

# Defining of the Covalence Parameters in $\text{Cs}_2\text{NaYCl}_6:\text{Eu}^{3+}$ and $\text{Cs}_2\text{NaEuCl}_6$ by the Optical Spectra

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The influence of the excited configurations with the charge transfer on the Stark structure of the multiplets of  $\text{Eu}^{3+}$  ion in the elpasolites was investigated with the help of the modified crystal field Hamiltonian.  $\text{Cs}_2\text{NaYCl}_6:\text{Eu}^{3+}$  and  $\text{Cs}_2\text{NaEuCl}_6$  crystal systems were chosen as the subjects of our research. The modified Hamiltonian of the crystal field was for the first time applied to the crystal systems activated by  $\text{Eu}^{3+}$  ions and its use has allowed to reduce considerably the root-mean-square deviation and to define the covalence parameters from optical spectra. The convenient algorithm for determination of errors of the modified Hamiltonian parameters is offered.

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## 1. Introduction

The crystals with the elpasolite structure type cause great interest while studying and growing laser crystals [1–21]. The elpasolites doped by rare-earth elements attract special attention. The main peculiarity of these compoundings is that impurity trivalent rare-earth ions can lay in cubical crystal environment without the need of charge compensation. The local symmetry of the rare-earth ions in these crystals is octahedral ( $O_h$ ), that is why the crystal field Hamiltonian has only two independent parameters  $B_0^4$  and  $B_0^6$ . The small amount of varied parameters and the sufficient quantity of experimental levels allows test various crystal field theories confidently. Therefore cubic elpasolites are convenient systems for studying the optical spectra and the electronic structure of  $4f$ -elements.

Theoretical investigation of similar systems is a vital problem, as it can help to synthesize crystals with pre-designed optical characteristics.

To improve the description of the Stark splitting of multiplets it was offered to apply a spin-correlated crystal field Hamiltonian [17, 18] and to consider the influence of some excited configurations [10, 13, 19, 20]. However, there still are the multiplets for which the description is not satisfactory, and the role of the opposite parity configurations and covalence effects is unclear. These problems can be avoided using the modified crystal field theory [22–24], which considers the influence of the excited opposite parity configurations and the covalence effects on the Stark structure of the multiplets. Such approach allows to receive the good agreement of the theory with experiment and thus define the covalence parameters and the odd crystalline field parameters from optical spectra.

The  $\text{Cs}_2\text{NaYCl}_6:\text{Eu}^{3+}$  and  $\text{Cs}_2\text{NaEuCl}_6$  crystal systems are chosen as the objects of the research. The position of the  $\text{Eu}^{3+}$  ion in these crystals is centrosymmetric. Therefore the adding of excited opposite parity configurations is forbidden and it is possible to investigate in detail only the influence of the excited configurations with a charge transfer and to define covalence parameters.

Accuracy of the determination of freely varied parameters on the basis of the experimental data carries the important information for the correctness of the experimental results and for the adequacy of the model. The Stark energy levels are the eigenvalues of the Hamiltonian and are a rather complicated function of the crystal field parameters. Therefore the determination of errors for the Hamiltonian parameters is a complex problem. In connection with this we offer a convenient algorithm to determine the errors of the parameters.

It was reported earlier about the successful application of the approximation of anomalously strong configuration interaction to the description of the Stark structure of other ions, such as  $\text{Tm}^{3+}$  [23],  $\text{Pr}^{3+}$  [24],  $\text{U}^{4+}$  [25] and to the description of the absorption spectra intensities of an  $\text{Eu}^{3+}$  ion in double tungstates [26, 27].

## 2. Basic formulae

To describe the Stark structure of the multiplets the Hamiltonian of the crystal field obtained in the approximation of weak configuration interaction is often used [28]:

$$H_{cf} = \sum_{k,q} B_q^k C_q^k, \quad (1)$$

where  $C_q^k = \sum_{i=1}^N c_q^k(i)$  is the spherical tensor operator. The crystal field parameters  $B_q^k$  are usually determined on the basis of the experimental data. With the help of the Hamiltonian (1) we can take into account the effect of the excited configurations adding functions of the corresponding excited configurations to the basis.

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However, the influence of the excited configurations can be taken into account easier using the effective Hamiltonian in the approximation of the intermediate configuration interaction [29]:

$$H_{cf} = \sum_{k,q} \underbrace{[B_q^k + (E_J + E_{J'}) G_q^k]}_{\tilde{B}_q^k} C_q^k, \quad (2)$$

where  $E_J$ ,  $E_{J'}$  are the energies of the multiplets,  $G_q^k$  is the parameter caused by the interconfiguration interaction. Here the operator  $C_q^k$  influences the angular variables of electrons only with basic configurations. The generalized crystal field parameters  $\tilde{B}_q^k$  depend linearly on the multiplet energy.

