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Angular and temperature dependencies of the electron paramagnetic resonance and paramagnetic relaxation characteristics in $KCuF_3$ single crystal

N. P. Fokina, E. Kh. Khalvashi, and M. O. Elizbarashvili

Abstract— The paper is devoted to the electron paramagnetic resonance (EPR) in the magnetically quasi one-dimensional crystal $KCuF_3$, the representative of the interesting family of the magnetically concentrated compounds with Cu^{2+} magnetic ions. In this crystal the dominating isotropic exchange coupling acting together with the spin-orbit interaction creates Dzyaloshinsky-Moriya interaction, which determines the anisotropic properties of EPR. The one-dimensionality brings about the short range spin correlations surviving to room and higher temperatures. These correlations are taken here into account not only in the analytical calculation of the EPR exchange narrowed linewidth by Kubo-Tomita method, but also in that of the resonance field. The temperature and angular dependencies of the EPR linewidth agreeing well with the experimental plots and the temperature dependence of the resonance field are plotted. The anisotropic zero-field relaxation rates measurable in the Gorter type of experiments are obtained analytically; their connection with the EPR linewidth is discussed, their angular dependencies are plotted. The zero-field relaxation rate measurements allow duplicating the EPR results at the availability of the both types of experiments with the help of the obtained formulae

Index Terms— EPR line width, quasi-1D magnetic systems, short range spin correlations, zero-field relaxation times.

I. INTRODUCTION

Magnetically concentrated compounds with Cu^{2+} magnetic ions attract much attention because of their interesting anisotropic properties. For instance, Moriya [1] had discovered anisotropic superexchange interaction, which is known also as Dzyaloshinsky-Moriya (DM) interaction, because of the impossibility to explain the magnetic properties of the $CuCl_2 \cdot 2H_2O$ compound without it. As is well-known [1], the DM interaction is a combined effect of spin-orbit coupling and isotropic exchange interaction. So, the role of the strong isotropic exchange interaction is crucial in the EPR experiments described and interpreted below.

Among the various compounds with the Cu^{2+} ions the quasi one-dimensional (1D) spin systems attract considerable interest owing to the quantum mechanical nature of their ground state. EPR of the quasi-1D cuprate $Sr_{0.73}CuO_2$ was studied in [2]. There the electron paramagnetic resonance (EPR) measurements provided evidence that a small spontaneous ferromagnetic moment (weak ferromagnetism typical for DM) occurs below T_C . So, the supposition was adduced that the DM interaction takes place in $Sr_{0.73}CuO_2$. Also in the quasi-1D spin-Peirls compound $CuGeO_3$ the anisotropic EPR characteristics are well explained in [3], when DM interaction is considered as a main perturbation term.

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The compound $KCuF_3$ has quasi-1D magnetic properties in spite of its pseudo-cubic crystal structure, too – the



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isotropic exchange constants in Cu^{2+} ion chains satisfy the relation $J_{interchain} / J_{intrachain} \approx 0.01$ [4]. As it was shown in [4] from both the theoretical and experimental point of view, the DM interaction with DM vector perpendicular to the c -axis of the crystal determines there the EPR linewidth.

Since the presence of the dominating isotropic exchange interaction is the important mutual feature of CuGeO_3 and KCuF_3 compounds, the Kubo-Tomita (KT) theory [5] of the magnetic resonance broadening in the case of the exchange narrowing was used for the interpretation of the temperature and angular dependencies of the EPR line in both papers [3], [4].

It appeared that the KT formula with the high-temperature second moment does not give the adequate quantitative interpretation. According to [3], [4], the short-order spin correlations surviving in these quasi-1D compounds till room and even higher temperatures are the reason of this discordance. The corresponding steps were made in analytical calculations of [3], [4]. So, we'd like to take these correlations into account in the EPR linewidth calculation in KCuF_3 , but in a slightly advanced way as compared to [3], [4] and apply this also to the resonance field calculation, thus obtaining the temperature dependencies of EPR linewidth and the resonance field.

