



THE INFLUENCE OF THE PHASE TRANSFORMATION: $\text{Fhyd} \rightarrow \text{Fe}_3\text{O}_4$, OCCURS DURING THE ANNEALING TREATMENT OF SOL-GEL SYNTHESIS, ON THE ABSORPTION PROPERTIES OF THE FINAL SYNTHESIS PRODUCT ($\text{Fe}_3\text{O}_4/\text{C}$ HOLLOW-SPHERE NANOPARTICLES)

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Abstract: In this paper, we perform a theoretical study of the possible improvement of the absorption properties ($> 8 \text{ GHz}$) of the sample consisting of $\text{Fe}_3\text{O}_4/\text{C}$ hollow sphere nanoparticles, prepared by sol-gel method. In order to achieve proposed goal, we examine the impact of the phase transformation of the sample, occurred during the annealing treatment. It was found that the presence of the $\text{Fhyd} \rightarrow \text{Fe}_3\text{O}_4$ phase transformation, occurred during the thermal treatment process, decreases the absorption power of the final synthesis product. Accordingly, we considered ways to mitigate the effects of phase transformation as a mean of improving absorption efficiency. For this purpose, we propose a theoretical framework that relies on a simple quantum mechanical Hamiltonian. We suggest that the absence of the $\text{Fhyd} \rightarrow \text{Fe}_3\text{O}_4$ phase transformation, could improve the absorption properties of the final sample ($\text{Fe}_3\text{O}_4/\text{C}$).

Keywords: sol-gel, Hamiltonian, quantum entanglement.

1. INTRODUCTION

Sol-gel synthesis method enables production of high-quality nanoparticles, which could be used for a numerous different applications.

Magnetic nanoparticles, prepared by the sol-gel method (mostly magnetite (Fe_3O_4)), can be used for various military purposes, among others, as adsorbing agents. Accordingly, one of the promising applications of use of material, containing Fe_3O_4 nanoparticles, prepared by sol-gel method, is the absorption of electromagnetic (EM) waves [1]. Based on the chemical composition, it is desirable that the magnetic absorbents are nanocomposite compounds, because single-phase absorbents do not show strong absorption, due to weak magnetic losses [1]. Various coated magnetic nanoparticles can participate in the process of radar absorption in the X-band area, or in the electromagnetic camouflage of submarines [2] (ferrites [1], magnetite, and Ni-ferrite nanoparticles coated by titanium dioxide [3], nanoparticle systems containing silicon nanoparticles [4], composites

containing gadolinium and iron nanoparticles [5], etc.). Literature has shown that radar-absorbing applications (important in the construction of stealth materials) could be improved by using nanoparticle systems, which change color in the presence of a magnetic field [6]. Note that the nanocomposite, consisting of Fe_3O_4 core-shell nanoparticles and mesoporous carbon hollow spheres ($\text{Fe}_3\text{O}_4/\text{C}$), synthesized by sol-gel method, are capable to achieve an ultra-wide absorption band of 8 GHz [7].

On the other hand, sol-gel synthesis procedure is thoroughly investigated with the aim of improving properties of the final synthesis product. For example, in the case of mentioned army application, it is of importance to improve materials absorbing properties. Accordingly, in this article will be discussed theoretical suggestions of the possible modifications of a mentioned synthesis, with the aim of improvement of the absorbing properties of the synthesized samples. Annealing process is step in a synthesis procedure, which determines size of the magnetic nanoparticles and matrix pores, diffusion of gases through the matrix, final structural properties of the

sample, etc. One of the aspects that affects growth of the silica matrix pores (as well as the nanoparticles within the pores), is the phase transformation of the different phases, of which are consisted annealed sample. Noteworthy, the role of phase transformation in sol-gel synthesis has not yet been unambiguously established. It is not known whether the presence of the phase transformations of the sample improves or hinders its final properties.

On the other hand, phase transformations occurs in a different stages of sol-gel synthesis, but in this article, accent will be on the phase transformations, triggered by annealing process. Brief overview on the annealing treatment confirms that annealing treatment represents essential part of sol-gel synthesis, during which nanoparticles growth in the matrix pores. Also, final structure and size of the pores is achieved via heating process. Since literature showed that annealing treatment presents part of sol-gel, which is of the highest importance for defining of absorption properties of the final synthesis product [7], here will be discussed presence/absence of phase transformation during annealing treatment, and its impact on the absorption properties of the final sample.