Sometimes the influence of the excited configurations is so strong that for an adequate description of the Stark structure it is necessary to use the crystal field Hamiltonian in the approximation of strong configuration interaction [29]:

$$H_{cf} = \sum_{k,q} \underbrace{\left[ B_q^k + \left( \frac{\Delta^2}{\Delta - E_J} + \frac{\Delta^2}{\Delta - E_{J'}} \right) \tilde{G}_q^k \right]}_{\tilde{B}_q^k} C_q^k, \quad (3)$$

where  $\Delta$  is the energy of the excited configuration.

It should be mentioned that the formula (3) is valid if a decisive contribution to interconfiguration parameters  $\tilde{G}_q^k$  is yielded only by one excited configuration or several excited configurations with close energy  $\Delta$ . If the excited configurations have substantially different energies, the effective Hamiltonian is more complicated [22]:

$$H_{cf} = \sum_{k,q} \left\{ B_q^k + \left( \frac{\Delta_d^2}{\Delta_d - E_J} + \frac{\Delta_d^2}{\Delta_d - E_{J'}} \right) \tilde{G}_q^k(d) + \sum_i \left( \frac{\Delta_{ci}^2}{\Delta_{ci} - E_J} + \frac{\Delta_{ci}^2}{\Delta_{ci} - E_{J'}} \right) \tilde{G}_q^k(c) \right\} C_q^k. \quad (4)$$

The usual defining contribution to the parameters  $\tilde{G}_q^k$  is yielded by  $4f^{N-1}5d$  configurations of opposite parity and configurations with charge transfer.

But as the elpasolites have cubic symmetry (space symmetry  $Fm\bar{3}m$ ), the summand  $\left( \frac{\Delta_d^2}{\Delta_d - E_J} + \frac{\Delta_d^2}{\Delta_d - E_{J'}} \right) \tilde{G}_q^k(d)$  which stands for the opposite parity configuration equals to null, in this case we have to consider only the influence of charge transfer processes, submitted in a Hamiltonian (4) by  $\left( \frac{\Delta_{ci}^2}{\Delta_{ci} - E_J} + \frac{\Delta_{ci}^2}{\Delta_{ci} - E_{J'}} \right) \tilde{G}_q^k(c)$ . Here  $\Delta_{ci}$  is the energy of the excited charge-transfer configurations [29]:

$$\tilde{G}_q^k(c) = \sum_b \tilde{J}^k(b) C_q^{k*}(\Theta_b, \Phi_b), \quad (5)$$

where

$$\begin{aligned} \tilde{J}^2(b) &\approx \frac{5}{28} [2\gamma_{\sigma f}^2 + 3\gamma_{\pi f}^2], \\ \tilde{J}^4(b) &\approx \frac{3}{14} [3\gamma_{\sigma f}^2 + \gamma_{\pi f}^2], \\ \tilde{J}^6(b) &\approx \frac{13}{28} [2\gamma_{\sigma f}^2 - 3\gamma_{\pi f}^2]. \end{aligned} \quad (6)$$

Here  $\gamma_{if}$  ( $i = \sigma, \pi$ ) are the covalence parameters.

During the investigations a great role is played by the determination of the errors of physical quantities, their value allows to make a conclusion about the adequacy of the obtained data and the possibility of its further application. In optical spectroscopy the basic spectroscopic characteristics are expressed through such parameters as the crystal field parameters, intensity parameters, etc. The calculation of these parameters on the basis of complex contemporary theories is a hard problem to solve. No wonder nowadays there are no convenient methods of calculating the standard errors of such parameters in scientific literature. In this context we suggest a new scheme of calculating the standard errors of the parameters of theory in indirect measurements.

We used the general principles of the theory of standard errors in indirect measurements during the formulating of the scheme [30]. Let us assume that the spectroscopic characteristic or any other physical quantity, for example, transition oscillator strength, the energy of the Stark component, etc.

$$f_i = f_i(b_1, b_2, \dots, b_{N_p}) \quad (7)$$

depends on the theory parameters  $b_1, b_2, \dots, b_{N_p}$ , altogether  $N_p$  parameters.

Applying computer methods of minimization or any other methods at first it is necessary to define the optimal parameters  $b_1^{\text{opt}}, b_2^{\text{opt}}, \dots, b_{N_p}^{\text{opt}}$ , which give the minimum root-mean-square deviation between the calculated values and the corresponding experimental data

$$\sigma_{\text{exp}} = \quad (8)$$

$$\sqrt{\frac{\sum_{i=1}^N \left( f_i(b_1^{\text{opt}}, b_2^{\text{opt}}, \dots, b_{N_p}^{\text{opt}}) - f_i^{\text{exp}} \right)^2}{N - N_p}},$$

where  $N$  is the number of experimental data.

