Determination of tetragonal crystalline electric field parameters for Yb^{3+} and Ce^{3+} ions from experimental *g*-factors values and energy levels of Kramers doublets

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The tetragonal crystalline electric field parameters for Yb^{3+} and Ce^{3+} ions are expressed via ground multiplet exited doublets energies and parameters defining doublets' wave functions. The crystalline electric field parameters for Yb^{3+} ion in $YbRh_2Si_2$, $YbIr_2Si_2$ and $KMgF_3$ crystals extracted from excited state doublets energies and *g*-factors of ground state doublet are compared with parameters determined in other works. {***No more than 200 words. Use the MRSej style of paragraph named "_MRSej_Abstract"*** 10 pt Times New Roman}

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Keywords: crystalline electric field parameters, *g*-factors, Yb-based intermetallides, heavy-fermion systems {***"_MRSej_Key"*** 10 pt Times New Roman}

1. Introduction {Use the MRSej style of paragraph named "*_MRSej_Section*" 12 pt Times New Roman Bold}

Our work was initially stimulated by investigation of heavy-fermion Kondo lattice compounds. Very peculiar magnetic, thermal and transport properties of 4*f*-electron based heavy-fermion systems are determined by the interplay of the strong repulsion of 4*f*-electrons on the rare-earth ion sites, their hybridization with wide-band conduction electrons and an influence of the crystalline electric field. {***For first paragraph use the MRSej style of paragraph named "_MRSej_TextNonIndent"*** 11 pt Times New Roman}

In this paper we present the detailed calculation of CEF parameters from energies of ground multiplet exited Kramers doublets and *g*-factors of ground state Kramers doublet. {***Use the basic MRSej style of paragraph named "_MRSe_Text_Main"*** 11 pt Times New Roman with factor 1.1 determining the line spacing}

2. Diagram of Yb^{3+} *g*-factors

A free Yb^{3+} ion has a $4f^{13}$ configuration with one term ^2F. The spin-orbit interaction splits the ^2F term into two multiplets: $^2F_{7/2}$ with $J = 7/2$ and $^2F_{5/2}$ with $J = 5/2$, where J is value of the total momentum $\mathbf{J} = (J_x, J_y, J_z)$. Multiplets are separated by about 1 eV [4]. The Hamiltonian of the Yb^{3+} ion interaction with the tetragonal CEF could be written via equivalent operators $O_k^q(\mathbf{J})$ [4]: {***Below we use the MRSej style of paragraph named "_MRSej_Eq"***}

{***One tab before***} $$H = \alpha B_2^0 O_2^0 + \beta (B_4^0 O_4^0 + B_4^4 O_4^4) + \gamma (B_6^0 O_6^0 + B_6^4 O_6^4), \qquad (1)$$ {***One tab after***}

where B_k^q are the CEF parameters, $\alpha = 2/63$, $\beta = -2/1155$, $\gamma = 4/27027$ [4]. {***After equations we use "_MRSej_TextNonIndent" or "_MRSe_Text_Main"***}

As follows from the group theory, the two-valued irreducible representation $D^{7/2}$ of rotation group contains two two-dimensional irreducible representations of the double tetragonal group [4]. ……………………………………..

Table 1. Energies, wave functions and *g*-factors of Yb^{3+} ion in tetragonal crystalline electric field. {**"*_MRSej_table_caption*"** 11 pt Times New Roman}

$E_{1,2} = -D \pm C/\cos\varphi_7$	$E_{3,4} = D \pm A/\cos\varphi_6$ {**"*_MRSej_tableIn*"**}
$\lvert {}^1\Gamma_7^t \uparrow,\downarrow\rangle = \pm c_1 \lvert \pm 5/2\rangle \pm c_2 \lvert \mp 3/2\rangle$	$\lvert {}^3\Gamma_6^t \uparrow,\downarrow\rangle = \pm a_1 \lvert \mp 7/2\rangle \pm a_2 \lvert \pm 1/2\rangle$
$\lvert {}^2\Gamma_7^t \uparrow,\downarrow\rangle = \mp c_2 \lvert \pm 5/2\rangle \pm c_1 \lvert \mp 3/2\rangle$	$\lvert {}^4\Gamma_6^t \uparrow,\downarrow\rangle = \mp a_2 \lvert \mp 7/2\rangle \pm a_1 \lvert \pm 1/2\rangle$
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The former results correspond to bases $|5/2\rangle$, $|-3/2\rangle$ and $|-5/2\rangle$, $|3/2\rangle$, the latter corresponding to bases $|7/2\rangle$, $|-1/2\rangle$ and $|-7/2\rangle$, $|1/2\rangle$. It is convenient to introduce parameters *C*, *A* and *D*:

$$C = 4B_2^0/21 + 40B_4^0/77 - 560B_6^0/429, \quad A = 4B_2^0/7 + 8B_4^0/77 + 80B_6^0/143. \tag{2}$$

Since matrices (2) are diagonal in the bases of their eigenvectors we can find the relations between our angular parameters and CEF parameters: $\tan\varphi_7 = C_3/C$, $\tan\varphi_6 = A_3/A$, it is enough to take $-\pi/2 \le \varphi_7, \varphi_6 \le \pi/2$.

