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Polarization Effect in Electron Paramagnetic Resonance with Anisotropic Effective G-Tensor and Anisotropic Spin Relaxation

S. V. Demishev^{1,2} · A. V. Semeno¹

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Abstract

A simple semi-classical model of magnetization dynamics based on Landau-Lifshits equation of motion with anisotropic effective g-tensor and anisotropic spin relaxation is proposed and applied to the case of electron paramagnetic resonance (EPR). In the Faraday geometry, the model predicts polarization effect consisting in strong dependence of the EPR line shape and magnitude on orientation of vector **h** of oscillating magnetization with respect to the crystal structure, so that EPR may be suppressed for some directions, which are different from standard circular rotation of the magnetization vector around the direction of external magnetic field. In general case, the trajectory of the magnetization vector end is either elongated quasi-ellipse, the position of the main axis of which depends on the magnitude of the external magnetic field, or magnetic oscillations may acquire almost linear character. The model is successfully applied for the quantitative accounting of the polarization effect for EPR mode observed in CuGeO₃ doped with 2% of Co impurity, which remained unexplained for more than 15 years.

Keyword Electron paramagnetic resonance \cdot Anisotropic g-tensor \cdot Anisotropic spin relaxation \cdot Polarization effect \cdot CuGeO₃ \cdot Cobalt impurity

S. V. Demishev demis@lt.gpi.ru

¹ Prokhorov General Physics Institute of the Russian Academy of Sciences, Vavilov street, 38, 119991 Moscow, Russia

² National Research University Higher School of Economics, Myasnitskaya, 20, Moscow 101000, Russia

1 Introduction

In most cases, the anisotropy effects in electron paramagnetic resonance (EPR) are addressed in terms of the g-factor anisotropy. The influence of the structure of the complex with magnetic ion is described by the spin Hamiltonian $\hat{H} = \mu_B(H \cdot \hat{g} \cdot S)$ with the g-tensor \hat{g} (here **H** and **S** are magnetic field and the effective spin) [1]. Another type of anisotropic effects in EPR was reported in [2–4]. It was found that the amplitude of the magnetic resonance may be strongly affected by polarization of oscillating magnetic field **h**. In the germanium cuprate, CuGeO₃, doped with cobalt, for some directions of vector **h** with respect to crystal axes the EPR may be completely suppressed [2–4].

From the theoretical point of view, the considered phenomenon is difficult to explain. Indeed, in Faraday geometry $\mathbf{H} \perp \mathbf{h}$, no strong polarization dependence of the EPR is expected [5-11]. In a semi-classical approach, the magnetization M rotates around **H**, and the trajectory of the **M** vector is a circle in the plane perpendicular to H [5]. For that reason, the EPR mode may be excited by any direction of **h**. The doped CuGeO₃ is an S = 1/2 antiferromagnetic (AF) quantum spin chain system constructed of Cu²⁺ magnetic ions, thus looking quite anisotropic. However, the exact consideration of the EPR problem in this case shows that some polarization effects are possible but small [6, 7]. This is related to the fact that, even in the quantum case [6, 7], the spin Hamiltonian of S = 1/2 AF quantum spin chain containing only exchange and Zeeman terms commutes with the operators of the total spin and its z projection. Therefore, to a first approximation, the resonance will be observed at the frequency of an isolated spin and the excitation of this resonance is independent of **h** even in such strongly interacting system as the quantum spin chain [6, 7]. Some polarization effects may develop when the anisotropic terms determining the EPR line width and g-factor shift are accounted [6, 7]. However, both numerical simulation [8] and analytical computation [10, 11] shows that polarization effects are confined by the cases of the EPR line width and g-factor and no damping of the EPR-like mode for specific vector **h** direction is possible.

The unusual strong dependence of EPR amplitude in $CuGeO_3$:Co had no explanation for more than 15 years. The above argument was considered as a sign that Landau-Lifshits (LL) equation of magnetization motion may be violated in magnets or, at least, is not applicable in the case of cobalt-doped CuGeO₃ [12]. In the present work, we suggest a simple semi-classical model based on LL equation, which may explain magnetic resonance modes with anomalous polarization dependence.