In order to better understand the role of phase transformations during annealing treatment, this article will discuss whether it is more energetically advantageous to anneal a sample with or without the presence of its phase transformation.

Let us recall that the changes, resulting in phase transformations, are very sophisticated, and that the energy imbalance at the quantum level leads to the appearance of phase transformation. Accordingly, the discussed question will be examined at the quantum level, taking into account the change of the Hamiltonian (H) of the system model. Given Hamiltonian will be introduced to describe the samples, formed in the furnace, with and without the presence of their phase transformation, respectively.

2. RESULTS AND DISCUSSION

2.1. Comments on the synthesis

In a Ref. [7], sol-gel synthesis was performed, in order to synthesize 9 – 10 nm Fe₃O₄ nanoparticles, by using iron nitrate precursor (Fe(NO₃)₃). The solid phase of iron nitrate is unstable in air, and oxidizes very easily, so it is not convenient starting precursor. Iron nitrate in the liquid phase is not so easy to prepare reproducible, due to the difficulty in determining the exact concentration of iron cations, which varies, depending on how much nitric acid is evaporated, which is most often quite arbitrary. Noteworthy, literature has shown that an excess of nitrate anions slower the matrix crystallization (affects the diffusion of gases, and therefore the formation of pores) [8].

Another issue, characteristic for using iron nitrate precursor, is obtaining the final product through phase transformations, occurred during the annealing treatment. Before the annealing treatment, the sample consists of Fhyd nuclei, which is a common name for different types

of oxyhydroxide species. Fhyd formula is determined variously: 5Fe₂O₃·9H₂O, 2FeOOH·2.6H₂O, Fe₅HO₈·4H₂O, Fe₅O₃(OH)₉ or Fe₄O₅(OH)₂·2.6H₂O [9]. Although, from a practical point of view, all the given formulas are essentially equivalent (and for this reason are often reduced to the formula of hydrated iron oxyhydroxide, FeOOH·0.4H₂O), from a quantum point of view, the difference is significant. For simplicity, here will be assumed that only one phase transformation occurred, from Fhyd to Fe₃O₄ nuclei formation.

From the energetic point of view, question of phase transformations is not negligible. Taking into account quantum aspect, phase transformations, started from different formulas, will be characterized by different energy losses. Having on mind that, usually, the exact formula of Fhyd (of which the non-annealed sample consists) is not known, it is very difficult to get a precise insight into the path of the presented phase transformations, and therefore into the exact magnetostructural properties of the prepared magnetite nanoparticle. To avoid the use of iron nitrate precursors, and to improve the reproducibility of the synthesis, the use of other iron precursors, such as iron sulfate, could be suggested.

On the other hand, in order to improve the absorption properties of the samples, this study proposes a modification of the given synthesis. The proposed modification of the synthesis would be based on the application of the seeding method, as part of the sol-gel synthesis, which would enable the growth of magnetite nanoparticles, during the annealing process, without phase transformation (seeding would replace the growth of magnetite nanoparticles via the Fhyd → Fe₃O₄ phase transformation). So, the use of already prepared Fe₃O₄ of smaller size, and their consequent homogeneous dispersion in the alkoxide solution is recommended here, which allows avoiding the mentioned phase transformation during the annealing treatment. As a potential seeds, already prepared Fe₃O₄ nanoparticles, coated with oleic acid (which acts as a stabilizing agent), and synthesized by the solvothermal synthesis method, starting from the FeSO₄·7H₂O precursor [10], could be used. Once when the particles are prepared, the oleic acid layer can be removed by washing the nanoparticles with methanol [11]. Proposed nanoparticles have a regular, spherical shape, characterized by high monodispersity and a narrow particle size distribution of (4.8 ± 1.3) nm. During the annealing treatment, the given nanoparticles can be expected to grow to 9 – 10 nm, which was the size of the particles used in a discussed synthesis [7].