<u>*EPR spectra of Yb^{3+} ions*</u> {**"*_MRSej_SubSection*"** 12 pt Times New Roman Italic Underline}

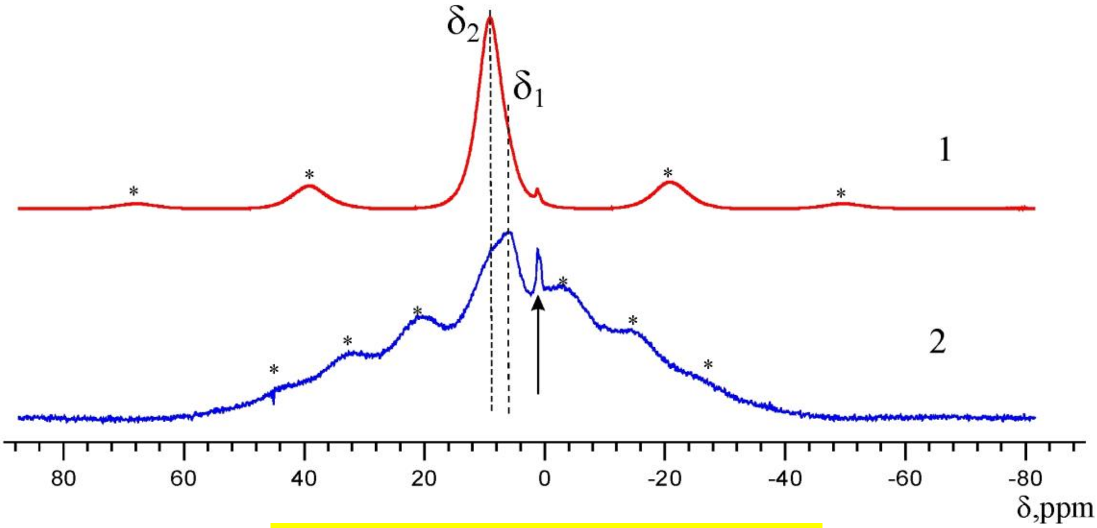
The Zeeman energy $g_J \mu_B \mathbf{HJ}$ in the basis $|\uparrow\rangle$, $|\downarrow\rangle$ of each doublet could be represented by matrix

$$H_{Zeeman} = g_{\parallel}\mu_B H_z S_z + g_{\perp}\mu_B (H_x S_x + H_y S_y), \tag{3}$$

where **H** is the magnetic field, **S** is the effective spin operator with S = 1/2, μ_B is the Bohr magneton, $g_{\parallel}$ and $g_{\perp}$ are *g*-factors when the field is applied parallel and perpendicular to the tetragonal *z*-axis, respectively (tab. 1) The field is applied parallel and perpendicular to the tetragonal *z*-axis, respectively.

<u>*EPR spectra in cubic symmetry case*</u>

In the case of cubic symmetry $\tan\varphi_7 = -\sqrt{3}$, $\tan\varphi_6 = -\sqrt{35}$, $c_1 = \sqrt{3}/2$, $c_2 = -1/2$, $a_1 = \sqrt{7/12}$, $a_2 = -\sqrt{5/12}$. In accordance with expansion $\Gamma_8 = \Gamma_7^t + \Gamma_6^t$ [4] the doublets ${}^2\Gamma_7^t$ and ${}^3\Gamma_6^t$



{**Figures and tables are inserted into tables**}

Figure 1. The diagram of *g*-factors of Yb^{3+} ion in tetragonal crystalline electric field and experimental *g*-points taken from literature (tab. 2). {**"*_MRSej_figure_caption*"** 10 pt Times New Roman}

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Using the least squares method the experimental values of g-factors (tab. 2) and experimental energy of whole ^2F term levels have been taken into account. Obtained CEF parameters satisfy the experimental energy scheme of ^2F term very well, but are reproduced by our expressions (9) only approximately (tab. 5).

Table 2. Experimental g-factors of Yb^{3+} ion in tetragonal crystalline electric field given in figure 1.