On the other hand, the investigations [6], in which also the paramagnetic relaxation anisotropy was first revealed, were carried out by means of the calorimetric version of the Gorter method in the zero external constant magnetic field on a $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ crystal. It was shown by De Jong and Verstelle [6] (see also the references therein) on the basis of their own experimental data and the data of the other authors that the EPR half width on the half height (hereafter – linewidth) in $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ at the constant field direction along any crystal axis is equal to the half sum of the zero-field relaxation rates measured by means of the Gorter method for the two other crystal axes, and the formula is brought describing this relation. So, in the given paper we suppose to obtain the zero-field relaxation rate angular dependencies for KCuF_3 , too.

II ANGULAR AND TEMPERATURE DEPENDENCIES OF THE EPR AND PARAMAGNETIC RELAXATION CHARACTERISTICS IN KCuF_3

Our consideration is based on the KT theory [5], which describes the influence of the perturbation \mathcal{H}' of the Zeeman interaction on the linewidth and the resonance field of the magnetic resonance. \mathcal{H}' can be generally presented, as

$$\mathcal{H}' = \sum_{\gamma} \mathcal{H}'(\omega_{\gamma}),$$

where $\mathcal{H}'(\omega_{\gamma})$ is the part of \mathcal{H}' causing the spin transitions with the frequency ω_{γ} . Since the strong isotropic exchange interaction along the Cu^{2+} chains takes place in KCuF_3 , the KT results for the strong exchange narrowing of EPR lines are relevant for our problem. According to the KT theory, at the strong exchange narrowing ($\omega_{\gamma}^2 \tau_c^2 \ll 1$, $\tau_c = J / \hbar \equiv \omega_{ex}^{-1}$ is the correlation time of the isotropic exchange interaction with the constant J), the following expression for the Zeeman resonance line contour is valid

$$I(\omega - \omega_z) = \frac{1}{\pi} \frac{(1/T_2)}{(\omega - \omega_z - \delta_z)^2 + (1/T_2)^2}. \quad (1)$$

Here $\omega_z = g\mu_B\mu_0 H_0 / \hbar$ is the Zeeman frequency, μ_B is the Bohr magneton; g -is the g -factor of the magnetic ions; \hbar and μ_0 are the Planck and magnetic constants, correspondingly. H_0 is the strength of the external constant field directed along the Z axis; it is supposed that it is weak compared to the exchange field, but the corresponding Zeeman frequency ω_z is insignificantly (2-3 times, as in [3,4]) larger compared to the value of



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Volume 5, Issue 3, May 2016

$1/T_2$. The \mathcal{H}' terms with $\gamma = 0$ (secular perturbation effect, $\omega_{\gamma=0} = 0$) and with $\gamma \neq 0$ (nonsecular effect, $\omega_{\gamma \neq 0} \neq 0$) enter the values of the line width $\Delta\omega_Z$ and the resonance frequency shift δ_Z :

$$1/T_2 = \Delta\omega_Z = \sum_{\gamma} \sigma_{Z\gamma}^2 \tau_c \quad \delta_Z = -\sum_{\gamma} \sigma_{Z\gamma}^2 \omega_{\gamma} \tau_c^2. \quad (2)$$

For the case, when the main term of the Hamiltonian is the Zeeman interaction of the equivalent spins, and for any perturbation \mathcal{H}' the following expression for $\sigma_{Z\gamma}^2$ is valid [7]:

$$\left(\sigma_{Z\gamma}^2\right) = -\hbar^{-2} \left\langle \left[S^+, \mathcal{H}'(\omega_{\gamma}) \right] \left[S^-, \mathcal{H}'(\omega_{\gamma}) \right] \right\rangle / \left\langle S^+ S^- \right\rangle, \quad (3)$$

where $S^{\pm} = S^X \pm iS^Y$ and $S^{X,Y}$ are the spin components along the X, Y axes.