In order to investigate whether the proposed modification of the synthesis method (use of already prepared Fe₃O₄ nanoparticles) can improve the absorption properties of the sample prepared in [7], the influence of phase transformations during the annealing treatment will be considered from a quantum point of view. Recall, Ref. [7] showed that annealing is the most important step to control the absorption properties of the absorber material (a sample annealed at 400°C absorbed in the bandwidth of 5.7 GHz, while annealing the sample, taken from the same bench, at 500°C, extended that bandwidth to 8

GHz). Accordingly, in this study, the sample prepared by annealing at 500°C , during which the phase transformation $\text{Fhyd} \rightarrow \text{Fe}_3\text{O}_4$ [7] occurs, will be designated as S1, and the theoretical sample, synthesized without phase transformation of iron species (which could be prepared by seeding Fe_3O_4 nanoparticles), will be labeled as S2.

2.2. Theoretical model

Heat is the main characteristic of the annealing process. It interacts with the sample (crystal lattice of a solid body) at the quantum level, through the vibrational energy of atoms. During the annealing treatment, heat transfer (via conduction, convection and radiation) occurs through the sample.

Note that this phenomenon is related to the electromagnetic (EM) radiation. During heating a sample, atoms vibrate, and a large number of electrons and other excitations are transported within the crystal lattice, inducing, in that way, changes at the quantum level. As a result of vibration, EM radiation is emitted. In the opposite case, during absorption of EM radiation, absorbed part of the EM wave is turned into heat, which consequently brings to the increased vibration of the atoms, of which is sample consisted of. The sample's temperature rises, with the aim to equalize emission and absorption, in order to achieve thermal equilibrium between the sample and its surroundings. These processes occur simultaneously, which additionally complicates examination of the impact of the phase transformations, induced by annealing treatment, on the energy changes at quantum level. Here is proposed that thermal treatment impacts sample through disturbing quantum entanglement (QE) of the system, which results in its phase transformation.

To get deeper insight in this issue, simplified theoretical model will be postulated.

2.2.1. The form of the model Hamiltonian

Recall here that different forms of Hamiltonian, postulated in literature, are proposed for the interaction between EM and atom. Despite, in the context of considered case (annealing process, as a part of sol-gel synthesis method, and formation of $\text{Fe}_3\text{O}_4/\text{C}$ nanoparticles), a satisfactory theoretical model to describe the Hamiltonian for interaction of heat radiation and atom, has not yet been postulated. In this study, mentioned model will be proposed.

Afterwards, with the aim to comment absorption properties of the investigated samples, it will be discussed Hamiltonian of the interaction of EM radiation and samples, considering contribution of their Hamiltonian, defined during annealing treatment. In order to simplify, it will be used the same form of Hamiltonian for description of interaction between different forms of radiation (heat and EM) with the discussed samples. Such Hamiltonians are consisted of a three terms, describing field, atom, and interaction between atom and field, respectively (in other words, Hamiltonian is defined as:

$$\hat{H} = \hat{H}_{\text{field}} + \hat{H}_{\text{atom}} + \hat{H}_{\text{int}}.$$

To chose appropriate form of model Hamiltonian, recall that temperature difference (i.e., entropy) is related to the quantum entanglement, which depicts to importance of the annealing process for the absorption properties of the samples. In 2021 was experimentally shown that temperature differences in superconductor can be used to entangle pairs of electrons [12], which confirmed that it is possible to entangle electrons with heat. Although it is much complicated to experimentally observe this in the case of high temperatures, it could be assumed that temperature difference (a main characteristic of an annealing treatment), affects quantum entanglement.

The entropy of entanglement is characterized by von Neumann entropy, which presents the Gibbs entropy in the quantum statistical mechanics [13]. On the other hand, as the lowest-order approximation to the von Neumann entropy, which would quantify the degree of coherence losses (in the case of the interaction of the atom, with EM radiation), the degree of entanglement between the subsystems can be quantified by linear entropy [14]. In the literature, coherence loss is understood as the purity loss [14]. Since the purity describes how much a state is mixed, this magnitude is related to the linear entropy, but also, to the phase transformations.