1.1 The Model

The EPR of the S = 1/2 paramagnetic centers with the strongly anisotropic g-tensor were considered in [13, 14] by Bloch equations and density matrix approximation with emphasis on comparison of the CW and pulsed ESR. Another source of the anisotropy of spin dynamics may arise from the anisotropy of relaxation time. The anisotropic spin relaxation was considered for electrons in heterostructures [15], but, to our best knowledge, was not investigated in detail for the case of EPR. In our treatment, we will take into account both anisotropy of the effective g-tensor and relaxation time assuming LL type semi-classical spin dynamics.

In the Landau-Lifshits equation

$$\frac{d\mathbf{M}}{dt} = \hat{\gamma}[\mathbf{M}, \mathbf{H}] - \mathbf{R}(\mathbf{M}) \tag{1}$$

the anisotropy of the effective g-tensor is accounted by tensor $\hat{\gamma}$ and the anisotropy of the relaxation term $\mathbf{R}(\mathbf{M})$ is assumed. When steady magnetic field H_0 and equilibrium static magnetization M_0 are aligned along z-axis, it is possible to write

$$\mathbf{H} = H_0 \mathbf{k} + \mathbf{h}(t)$$
$$\mathbf{M} = M_0 \mathbf{k} + \mathbf{m}(t)$$
(2)

Considering the linearized LL equation for the small oscillating at frequency ω magnetic field $\mathbf{h} \sim \exp(-i\omega \cdot t)$ and small oscillating magnetization $\mathbf{m} \sim \exp(-i\omega \cdot t)$ located in the *x*-*y* plane as described in [5], we come to the system of two equations for m_x and m_y projections of vector \mathbf{m}

$$\begin{pmatrix} -i\omega + v_x & -\omega_x \\ \omega_y & -i\omega + v_y \end{pmatrix} \begin{pmatrix} m_x \\ m_y \end{pmatrix} = \chi_0 \begin{pmatrix} -h_y \omega_x \\ h_x \omega_y \end{pmatrix}$$
(3)

Thus it is supposed that x and y axes in the model magnet are not equivalent in the general case, i.e., $\omega_x = \gamma_x H_0$ and $\omega_y = \gamma_y H_0$ may be different due to different effective gyromagnetic ratios γ_x and γ_y originating from the anisotropic g-tensor. Additionally, the relaxation frequencies ν_x and ν_y describe anisotropic spin relaxation. In accordance with the experimental geometry for the investigation of the anomalous polarization effect [2–4], the oscillating magnetic field is linearly polarized, so that $h_x = h_0 \cos\phi$ and $h_y = h_0 \sin\phi$ (h_0 is the magnitude of oscillating magnetic field). Hereafter, we use notation $\chi_0 = M_0/H_0$.

The solution of the Eqs. (3) is straightforward

$$m_{x} = \frac{h_{x}\omega_{x}\omega_{y} - h_{y}\omega_{x}(-i\omega + v_{y})}{-\omega^{2} + \omega_{x}\omega_{y} + v_{x}v_{y} - i\omega(v_{x} + v_{y})},$$

$$m_{y} = \frac{h_{y}\omega_{x}\omega_{y} + h_{x}\omega_{y}(-i\omega + v_{x})}{-\omega^{2} + \omega_{x}\omega_{y} + v_{x}v_{y} - i\omega(v_{x} + v_{y})}.$$
(4)

Once the components m_x and m_y of oscillating magnetization are known, it is possible to estimate power absorbed in the magnetic resonance $P \sim \text{Im}\{\mathbf{m}\cdot\mathbf{h}^*\}$ [5]. When dimensionless parameters $x = \sqrt{\gamma_x \gamma_y} H_0/\omega$, $\Gamma = \gamma_y/\gamma_x$, $b = v_y/v_x$, $a = v_x/\omega$ and $p = P/\chi_0 h_0^2$ are introduced it is possible to express the resonance absorption curve as

$$p = \operatorname{Im}\left\{\frac{x^2 + x \cdot \sin 2\varphi \sqrt{\Gamma[i(1-\Gamma) + a(\Gamma-b)]/2}}{x^2 - 1 - ia(1+b) + a^2b}\right\}$$
(5)

Thus the parameter Γ is responsible for description of the g-tensor anisotropy, whereas the deviation of the parameter *b* from unity is the measure of anisotropic spin relaxation. The scale *x* is the scale of the dimensionless magnetic field $x = H_0/h_1$, where $h_1 = \omega/\sqrt{\gamma_x \gamma_y}$. The parameter *a* sets the scale of spin relaxation magnitude in the units of the microwave frequency ω .