According to [4], DM with the Hamiltonian $\mathcal{H}_{DM} = \sum_{i<j} \mathbf{d}_{ij} [\mathbf{S}_i \times \mathbf{S}_j]$ with $\omega_{\gamma} = 0, \pm\omega_z$ is the solely

dominating interaction of \mathbf{S}_i and \mathbf{S}_j spins in \mathcal{H}' of the $KCuF_3$ crystal. So, according to [5,7], in our case

$$\sum_{\gamma} \left(\sigma_{Z\gamma}^2\right)_{DM} = \left(M_2^{+-}\right)_{DM} = -\hbar^{-2} \left\langle \left[S^+, \mathcal{H}_{DM} \right] \left[S^-, \mathcal{H}_{DM} \right] \right\rangle / \left\langle S^+ S^- \right\rangle,$$

the linewidth is equal to $\Delta\omega_Z = \left(M_2^{+-}\right)_{DM} / \omega_{ex}$, and the resonance frequency shift is equal to

$$\delta_Z = -\left\{ \sigma_{z(+1)}^2 \Big|_{DM} - \sigma_{z(-1)}^2 \Big|_{DM} \right\} \omega_Z \tau_c^2. \quad (4)$$

The following should be taken into account at the calculation of $\left(M_2^{+-}\right)_{DM}$ and $\left(\sigma_{Z\gamma}^2\right)_{DM}$: According to [3], [4], the short-order spin correlations survive in $KCuF_3$ (as well as in $CuGeO_3$) till the room and even higher temperatures. For taking into account the effect of these correlations at the calculation of $\left(M_2^{+-}\right)_{DM}$ we use the results of [3] for the four-spin correlations, as well as at the calculation of $\left\langle S^+ S^- \right\rangle = 2 \left\langle \left(S^X\right)^2 \right\rangle$ – the Fisher's results [8] for one-dimensional magnetic systems:

$$\left\langle \left(S^X\right)^2 \right\rangle = \left\langle \left(S^Y\right)^2 \right\rangle = \frac{NS(S+1)}{3} \frac{1+u(K_Y)}{1-u(K_Y)}, \quad (5)$$

where $u(K_Y) = \coth(K_Y^{-1}) - K_Y$, $K_Y = k_B T_L / 2|J|S(S+1)$, k_B is the Boltzmann constant, T_L is the lattice temperature. Here we'd like to note that the subscript "Y" in K_Y means that we use the definition of K according to [3], which is different from that of [8]. As a result, the following temperature dependence of the DM second moment is obtained:

$$M_{2DM}^{+-}(K_Y) = M_{2DM}^{+-}(T_L \rightarrow \infty) f(K_Y), \quad (6)$$

where

$$f(K_Y) = 3K_Y u(K_Y) \frac{1-u(K_Y)}{1+u(K_Y)},$$

$$M_{2DM}^{+-}(T_L \rightarrow \infty) = \frac{S(S+1)}{3\hbar^2 N} \sum_{i<j} \left\{ \left(d_{ij}^X\right)^2 + \left(d_{ij}^Y\right)^2 + 2\left(d_{ij}^Z\right)^2 \right\}. \quad (7)$$

The second moment can be presented as $M_{2DM}^{+-}(K_Y) = (1/2) \left[M_{2DM}^X(K_Y) + M_{2DM}^Y(K_Y) \right]$, where

$$M_{2DM}^{X,Y}(K_Y) = -\hbar^{-2} \left\langle \left[S^{X,Y}, \mathcal{H}_{DM} \right]^2 \right\rangle / \left\langle \left(S^{X,Y}\right)^2 \right\rangle = \frac{S(S+1)}{3\hbar^2 N} \sum_{i<j} \left\{ \left(d_{ij}^Z\right)^2 + \left(d_{ij}^{Y,X}\right)^2 \right\} f(K_Y).$$



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Volume 5, Issue 3, May 2016

The second moment $M_{2DM}^{+-}(K_Y)$ determines the linewidth of an EPR with the spin quantization along the constant field (Z axis) and precession with the varying magnetization components M^X and M^Y .