One of the first models, suitable to describe phenomena, appeared in a quantum system as a consequence of the interaction between a two-level atom and an EM field within a cavity, is Tavis-Cummings model. Such a model could be extended to the application in a various solid state systems, and it is used in literature to discuss the interacting Heisenberg spin chain [15]. If a given Hamiltonian is modified, in order to be applicable for N-level atoms (with the constant coupling parameters), Hamiltonian takes a form [14]:

$$\hat{H} = \hbar\eta\hat{\sigma}_z + \hbar\mu\hat{j}_z + \lambda_1\hbar(\hat{\sigma}_- + \hat{\sigma}_+)(\hat{j}_+ - \hat{j}_-) + \lambda_2\hbar(\hat{j}_+ - \hat{j}_-) + \lambda_3\hbar(\hat{\sigma}_- + \hat{\sigma}_+) \quad (2.2.1.1)$$

Here, λ_{1-3} are coupling parameters, $\hat{\sigma}_+$ and $\hat{\sigma}_z$ are two-level atom Pauli operators. In a given expression, third term is given as (2.2.1.2):

$$\hat{H}_{\text{int}} = \lambda_1\hbar(\hat{\sigma}_- + \hat{\sigma}_+)(\hat{j}_+ - \hat{j}_-) + \lambda_2\hbar(\hat{j}_+ - \hat{j}_-) + \lambda_3\hbar(\hat{\sigma}_- + \hat{\sigma}_+) \quad (2.2.1.2)$$

In this case, linear entropy could be recognized as a suitable criterion to monitor the purity loss of the investigated quantum system [14], and it is defined by expression [16]:

$$P(t) = \text{Tr}[\hat{\rho}_a(t)(1 - \hat{\rho}_a(t))] \quad (2.2.1.3)$$

where is $\hat{\rho}_a(t)$ - atomic density operator.

After discussion of the wave function, and calculations of the expectation values of the Pauli operators, equation for the linear entropy for the atomic state is expressed in the form of eq. (2.2.1.4) [14]:

$$P(t) = \frac{1}{2} \left(1 - (\sigma_x^2(t) + \sigma_y^2(t) + \sigma_z^2(t)) \right) \quad (2.2.1.4)$$

This equation clearly indicates importance of a linear entropy (through the impact of Pauli operators) for the total Hamiltonian of a modeled quantum system. Having on mind that linear entropy can be understand as a Shannon entropy, which is directly analogous to the entropy in statistical thermodynamics [17], it is

understandable that it presents important parameter of a closed thermodynamic system in the annealing furnace, which impacts the absorption power of the annealed samples.

2.2.2. The form of the model Hamiltonian, applied in a discussed case

Theoretical model proposed within this study will be discussed relying on second quantization, which enables postulation of the Hamiltonian for solid state body. To discuss impact of the quantum effects on the investigated samples (in this case, nanocomposite absorber, containing Fe_3O_4 nanoparticles), it should be performed formal quantization of the excitation, and quantum corrections will be introduced into the defined form of the Hamiltonian. In other words, instead of position and momentum operators, interacting part of the samples will be described by creation and annihilation operators.

Although heat interacts with the atom through the vibrations of its crystal lattice, and contributes more to the second Hamiltonian term ($\hat{H}_{\text{atom}}$), the emphasis of the postulation of the model Hamiltonian will be on the third term, since it is the term of the Hamiltonian, which describes the interaction between radiation and atoms in the case of heat, as well as in the case of EM radiation. In other words, it is the term of Hamiltonian, whose variation impacts absorption power of the annealed samples. Another reason for devoting special attention to this term of Hamiltonian is a fact, that variation of a synthesis parameters impacts absorption properties of the final sample, while absorption occurs as a result of interaction between atom and radiation. Accordingly, it will be postulated that modification of a third term of a model Hamiltonian ($\hat{H}_{\text{int}}$) is affected due to phase transformation, which occurs as a result of the radiating of a sample with the heat, during annealing treatment. Consequently, in the case of all Hamiltonian investigated in this study, the term describing interaction between radiation and atom, will be considered.

In the postulated model, as part of the sample that interacts with radiation, the excitation (in the form of exciton, created during the annealing treatment, as a result of the interaction of radiation and atoms) of a two-level atom, will be considered. Exciton Hamiltonian will be described as $H(S1)$, in the case of a model system, where a given model atom was subjected to the phase transformation under the impact of the annealing treatment. It will be postulated that the form of $H(S1)$ defines the absorption capability of a sample S1 (literature sample, capable to absorb in a wave range of 8 GHz [7]). Exciton of a model atom, described as $H(S2)$, will refer to the theoretical sample in the system of the furnace, whose formation is not characterized by the presence of phase transformation during annealing treatment.

