close,[18] but in the latter case the transition is from TP to NMP.

Both for TbMn$_2$ and GdMn$_2$, the critical pressures are to be obtained from Eq. (15). There, in principle, we need information on the effective magnetic field Γ. However, generally, the magnetic coupling to the RE sublattice is small, and the range of order of magnitude for p_c should not depend on Γ. Setting $\Gamma = 0$, we get p_c equal to 30 kbar for GdMn$_2$ and to 33 kbar for TbMn$_2$, while the experimental values are 11.3 and 2.0 kbar, respectively.[19]

The overall model results on $\Delta\omega$ for the $R\mathrm{Mn}_2$ intermetallics are summarized in Fig. 1, where we include the experimental values for comparison. The agreement is rather good. Results are given for the cases where we dispose of the elastic stiffness constants c; an exception is DyMn$_2$, but our result $\Delta\omega = 0$ for this compound does not depend on the elastic constant value. For intermediate concentrations (e.g., Dy$_x$Y$_{1-x}$Mn$_2$) the results are expected to depend on x roughly linearly, and the general accordance with experiment is preserved.
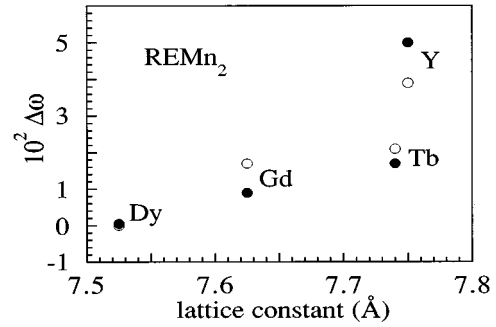

FIG. 1. Experimental (full circles) and theoretical (empty circles) values for the volume magnetostriction change $\Delta\omega$ between two magnetic phases, for $R\mathrm{Mn}_2$ cubic Laves phase intermetallics (see details about the meaning of $\Delta\omega$ in text).

IV. DISCUSSION AND CONCLUSIONS

As it has been shown, good accordance is obtained with the experimental magnetovolume data, but the formulas for transition pressure are not fully reliable. Our results on the strains reproduce well an experimental rule, that the volume magnetostriction increases when the Mn magnetic moments become more and more stable. We suppose that the small but existing discrepancy between the calculated values and the experimental data on volume strains has its source in the fact that the phase transition does not encompass the whole sample, in contrast to the theoretical calculations. The problem of the transition pressure is a difficult many-body problem, which includes the magnetic contribution to compressibility and mutual interaction of band electrons as dependent on pressure. The latter effect cannot be described within an Ising phenomenology. What is still remarkable is that for larger volumes per atom, i.e., for $C14$ hexagonal Laves phase PrMn$_2$ or NdMn$_2$ compounds, the transition pressures are of the order of 20–40 kbars.[19] This could mean that the ranges of magnitude of our model parameters are proper.

A weak point of the model is that the angular degrees of freedom are not taken into account. As was pointed out in Sec. I, the discussion on the validity of a localized picture is one of the classical matters of the theory of magnetism; it is not our aim to give new arguments here. However, the imposed restriction from the Heisenberg to the Ising model does not allow us to discuss possible variation of the direction of R^{3+} and Mn moments with pressure. The Ising ap-

proximation is a deficiency of both this work and the approach of Refs. (11)–(13), which we use as the starting point.

In conclusion, we have calculated the magnetovolume strains and transition pressures for the cubic $C15$ RMn$_2$ intermetallic compounds at $T=0$ within a very simple Ising-like model with one adjustable parameter, i.e., the intra-atomic Coulomb interaction between d electrons. Reasonable accordance with experiment is obtained for volume magnetostriction in YMn$_2$, DyMn$_2$, GdMn$_2$, and TbMn$_2$, and for some of their alloys. Volume magnetostriction is found to be essential for the stability of Mn magnetic moments.

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