However, in the Gorter type experiments in the zero constant field and the low-frequency field $\vec{H}_1 \cos \omega t$ (with ω of the relaxation rate order) along the X axis, the spin quantization is along the X axis, at that magnetization relaxes to its instantaneous value $\chi_0 H_1 \cos \omega t$ [9]. Both the forced and relaxational motions are effectively linear along this axis. This fact is reflected by the Debye-like formulae of the corresponding dynamic susceptibilities [9]. So, the relaxation rates measurable in the zero-field Gorter type experiments with the low-frequency field along X,Y axes are equal to $T_{X,Y}^{-1} = M_{2DM}^{X,Y}(K_Y) / \omega_{ex}$. The comparison of Eqs.

$$\Delta\omega_Z = (M_{2DM}^{+-}) / \omega_{ex}, \quad M_{2DM}^{+-}(K_Y) = (1 / \sqrt{2}) [M_{DM}^X(K_Y) + M_{DM}^Y(K_Y)] \quad \text{and}$$

$T_{X,Y}^{-1} = M_{2DM}^{X,Y}(K_Y) / \omega_{ex}$ gives the relation $(\Delta\omega_Z^{EPR})_{DM} = (T_X^{-1} + T_Y^{-1}) / 2$, which was observed experimentally [6], [10]. The corresponding values for quasi-1D crystals can be obtained accounting for only two nearest neighbors in a spin chain in the second moment expressions. Then, after passing to the crystallographic frame of reference (CFR) with the axes a, b, c in $KCuF_3$ crystal, where according to [4] the DM vector d_{ij}

components are equal to $d_{ij}^c = 0$; $(d_{ij}^a)^2 = (d_{ij}^b)^2 = d_{ij}^2 / 2$, the value of $M_{2DM}^{+-}(K_Y)$ is reduced to

$$M_{2DM}^{+-}(K_Y) = d_{ij}^2 (2 + \sin^2 \theta) f(K_Y) / 8\hbar^2.$$

It should be mentioned that here, as in the paper of Yamada [4], the c axis is perpendicular to the ferromagnetically ordered ab planes; θ is the angle between the constant field and the c axis directions. Taking the g -factor anisotropy existing in $KCuF_3$ into account, the final expression for the peak-to-peak EPR linewidth in kOe in $KCuF_3$ in CFR reads (all values in the r.h.s. are in SI units, as throughout the paper)

$$\Delta H_{pp}^{EPR} (kOe) = l d_{ij}^2 (2 + \sin^2 \theta) f(K_Y) / 8|J|, \quad (8)$$

where l is equal to $l = 20 \sqrt{3} g(\theta) \mu_B$ with $g(\theta) = \sqrt{g_{\square}^2 \cos^2 \theta + g_{\perp}^2 \sin^2 \theta}$ being the g -factor in CFR. Substituting $d_{ij}^c = 0$, $d_{ij}^a = d_{ij}^b$; $d_{ij} = |J| \Delta g / g$; $\Delta g \equiv g_a - 2$, $g_a = (g_{\square} + g_{\perp}) / 2$; $g_{\square} = 2.15$; $g_{\perp} = 2.39$; $|J| = 203K$ from [4], we obtain the following temperature dependence of the $KCuF_3$ EPR linewidth.

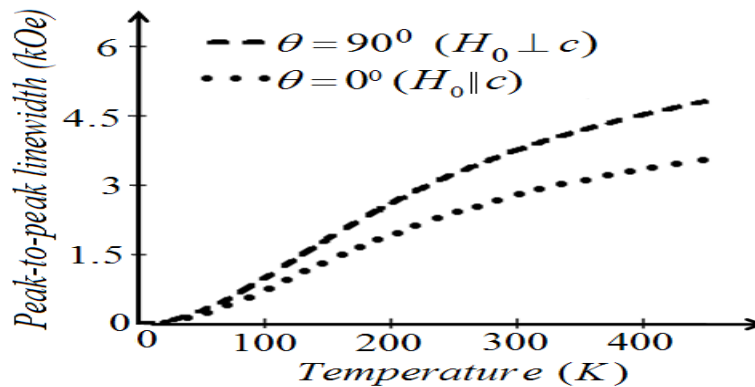


Fig. 1. Temperature dependence of the EPR peak-to-peak linewidth in kOe in $KCuF_3$ for the two directions of the constant magnetic field with respect to the c axis of the sample.