The second term in the nominator of Eq. (5) describes the polarization effect. The angular dependence of the resonance magnitude in (5) vanishes in the isotropic case $\gamma_x = \gamma_y$ and $\nu_x = \nu_y$ ($\Gamma = 1$, b = 1) so that EPR may be equally excited by any direction of **h** with respect to x and y axes. It is worth noting that besides anisotropy of g-tensor ($\Gamma \neq 1$), the polarization effect develops even for isotropic g-tensor $\gamma_x = \gamma_y$ ($\Gamma = 1$) when spin relaxation is anisotropic $\nu_x \neq \nu_y$ ($b \neq 1$).

In the case of small anisotropy $\Gamma = 1 + \delta\Gamma$, $b = 1 + \delta b$ ($\delta\Gamma$, $\delta b < <1$) and weak resonance damping, the angular dependent amplitude of absorbed power at the resonant field may be estimated as

$$\delta p_{res} \approx \frac{1}{4} (1 + a^2) (\delta \Gamma - \delta b) \sin 2\varphi$$
 (6)

Corrections $\delta\Gamma$ and δb have opposite sign. Therefore, the anisotropy of the parameter γ and anisotropy of spin relaxation in real systems may compensate each other, thus reducing possible polarization effects in real systems. In general case, the influence of various parameters describing anisotropy of spin dynamics may be different and below we will address this problem in more detail.

1.2 Comparison with the Experiment

Before discussing the general properties of the suggested model, it is worth to consider its applicability to available experimental data. Here we consider strong polarization effect reported in CuGeO₃ doped with 2% of Co impurity substituting Cu in chains [2–4]. Spin-Peierls state in CuGeO₃:Co at this doping level is suppressed and temperature dependence of magnetic susceptibility is close to Curie–Weiss law [2–4]. The magneto-optical response of this material is formed by two EPR modes, one of which, mode A corresponding to lower magnetic field, demonstrate polarization dependence. The second mode B observed at higher field behaves as an ordinary EPR almost independent of orientation of oscillating magnetic field **h**. The most pronounced polarization effect corresponds to the case, when external magnetic field is aligned along **a**-axis, i.e., is perpendicular to the **b**-**c** planes where S = 1/2 Cu²⁺ chains are located (Fig. 1a). Hereafter, we will characterize orientation of the vector **h** by an angle ϕ from the **c**-axis (chain direction).

EPR lines A and B are broad and may be resolved without overlapping at frequencies $\omega/2\pi$ exceeding ~ 150–200 GHz in quasi-optical transmission experiments [2–4]. However, in this type of measurement the precise fixing of the **h** direction Fig. 1 Polarization effect in CuGeO₃ doped with 2% of Co impurity. a Orientation of external magnetic field H and oscillating magnetic field h with respect to CuGeO3 crystal axes. b Experimental layout for the polarization effect angular dependence measurement in the cavity experiment. c Experimental EPR spectra (points) and model approximation (lines) for the angular dependence of the polarization sensitive mode of magnetic oscillations. Experimental data are from Ref. [4]. See text for details



may be difficult, and cavity experiments look more prospective. That is why in [2–4] the high-frequency cavity measurements were performed. For enhancement of the microwave frequency to $\omega/2\pi \sim 100$ GHz, the cylindrical cavity was tuned to TE₀₁₄ mode [2–4]. At this frequency, the modes A and B were somewhat overlapped, but spectral resolution was sufficient for performing the polarization measurements.

The anisotropic model considered in the previous section should be compared with the data obtained for anomalous EPR mode A. As long as temperature dependences of parameters in Eq. (5) are unknown, it is reasonable to limit analysis to the case of EPR line angle dependence at fixed temperature. This experiment was performed by rotation of a small CuGeO₃:Co sample placed at the oscillating magnetic field maximum [3] (see the layout in Fig. 1a). The result of this experiment (Fig. 1c) suggests that the mode A reaches maximal amplitude for $\phi = 0$, i.e., when vector **h** is parallel to chain direction (**c**-axis), and this mode may be almost damped in the case hllb. At the same time, the amplitude of the mode B slightly vary with angle ϕ as compared to the behavior of the mode A (Fig. 1c). The "visible" g-factors calculated from the positions of the absorption maxima in Fig. 1c are $g_A \sim 4.4$ and $g_B \sim 2.3$ for the modes A and B, respectively.

