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

{***Use style"_MRSej_Title"*** 14 pt Times New Roman Bold}

A.S. Kutuzov [1,] *, A.M. Skvortsova [2] {***"_MRSej_Authors"*** 11 pt Times New Roman}

[1] Kazan State University, Kremlevskaya 18, Kazan 420008, Russia {***"_MRSej_Address"***}

[2] Kazan Federal University, Kremlevskaya 18, Kazan 420008, Russia {***"_MRSej_Address"***}

E-mail: Alexander.Kutuzov@gmail.com* {*"_MRSej_Address"*** *for corresponding author*}

(Received ???, accepted ????) {***"_MRSej_Date"*** 10 pt Times New Roman}

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}

PACS: 75.10.Dg, 76.30.-v, 75.20. {***"_MRSej_PACS"*** 10 pt Times New Roman}

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"***}
$\lvert {}^1\Gamma_7^t\ \uparrow,\downarrow\rangle = \pm c_1 \lvert \pm 5/2\rangle \pm c_2 \lvert \mp 3/2\rangle$	$\lvert {}^3\Gamma_6^t\ \uparrow,\downarrow\rangle = \pm a_1 \lvert \mp 7/2\rangle \pm a_2 \lvert \pm 1/2\rangle$
$\lvert {}^2\Gamma_7^t\ \uparrow,\downarrow\rangle = \mp c_2 \lvert \pm 5/2\rangle \pm c_1 \lvert \mp 3/2\rangle$	$\lvert {}^4\Gamma_6^t\ \uparrow,\downarrow\rangle = \mp a_2 \lvert \mp 7/2\rangle \pm a_1 \lvert \pm 1/2\rangle$
……	……
……	……

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 $-\pi/2 \le \varphi_7, \varphi_6 \le \pi/2$.

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

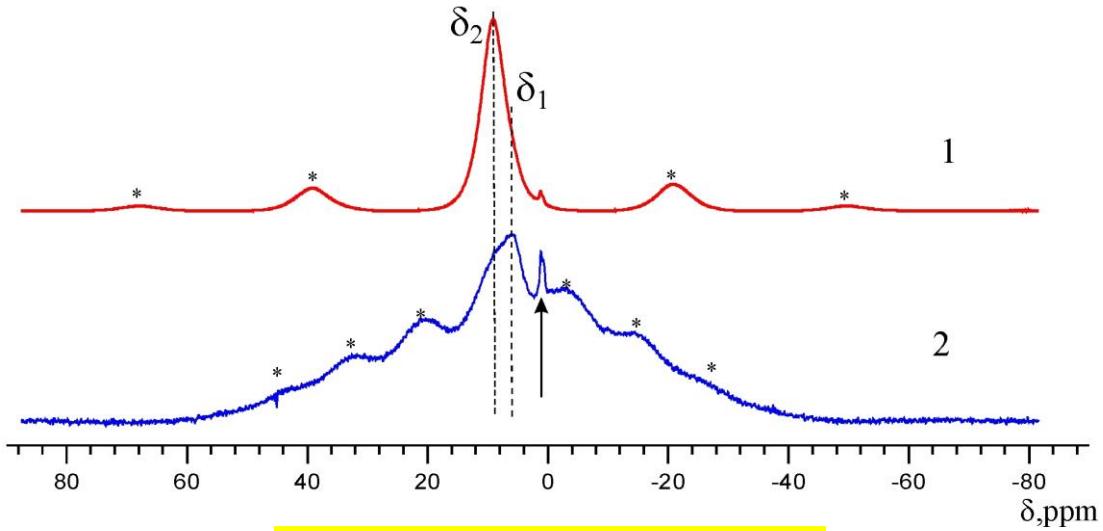
The Zeeman energy $g_J \mu_B \mathbf{HJ}$ in the basis $|\uparrow\rangle$, $|\downarrow\rangle$ of each doublet could be represented by matrix

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

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.

EPR spectra in cubic symmetry case

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}

……

……

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.