At atomic level, phase transformation represents re-arrangement of the atoms within crystal lattice, and consequently, re-arrangement of the crystal lattice structure. It is important to note that, at quantum level, phase transformation could be considered only as the energy contribution, appeared due incorporation of the atom in a different position within the crystal lattice.

To simplify model, it will be neglected: changes in the crystal lattice structure (structural transformation of the crystal lattice, due rearrangement of the atoms during phase transformation), exciton-vibron interaction [18], heat quantum nature and interpretation (it will be considered only as radiation), and temporal dimension of the problem (exact time-dependent dynamical operators are still not defined, and due to the simplicity of the problem, time component of the problem will be neglected). Consequently, presence/absence of a phase transformation, occurred during annealing treatment within the sample, will be considered as the only difference between the samples. Model Hamiltonian will be defined with reference on phase transformation. In order to simplify, all discussed Hamiltonian will have the same form.

The form of Hamiltonian of interaction between atom and radiation, described by eq. (2.2.1.2) and applied on the case of interaction between exciton and radiation, will be used to describe $H(S1)$ and $H(S2)$. Formation of exciton triggers re-arrangement of the atoms, atoms positions in crystal lattice, and structure of crystal lattice. Having on mind that energy contribution of the phase transformation appears under the impact of the heat radiation, it contributed to the 3rd term of Hamiltonian, which describes interaction between radiation and atom.

Accordingly, model will discuss energy contribution of appearance of phase transformation, on the Hamiltonian of the excitation. Because of the presence of phase transformation, energy dissipation occurs on the node of the crystal lattice, which reflects on the energy of the atom system, and on the transfer of excitation. Recall that dissipated energy presents a difference between kinetic and potential energy, and describes how much potential energy was irreversible converted into heat. Accordingly, dissipation energy presents energy loss, appeared via phase transformation of the sample S1, during annealing treatment, which is not presented in the case of the S2. This, mathematically, reflects on the Hamiltonian of the samples, during annealing treatment ($H(S1)^{\text{ann}}$ and $H(S2)^{\text{ann}}$), and $H(S1)^{\text{ann}}$ will be characterized by modified $\hat{H}_{\text{int}}$ term, describing energy losses due to the appearance of phase transformation in the system:

$$H(S1)^{\text{ann}} = \hbar\eta\hat{\sigma}_z + \hbar\mu\hat{j}_z + \lambda_1\hbar(\hat{\sigma}_- + \hat{\sigma}_+)(\hat{j}_+ - \hat{j}_-) + [\lambda_2\hbar(\hat{j}_+ - \hat{j}_-) + \lambda_3\hbar(\hat{\sigma}_- + \hat{\sigma}_+) + \hat{H}_{\text{phase tr.}}] \quad (2.2.2.1)$$

$$H(S2)^{\text{ann}} = \hbar\eta\hat{\sigma}_z + \hbar\mu\hat{j}_z + \lambda_1\hbar(\hat{\sigma}_- + \hat{\sigma}_+)(\hat{j}_+ - \hat{j}_-) + [\lambda_2\hbar(\hat{j}_+ - \hat{j}_-) + \lambda_3\hbar(\hat{\sigma}_- + \hat{\sigma}_+)] \quad (2.2.2.2)$$

Modification of the expression for Hamiltonian, by considering the energy changes, induced by phase transformation $\text{Fhyd} \rightarrow \text{Fe}_3\text{O}_4$, requires considering of the dissipation energy, which is estimated by eq. (2.2.2.3):

$$\hat{H}_{\text{phase tr.}} = \hat{T} - \hat{V} \quad (2.2.2.3)$$

Where is $\hat{T}$ - operator of the exciton kinetic energy, and $\hat{V}$ - operator of exciton potential energy. Described dissipation energy represents energy loss, for which reason is $\hat{H}_{\text{int}}^{\text{modif}} < \hat{H}_{\text{int}}$. Accordingly, $H(S2)^{\text{ann}} > H(S1)^{\text{ann}}$. Post-annealed atoms (modeled samples) already contains defined energy

content in the form of Hamiltonians, formed during annealing, and its value is further modified by the same amount of modifications for both atoms, so interaction with EM radiation, reflecting in the form of absorption capability, involves only further modification of the third term.