The approximation of the experimental data by Eq. (5) requires implementation of several fitting parameters. Besides the ratios a, b and Γ it is necessary to set the magnetic field scale which may be done by introducing an adjustment coefficient h_1 and replacing x in (5) by xh_1 . In addition, it is necessary to assume that the angle scale may be shifted as $\phi - \phi_0$, where angle ϕ_0 mark some "physical" direction in the **b**-**c** plane relevant to EPR parameters anisotropy. In our modeling of experimental data, we assume that if the parameters $a, b, \Gamma, h_0, \phi_0$ are known, the set of the angular dependences must be reproduced just by variation of the angle ϕ , once the amplitude of one curve $P(\phi)$ is fixed. Therefore, strictly speaking, the formal analysis of data in Fig. 1c is six parameters fitting.

However, it is natural to expect that the angle ϕ_0 is connected with some selected direction in CuGeO₃ structure. For the **b**-**c** plane, this direction may be either chain or perpendicular to the chain direction. Our calculations confirmed this idea; the fit of experimental data is possible when $\phi_0 = 0$ and parameter ϕ in theoretical expression (5) is the angle between **h** and **c**-axis as in the experiment (Fig. 1). The amplitude $P(\phi)$ sets the scale of the theoretical dependences magnitude corresponding to various ϕ and therefore appears as an adjustment parameter, which can be easily found if the values of *a*, *b*, Γ and h_1 are fixed. Consequently, the latter four are the most important parameters for experimental data simulation.

It is found that the values a=0.52, b=4, $\Gamma=1.95$ and $h_1=0.143$ provide a reasonable approximation of the resonance A (solid lines in Fig. 1c; the value of h_1 is chosen to give the magnetic field scale in kOe). The calculated value of the error in determining the specified parameters was about 20%. The parameters *a* and *b* give the relaxation frequencies $\nu_x \sim 3 \cdot 10^{11}$ Hz and $\nu_y \sim 1.2 \cdot 10^{12}$ Hz exceeding the excitation microwave frequency $\omega/2\pi \sim 10^{11}$ Hz. The values Γ and h_0 allow estimating γ_x and γ_y corresponding to the anisotropic components of g-tensor in x–y plane: $g_x \sim 8.9$ and $g_y \sim 4.6$. As long as $x = \sqrt{\gamma_x r_y} H_0 / \omega$, the effective g-factor for the A mode is $g_{eff} = \sqrt{g_x g_y} \sim 6.4$, thus being noticeably different from the "visible" value $g_A \sim 4.4$. This discrepancy is apparently due to the expected development of the mode A in conditions $\omega / \nu_x \sim 2$ and $\omega / \nu_y \sim 0.5$ when the resonance position shifts from the resonant field $H_{res} = \omega / \sqrt{\gamma_x r_y} \rho_z$.

The above analysis shows that the model proposed in this paper may describe the polarization effect in real systems. We expect that both anisotropy of the g-tensor and spin relaxation are essential for the quantitative accounting of the angular dependence of the anomalous mode. At the same time, the EPR line shape given by Eq. (5) is noticeably sensitive to the variation of fitting parameters and therefore the exact values of a, b, Γ and h_0 should be verified by first principle microscopic evaluation of anisotropy of EPR parameters in CuGeO₃:Co, which is missing to date. Another source of uncertainty of the model parameters lies in the experimental EPR line shape, which may be dependent on the procedure of baseline subtraction chosen in [2–4].

The analysis provided in the previous section suggests that EPR mode may either demonstrate polarization effect if ($\Gamma \neq 1$, $b \neq 1$) or be independent of orientation of the vector **h** ($\Gamma = 1$, b = 1). Both types of spin dynamics are observed in CuGeO₃:Co. In addition, magnetic oscillations in the modes A and B occur at different frequencies. Interesting that according to universal scenario of doping, impurity substituting Cu in CuGeO₃ does not give rise to an extra EPR mode but modifies the EPR on S=1/2 AF quasi-one = dimensional spin chain [12, 16]. As long as Co impurity in CuGeO₃ replaces Cu²⁺ site [12], the presence of two EPR modes A and B indicates the departure from the expected behavior. According to the available data, the mode B may be attributed to collective EPR on Cu²⁺ spin chains [2–4, 12]. Consequently,

the mode A may be associated with either Co^{2+} impurity in strongly anisotropic surroundings or with some anisotropic regions of the CuGeO₃ matrix caused by doping. In any case, the CuGeO₃ doped with Co is supposed to contradict to the universal scenario of CuGeO₃ doping [12, 16]. The experimental check of this opportunity is the subject of future investigations.