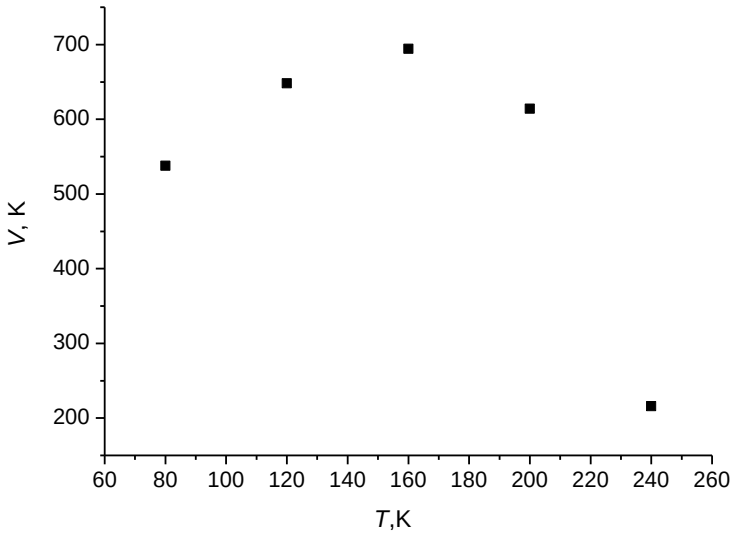


Figure 2. Temperature dependence of the strength of the potential *V* (see (6)), obtained from the simulation of C_{60} EPR spectra.

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).

Acknowledgments

We wish to acknowledge professor ……

This work was supported by ……..

References

1. Sichelschmidt J., Ivanshin V.A., Ferstl J., Geibel C., Steglich F. *Phys. Rev. Lett.* **91**, 156401 (2003) {**"_*MRSej_Reference*"** 11 pt Times New Roman}
2. Abragam A., Bleaney B. *Electron Paramagnetic Resonance of Transition Ions*, Clarendon Press, Oxford (1970)
3. Altshuler S.A., Kozyrev B.M., *Electron Paramagmetic Resonance in Compounds of Transition Elements*, Wiley, New York (1974)
4. Kutuzov A.S., Skvortsova A.M., Belov S.I., Sichelschmidt J., Wykhoff J., Eremin I., Krellner C., Geibel C., Kochelaev B.I. *J. Phys.: Condens. Matter* **20**, 455208 (2008)
5. Leushin A.M., Ivanshin V.A., Kurkin I.N. *Phys. Solid State* **49**, 1417 (2007) (*Fizika Tverdogo Tela* **49**, 1352 (2007), *in Russian*)
6. Bednorz J.G., Müller K.A. *Z. Phys. B* **64**, 189 (1986)
7. See, for instance, *Phase separation in Cuprate Superconductors*, edited by K.A. Müller and G. Benedek (World Scientific, Singapore, 1993)
8. Mihailovic D., Müller K.A., in *High-Tc Superconductivity: Ten Years After the Discovery*, E. Kaldis *et al*., eds. Kluwer Academic Publishers (1997) p. 243
9. Levin L.I., Cherepanov V.I. *Fizika Tverdogo Tela* **25**, 700 (1983) (*in Russian*)
10. Izyumov Yu.A., Proshin Yu.N., Khusainov M.G. *Physics – Uspekhi* **45**, 109 (2002)
11. Elschner B., Loidl A. in *Handbook of the Physics and Chemistry of Rare Earth*, edited by K.A. Gschneidner Jr., L. Eyring, and M.B. Maple (Elsevier Science, Amsterdam, 2000), Vol. **30**, p. 375
12. Kochelaev B.I., Kharakhash'yan E.G., Garifullin I.A., Alekseevskii N.E. *Proceedings of the 18th Congress AMPERE, Nottingham, 1974*, p. 23 (1974)
13. Kochelaev B.I., Tagirov L.R. Khusainov M.G. *Sov. Phys. JETP* **49**, 291 (1979)
14. Vojta M., Sachdev S. *Cond-mat*/9906104
15. Kuhns P. *Proc. of the Physical Phenomena in High Magnetic Fields-IV. Santa Fe, NM, World Scientific*, p. 297 (2002)
16. Grechishkin V.S., *Nuclear Quadrupole Interactions in Solid*, Nauka, Moscow (1973) (*in Russian*)