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ISO 9001:2008 Certified

International Journal of Engineering Science and Innovative Technology (IJESIT)

Volume 5, Issue 3, May 2016

One can see that the plot for EPR linewidth quantitatively coincides with the experimental one from [4], except for the vicinity of T_N (this area is out of our consideration). As soon as the experimental EPR linewidth is successfully interpreted in terms of the KT method, it seems to us logically to use the latter also for the EPR resonance field calculation. First, we calculate the resonance frequency shift (4). The straightforward calculation gives:

$$\sigma_{Z(+1)}^2 \Big|_{DM} = 0; \quad \sigma_{Z(-1)}^2 \Big|_{DM} = \frac{S(S+1)}{3\hbar^2 N} \sum_{i < j} \left\{ (d_{ij}^y)^2 + (d_{ij}^x)^2 \right\} f(K_Y). \quad (9)$$

Knowing the resonance frequency shift, the expression for the resonance field corresponding to the resonance frequency $\omega_Z - \delta_Z$ is easily found as (all values in the r.h.s. are in SI units, as throughout the paper)

$$H_{res}^{EPR} (kOe) = (10\hbar\nu / g(\theta)\mu_B) \left\{ 1 - f(K_Y) \left(d_{ij}^2 / 8J^2 \right) (2 - \sin^2 \theta) \right\}^{-1}. \quad (10)$$

Equation (10) can be approximately (accounting for the inequality $d_{ij}^2 / J^2 \ll 1$) rewritten in the form

$$H_{res}^{EPR} (kOe) = (10\hbar\nu / g(\theta)\mu_B) \left\{ 1 + f(K_Y) \left(d_{ij}^2 / 8J^2 \right) (2 - \sin^2 \theta) \right\}.$$

We'd like to note that the shift of the ESR resonance frequency in the saturated antiferromagnet Cs_2CuCl_4 caused by the DM interaction and proportional to d_{ij}^2 / J^2 was theoretically predicted in [11] and experimentally observed in [12]. However, the influence of the DM interaction on the resonance field in $KCuF_3$, predicted by (10) is negligibly small

$$g(\theta) - g_{eff}(\theta) = \sqrt{g_{\parallel}^2 \cos^2 \theta + g_{\perp}^2 \sin^2 \theta} f(K_Y) \left(d_{ij}^2 / 8J^2 \right) (2 - \sin^2 \theta) \square 10^{-3}.$$

According to (10), this resonance field has the following temperature dependence for the experimental conditions of [4]