1.3 EPR with anisotropic g-tensor and spin relaxation

Although the model considered is very simple, our investigation of EPR polarization dependence in CuGeO₃:Co shows that it may apply to real experimental systems. The general character of suppositions leading to spin dynamics expressed by the system (3) may indicate that some magnets apart doped germanium cuprate may also demonstrate polarization effect. For that reason, in the present section some general properties of EPR described by Eq. (5) are considered. For simplification of our analysis, the values a=0.52 and $h_1=0.143$ found for CuGeO₃:Co are fixed and we will vary parameters b and Γ responsible for the angular dependence. Below in



Fig. 2 Angular dependence of the EPR line (left) and corresponding contour maps (right) for the model parameters a=0.52 and $h_1=0.143$. **a** Isotropic case ($b=1, \Gamma=1$). **b** Anisotropic case ($b=4, \Gamma=1.95$)

Figs. 2–4, the origin of the angle axis $\phi = 0$ corresponds to the maximum amplitude of the magnetic resonance.

The comparison of evolution of the EPR line shape with the angle ϕ for the trivial case (*b*=1 and Γ =1) and for the best fit describing experiment in CuGeO₃:Co (*b*=4 and Γ =1.95) is presented in Fig. 2. It is visible in contrast to the isotropic case (Fig. 2a) that anisotropic g-tensor and spin relaxation leads to strong modulation of the EPR magnitude and 180° symmetry of polarization effect in reasonable agreement with the available data (Fig. 2b).

It is interesting to consider influence of the parameters b and Γ separately, i.e., to compare cases of anisotropic g-tensor and isotropic spin relaxation $(b=1 \text{ and } \Gamma \neq 1)$ and anisotropic spin relaxation and isotropic g-tensor $(b \neq 1 \text{ and } \Gamma = 1)$. To start, let us estimate EPR line shape for b=1, $\Gamma=1.95$ (Fig. 3a) and b=4, $\Gamma=1$ (Fig. 3b). Both parameters provide the same type of polarization effect symmetry, but, as it was pointed out above, anisotropy of b and Γ affect line shape in the opposite way: the maxima for b=4, $\Gamma=1$ (Fig. 3b) corresponds to the minima for b=1, $\Gamma=1.95$ (Fig. 3a) of EPR line amplitude [see also Eq. (6)]. In addition, the



Fig. 3 Angular dependence of the EPR line (left) and corresponding contour maps (right) for the model parameters a=0.52 and $h_1=0.143$. Panels **a-b** display influence of the g-tensor and spin relaxation anisotropy. **a** b=1, $\Gamma=1.95$. **b** b=4, $\Gamma=1$

anisotropic g-tensor may provide strong angular dependence of the EPR line width without introducing any anisotropy of spin relaxation (Fig. 3a). For cases of bigger anisotropy b=1, $\Gamma=10$ (Fig. 4a) and b=10, $\Gamma=1$ (Fig. 3b), the effect of EPR line distortion caused by anisotropy of the g-tensor becomes more pronounced, whereas the polarization effect caused by anisotropic spin relaxation remain qualitatively the same, although line become broaden with respect to the situation when b=4 (Fig. 4b and Fig. 3b).