	$YbRh_2Si_2$ [1]	$YbIr_2Si_2$ [2]
$\lvert g_{\parallel} \rvert$	0.17(7)	0.85(1)
$\lvert g_{\perp} \rvert$	3.561(6)	3.357(5)

4. Calculation of CEF parameters for Yb^{3+} ion. Comparison with another papers

Let us calculate the CEF parameters for the given exited state doublets energies $\Delta_1 < \Delta_2 < \Delta_3$. It follows from (3) that we find:

$$B_2^0 = \frac{1}{8}b + \frac{3}{4}b_6\cos\varphi_6 + \frac{1}{4}b_7\cos\varphi_7\,,$$

$$B_4^0 = -\frac{1}{4}b + \frac{1}{32}b_6\cos\varphi_6 + \frac{5}{32}b_7\cos\varphi_7\,,\quad B_4^4 = -\frac{7\sqrt{35}}{32}b_6\sin\varphi_6 - \frac{35\sqrt{3}}{32}b_7\sin\varphi_7\,, \tag{9}$$

$$B_6^0 = -\frac{13}{160}b + \frac{39}{320}b_6\cos\varphi_6 - \frac{91}{320}b_7\cos\varphi_7\,,\quad B_6^4 = \frac{117\sqrt{35}}{320}b_6\sin\varphi_6 - \frac{273\sqrt{3}}{320}b_7\sin\varphi_7\,,$$

In paper [7] CEF parameters of Yb^{3+} ion in $KMgF_3$ crystal have been found (tab. 5). Using the least squares method the experimental values of g-factors (tab. 2) and experimental energy of whole ^2F term levels have been taken into account. Obtained CEF parameters satisfy the experimental energy scheme of ^2F term very well, but are reproduced by our expressions (9) only approximately (tab. 5).

In this case $g_{\parallel}$ and $g_{\perp}$ are related by the equation $g_{\parallel} + 2g_{\perp} + 8 = 14p_3^2$, but as the admixture of excited $^2F_{5/2}$ multiplet is small (p_3 = 0.00551 [7]) we obtain previous relation $g_{\parallel} + 2g_{\perp} + 8 = 0$. On the diagram (fig. 1) we marked experimental values of Yb^{3+} g-factors in several crystals (see also tab. 2). This allows us to estimate the signs of g-factors and to make assumptions about the ground state Kramers doublet on the basis of measured absolute values of g-factors. For example, it is evident that the ground state doublet of Yb^{3+} ion in $HfSiO_4$ is $^3\Gamma_6^t$ and both parallel and perpendicular g-factors have a negative sign. The ground state doublet of Yb^{3+} ion in $KMgF_3$ is $^4\Gamma_6^t$, the sign of $g_{\parallel}$ is positive, the sign of $g_{\perp}$ is negative. In CaF_2 crystal the tetragonal center of Yb^{3+} is in state $^1\Gamma_7^t$ and the sign of $g_{\parallel}$ is positive but the sign of $g_{\perp}$ can be both positive and negative.

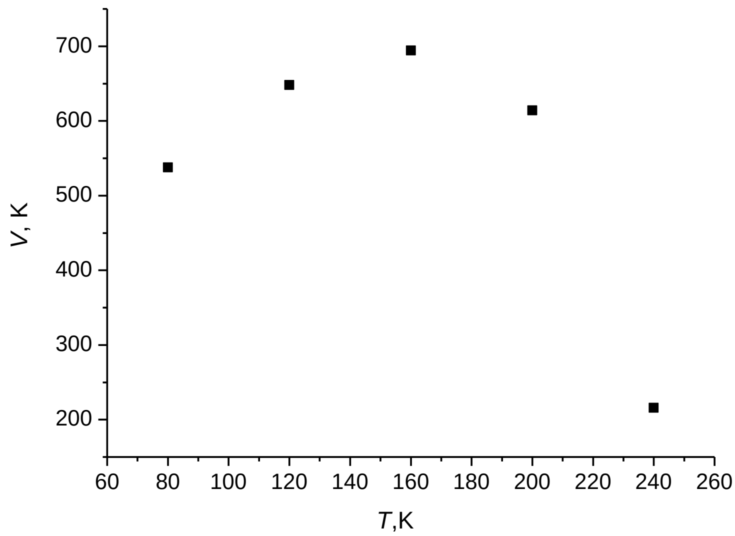

Figure 2. Temperature dependence of the strength of the potential V (see (6)), obtained from the simulation of C_{60} EPR spectra.

4. Summary

For Yb^{3+} and Ce^{3+} ions all possible sets of tetragonal crystalline electric field parameters that satisfy the given experimental energy scheme of ground multiplet are defined.

The earlier published CEF parameters for Yb^{3+} ion in $YbRh_2Si_2$ and $YbIr_2Si_2$ crystals calculated with the use of least squares method could be obtained from our formulas (see tab. 5).

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