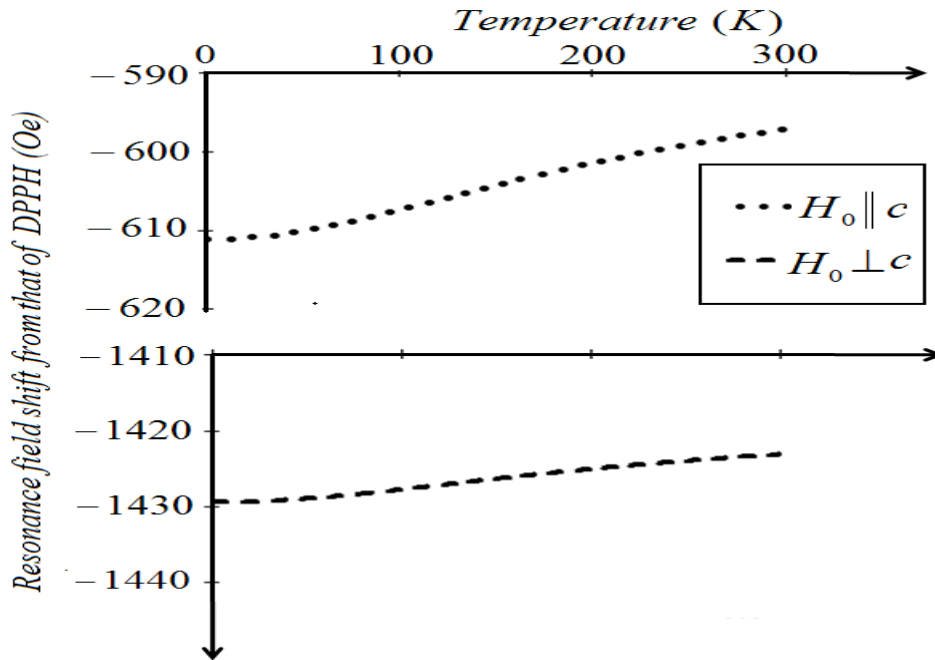


Fig. 2. The temperature dependence of the resonance field H_{res} shift in $KCuF_3$ from that in DPPH

($g = 2$; $d_{ij} = 0$) for the experimental conditions of [4].



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Volume 5, Issue 3, May 2016

It is seen from Fig. 2 that the resonance field is practically temperature independent, as it was really observed for the compound CuGeO_3 , similar to KCuF_3 , in [3].

Now we'd like to discuss the angular dependences of T_X^{-1}, T_Y^{-1} . As it was shown in [13], at EPR T_X^{-1}, T_Y^{-1} are the relaxation rates of X, Y components of the sample macroscopic magnetization. The angular dependencies of T_X^{-1}, T_Y^{-1} with accounting for the short range spin correlations can be easily obtained analytically, giving the following results in CFR:

$$T_X^{-1} = d_{ij}^2 (1 + \sin^2 \theta) f(K_Y) / 4\hbar |J| \quad T_Y^{-1} = d_{ij}^2 f(K_Y) / 4\hbar |J|. \quad (11)$$

For the obtaining the connection between the zero-field relaxation rates $T_a^{-1}, T_b^{-1}, T_c^{-1}$ in CFR measurable by the low-frequency field along the three crystallographic axes a, b, c from [6], [14], [15] and T_X^{-1}, T_Y^{-1} from [10], [16] we consider the following rotations of the constant field (Z axis) with respect to the CFR axes:

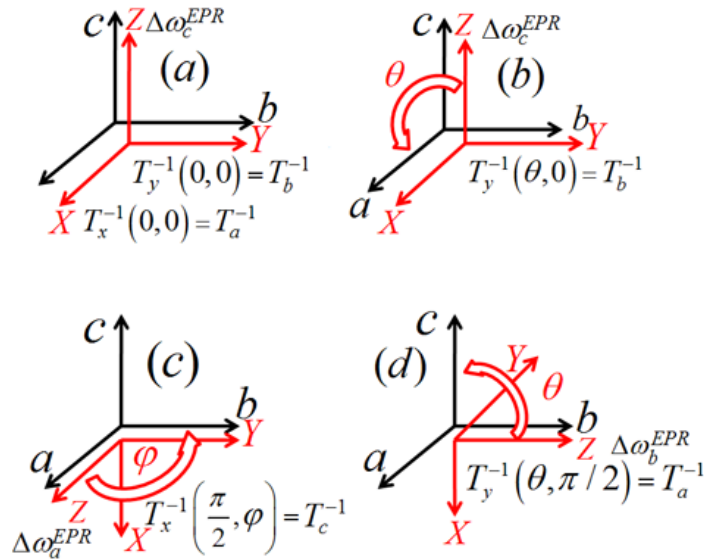


Fig. 3. Connection between the rotation of the laboratory frame of reference ($H_0 \parallel Z$) relatively to CFR and the obtaining $T_a^{-1}, T_b^{-1}, T_c^{-1}$ from T_X^{-1}, T_Y^{-1} .