$$H(S1) = \begin{matrix} H(S1)_{\text{ann}}^{\text{modif}} + \hat{H}_{\text{radiation of the field}}^{\text{modif}} + \hat{H}_{\text{atom}}^{\text{modif}} + \hat{H}_{\text{int}}^{\text{modif}} \end{matrix} \quad (2.2.2.4)$$

$$H(S2) = \begin{matrix} H(S2)_{\text{ann}}^{\text{modif}} + \hat{H}_{\text{radiation of the field}}^{\text{modif}} + \hat{H}_{\text{atom}}^{\text{modif}} + \hat{H}_{\text{int}}^{\text{modif}} \end{matrix} \quad (2.2.2.5)$$

Since the further modified terms of the discussed Hamiltonians are changed for the same amounts, it depicts to the fact that $H(S2) > H(S1)$. In other words, $\hat{H}_{\text{int}}(S2) > \hat{H}_{\text{int}}(S1)$, which depicts to the stronger interaction between atom S2 and radiation at quantum level. Having on mind that the same form of Hamiltonian is applied for description of the post-annealed atoms, it could be proposed that the higher value of $H(S2)$, initiated by the higher value of $\hat{H}_{\text{int}}(S2)$ term, should reflect in the improved absorption properties of the atom S2. In classical Newton mechanics, S2 represents absorber, synthesized by the same synthesis method as sample S1, but during the annealing process, the preparation of sample S2 did not involve its phase transformation.

Here could be considered mechanism, proposing explanation of the QE impact on absorption properties of the sample. It is proposed that the disturbance of QE, and consequent changes in the purity losses, could be described by variations in linear entropy, which contributes to the 3rd term of Hamiltonian, also [15]. On the other hand, 3rd term is a term of Hamiltonian, which describes interaction between atom and radiation. Since disturbance in QE is triggered by heat treatment, which induces, at the atomic level, occurrence of phase transformation, and, in the same time, contributes to the term of Hamiltonian which describes interaction between atom and radiation, it means that QE disturbance impacts absorption properties of the annealed sample.

Recall that linear entropy is related to the entire value of Hamiltonian through Pauli operators (eq. (2.2.1.4), characteristic for 3rd term of Hamiltonian), which means that variations in Hamiltonian value (induced by rearrangement of the atoms, during phase transformation), impacts linear entropy value. Changes in linear entropy, in the same time, reflects changes in the purity losses, induced by variation of QE. Heat in the furnace induces growth of the nanoparticles, and arranges the atoms in various positions, in different crystal lattices (observed as phase transformation). Accordingly, it could be proposed that changes in QE at quantum level (reflected in the variations of a purity losses, and, consequently, linear entropy of the quantum system) impacts absorption properties of the sample.

In other words, in this study is postulated that the improvement of the absorption properties of the samples

prepared by the sol-gel method could be achieved, if the annealing process took place without the phase transformation of the annealed samples. It is proposed that presence of phase transformation affects QE of the samples at the quantum level, which impacts linear entropy. Note that phase transformation and linear entropy are described by the third term of Hamiltonian, which describes interaction between excitation and radiation. Consequently, interaction between heat radiation and excitation induces changes in the linear entropy value, which contributes to the value of the macroscopic entropy magnitude, describing the investigated thermodynamic system of the annealed samples. Impact of presence/absence of phase transformation, on the QE of the investigated samples, is transmitted via entropy value, which further affects final absorption ability of the examined samples (mathematically described, again, via a third term of Hamiltonian).

3. CONCLUSION

This theoretical research was done in order to get deeper insight in the impact of the annealing treatment on the final properties of the annealed samples. It was investigated impact of the presence/absence of phase transformation of samples, prepared by the sol-gel method, on their quantum properties (quantum entanglement), as well as on their macroscopic properties (absorption power of the final samples). The study revealed that quantum entanglement affects the absorption properties of the finite sample, via linear entropy and through the contribution of the third term of the Hamiltonian.

A modification of the sol-gel method, in which the seed method would be used, instead of the synthesis of magnetic nanoparticles from iron precursors, is proposed. This approach would ensure the absence of phase transformation of Fe_3O_4 nanoparticles during the annealing treatment, which has been shown to be energetically more favorable for improving absorption properties of the sample ($> 8 \text{ GHz}$).

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