It is worth noting that in the general anisotropic case $b \neq 1$ and $\Gamma \neq 1$ the function $p(H_0,\phi)$ defined by Eq. (5) may change sign and become negative for certain regions of the angle and magnetic field (Figs. 2, 3, 4). The corresponding areas marked by dark violet may be clearly seen, for example, in the contour maps at the right parts of Fig. 4a-b. Therefore, the interaction of oscillating magnetic moment with the alternating electromagnetic field in the anisotropic case appears complicated. The case $p(H_0,\phi) > 0$ corresponds to absorption of radiation energy in EPR, which is always the case for a spin system with isotropic parameters (Fig. 2a). The opposite situation



Fig. 4 Angular dependence of the EPR line (left) and corresponding contour maps (right) for the model parameters a=0.52 and $h_1=0.143$. Panels **a-b** display influence of the g-tensor and spin relaxation anisotropy. **a** b=1, $\Gamma=10$. **b** b=10, $\Gamma=1$

 $p(H_0,\phi) < 0$ suggests that energy is transferred from oscillating magnetic moment to alternating electromagnetic field. This specific situation is a direct consequence of anisotropy of the g-tensor and spin relaxation. As a result, in a magnet, where anisotropic model of spin dynamics is applicable, it is possible to expect extra opportunities for radiation control, and likely for construction of amplifiers. However, in a real magnet with anisotropy the complicated $p(H_0,\phi)$ dependence may be superimposed on non-resonant background caused by different absorption mechanisms and, for that reason, may be masked. The detailed examination of the opportunities provided by possibility reach condition $p(H_0,\phi) < 0$ is the subject of separate publication, which is beyond the scope of the present work.

The EPR with anisotropic parameters possesses specific magnetic oscillations, which are different from standard circular rotation of the magnetization vector around the direction of the external magnetic field. The projection of the vector M onto x-y plane is given by Eq. (4). For definiteness, we use below the parameter values found above to describe the experimental data for $CuGeO_3$:Co, where x-y plane corresponds to b-c plane in the crystal structure. The result of computation for several angles ϕ is presented in Fig. 5a–b. In the general case, the trajectory given by $m_{\rm c}$ and m_{y} is an elongated quasi-ellipse, the position of the main axis of which depends on the magnitude of the external magnetic field (Fig. 5). This result is in agreement with the spin dynamics obtained in anisotropic [13, 14] models earlier. For some angles, the magnetic oscillations acquire almost linear character (Fig. 5a-b). It is worth noting that the hypothesis about linear magnetic oscillations responsible for polarization effect in CuGeO₃:Co was first introduced in [2]. However, the character of magnetic oscillations, which follows from the proposed model, is more complicated than simple linear motion of the vector **M** expected from the qualitative analysis of the experimental data [2].

2 Conclusion

In the present work, we considered a simple model of magnetization dynamics based on Landau-Lifshits equation of motion with anisotropic g-tensor and spin relaxation and applied it to the case of electron paramagnetic resonance. In the Faraday geometry, the model predicts polarization effect consisting in strong dependence of the EPR line shape and magnitude on orientation of vector **h** of oscillating magnetization with respect to the crystal structure, so that EPR may be suppressed for some directions of h. The EPR with anisotropic parameters possesses specific magnetic oscillations, which are different from standard circular rotation of the magnetization vector around the direction of external magnetic field. In the general case, the trajectory of the magnetization vector end is either an elongated quasi-ellipse, the position of the main axis of which depends on the magnitude of the external magnetic field, or magnetic oscillations may acquire almost linear character. The model is successfully applied for the quantitative accounting of the polarization effect for EPR mode observed in CuGeO₃ doped with 2% of Co impurity, which remained unexplained for more than 15 years. The simplicity and general character of the proposed model indicates that the search for the magnets with anisotropic EPR parameters causing



Fig.5 Magnetic oscillations for different alignments of vector **h**. Simulation is made for parameters a=0.52, $h_1=0.143$, b=4, $\Gamma=1.95$ relevant to the case of CuGeO₃:Co. a $-\phi=22.5^{\circ}$, b $-\phi=45^{\circ}$. Together with the m_x , m_y plot the resonance curve with the same parameters is drawn

polarization effect could be rewarding. It is worth noting that our consideration should correspond to the quantum mechanical treatment of anisotropic S=1/2 paramagnetic particle problem studied in [13, 14] although we accounted effect of the

effective g-tensor anisotropy in a more simple form. The comparison of the different anisotropic models may be another interesting topic for investigation of the anisotropic EPR, which will be the subject of further study.

Author Contributions SVD and AVS: wrote the text and designed figures. SVD: proposed the model and analyzed it general properties. AVS: applied the proposed model for explaining of the experimental data for CuGeO₃ crystals doped with Co impurity.

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Declarations

Conflict of Interest The authors declare no competing financial or non-financial interests.

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