From the Fig. 3 the following relation between $T_a^{-1}, T_b^{-1}, T_c^{-1}$ and T_X^{-1}, T_Y^{-1} can be written:
 $T_a^{-1} = T_b^{-1} = T_Y^{-1}(\theta)$ in the whole θ variation interval in the planes ac and bc ;
 $T_c^{-1} = 2T_a^{-1} = 2T_b^{-1} = T_X^{-1}(\varphi)$ in the whole φ variation interval in the plane ab .

However, the following instruction of the $T_a^{-1}, T_b^{-1}, T_c^{-1}$ obtaining from the EPR linewidth measured in the samples similar to KCuF_3 at the definite constant field angles relatively to c axis is, to our mind, of the most interest:

$$T_a^{-1} = T_b^{-1} = T_c^{-1} / 2 = \Delta\omega_c^{EPR}(\theta=0) = (2/3)\Delta\omega_a^{EPR}(\theta=\pi/2) = (2/3)\Delta\omega_b^{EPR}(\theta=\pi/2).$$

All these features are seen from the following plot:

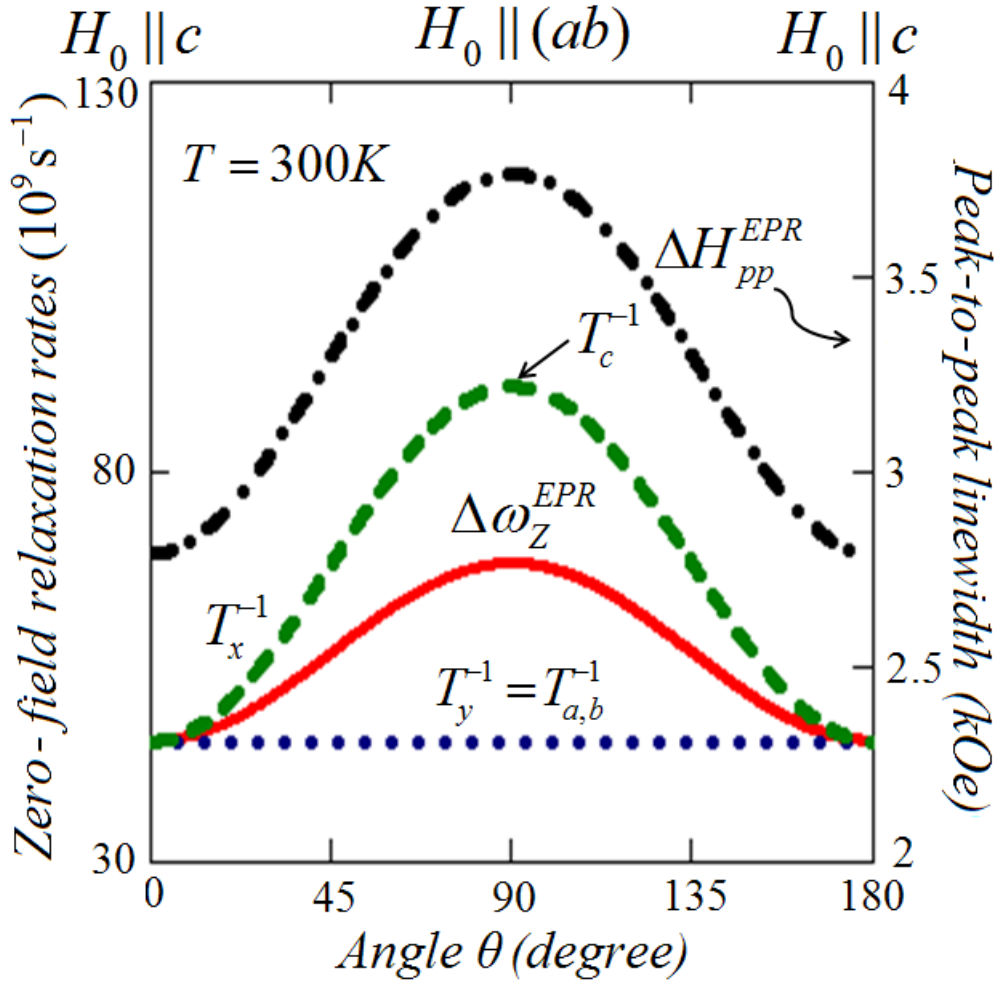


Fig. 4. Dashed green and dotted blue curves describe the angular dependencies of the relaxation rates in of the X and Y Cu^{2+} ion magnetization components T_x^{-1}, T_y^{-1} in KCuF_3 , correspondingly, at the rotation of LFR relatively to the crystallographic axes of the sample for the experimental conditions of [4]. Solid red curve and dash-dotted black curve describe the angular dependence of the EPR linewidth in $\text{s}^{-1}10^9$ and peak-to-peak EPR linewidth in kOe, correspondingly. At the top the positions of LFR Z axis relatively to CFR axes are pointed, $T_L = 300\text{K}$.

The angular dependence of EPR linewidth drawn in Fig. 4 quantitatively agrees with the experimental result of Yamada et al [4].

The measurements of the large relaxation rates in concentrated magnetic compounds was carried out in [17] by the longitudinal response method. The corresponding signals are induced in a longitudinal coil (oriented along the constant magnetic field) under low-frequency modulation of microwave power, which saturates EPR. In the future we suppose to study the relaxation rates obtained by the longitudinal response in the anisotropic materials at the magnetic field rotation with respect to the crystal axes.

III. CONCLUSIONS

Summarizing, the following results are obtained in the given paper under the conditions of experiments of [4] for the single crystal of KCuF_3 :



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Volume 5, Issue 3, May 2016

- the temperature and angular dependencies of the EPR linewidth in $KCuF_3$, plotted according to the KT exchange narrowing theory in the absolute values, however, with the subsequently accounted short-range spin correlations, agree quantitatively with the experimental plots of [4];
- it is shown that the resonance field in $KCuF_3$ experiments is practically temperature independent;
- the anisotropic zero-field relaxation rates measurable in the Gorter type experiments in magnetically concentrated paramagnets under condition of the strong exchange narrowing are obtained. They demonstrate the experimentally observed relation between these quantities and the EPR linewidth. The method of the experimental data processing for the obtaining the values $T_a^{-1}, T_b^{-1}, T_c^{-1}$ from the angular dependence of T_X^{-1}, T_Y^{-1} and that of the EPR linewidth in $KCuF_3$ is described.
- differing from [6], [14], [15], where the consideration was restricted to the CFR, and [10], [16], where the consideration was fulfilled in the LFR, our investigation combines the both approaches.

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AUTHOR BIOGRAPHY

Nathalie P. Fokina:

date of birth – 1947; place of birth – Tbilisi, Georgia; education – M.S. Physics, 1970, Tbilisi Javakhishvili State University, Georgia; Dr. Sci. (Phys.&Math.) – 1995, Institute of Applied Problems of Physics, Erevan, Armenia; employment: 1974-2006 – researcher and senior researcher in Tbilisi Javakhishvili State University; 2013 till now – researcher in Georgian Technical University. Publications: about 50 papers in peer-reviewed scientific journals; member of International ESR (EPR) Society.

Enver Kh. Khalvashi:

date of birth – 1945; place of birth – Batumi, Georgia; education – M.S. Physics, 1968, Tbilisi Javakhishvili State University, Georgia; Dr. Sci. (Phys.&Math.) – 1993, Tbilisi Javakhishvili State University, Tbilisi, Georgia; employment: 1974-2006 – Georgian Technical University Batumi Polytechnic Institute; 2006 till now – Batumi Shota Rustaveli State University. Publications: about 30 papers in peer-reviewed scientific journals.

Maia O. Elizbarashvili:

date of birth – 1970; place of birth – Tbilisi, Georgia; education – M.S. Physics, 1992, Tbilisi Javakhishvili State University, Georgia; Ph.D. – 2005, Tbilisi Javakhishvili State University, Georgia; employment: 2003 till now – researcher and senior researcher in Vladimir Chavchanidze Institute of Cybernetics of the Georgian Technical University. Publications: 6 papers in peer-reviewed scientific journals.