According to [30] we will write down Eq. (8) with the help of the optimal values of parameters and their errors  $\Delta b_1, \Delta b_2, \dots, \Delta b_{N_p}$ :

$$\sigma_{\text{exp}} = \quad (9)$$

$$\sqrt{\frac{\sum_{k=1}^{N_p} \sum_{i=1}^N \left( \partial f_i(b_1^{\text{opt}}, b_2^{\text{opt}}, \dots, b_{N_p}^{\text{opt}}) / \partial b_k \right)^2 (\Delta b_k)^2}{N - N_p}}.$$

Here  $\frac{\partial f_i(b_1^{\text{opt}}, b_2^{\text{opt}}, \dots, b_{N_p}^{\text{opt}})}{\partial b_k}$  stands for the derivative calculated with the optimal values of the parameters  $b_1^{\text{opt}}, b_2^{\text{opt}}, \dots, b_{N_p}^{\text{opt}}$ .

In the assumption of the equiprobable contribution of each parameter into the value of  $\sigma_{\text{exp}}$ , we will write down the formula (8) approximately with the help of the error  $\Delta b_j$  of any parameter

$$\sigma_{\text{exp}} \approx \quad (10)$$

$$\sqrt{N_p} \sqrt{\frac{\sum_{i=1}^N \left( \partial f_i(b_1^{\text{opt}}, b_2^{\text{opt}}, \dots, b_{N_p}^{\text{opt}}) / \partial b_j \right)^2}{N - N_p}} \Delta b_j.$$

Using Eq. (10) we can easily calculate the  $\Delta b_j$  error.

However, the basic difficulty is represented by the calculation of the derivatives as it is not always possible to establish the analytical dependence  $f_i = f_i(b_1, b_2, \dots, b_{N_p})$ . For example, the energies of the Stark components are calculated as the result of high-order matrix diagonalization by numerical techniques. Therefore it is more suitable to calculate  $\sqrt{\frac{\sum_{i=1}^N (\partial f_i(b_1^{\text{opt}}, b_2^{\text{opt}}, \dots, b_{N_p}^{\text{opt}})/\partial b_j)^2}{N - N_p}}$  using computer methods. To do this, according to the definition of the differential, we shall write down the approximate equation

$$\frac{1}{\delta b_j} \sqrt{\frac{\sum_{i=1}^N (\partial f_i(b_1^{\text{opt}}, b_2^{\text{opt}}, \dots, b_{N_p}^{\text{opt}})/\partial b_j)^2}{N - N_p}} \approx \frac{1}{\delta b_j} \sigma_{\text{theor}}, \quad (11)$$

where

$$\sigma_{\text{theor}} = \left[ \frac{1}{N - N_p} \sum_{i=1}^N \left( f_i(b_1^{\text{opt}}, b_2^{\text{opt}}, \dots, b_j^{\text{opt}} + \delta b_j, \dots, b_{N_p}^{\text{opt}}) - f_i(b_1^{\text{opt}}, b_2^{\text{opt}}, \dots, b_j^{\text{opt}}, \dots, b_{N_p}^{\text{opt}}) \right)^2 \right]^{\frac{1}{2}}. \quad (12)$$

$\sigma_{\text{theor}}$  can be calculated using the same computer program as  $\sigma_{\text{exp}}$ . For this purpose it is enough to substitute  $f_i^{\text{exp}}$  by  $f_i(b_1^{\text{opt}}, b_2^{\text{opt}}, \dots, b_j^{\text{opt}} + \delta b_j, \dots, b_{N_p}^{\text{opt}})$ , calculated using fixed parameters  $b_1^{\text{opt}}, b_2^{\text{opt}}, \dots, b_j^{\text{opt}} + \delta b_j, \dots, b_{N_p}^{\text{opt}}$  (an increment  $\delta b_j$  is added to the parameter  $b_j^{\text{opt}}$ ).

After the substitution of (11) into (10) for the estimation of the error of  $b_j$  parameter we shall have a convenient formula

$$\Delta b_j = \frac{\sigma_{\text{exp}} \delta b_j}{\sigma_{\text{theor}} \sqrt{N_p}}. \quad (13)$$

Satisfactory results for parameter errors are obtained if we choose the parameter increments  $\delta b_j = 0.001 b_j^{\text{opt}}$  for the calculation of  $\sigma_{\text{theor}}$ .

### 3. The discussion of the results

The local symmetry of  $\text{Eu}^{3+}$  ion in  $\text{Cs}_2\text{NaYCl}_6:\text{Eu}^{3+}$  and  $\text{Cs}_2\text{NaEuCl}_6$  is octahedral  $O_h$ . Therefore the crystal field Hamiltonian has only two independent parameters  $B_0^4$  and  $B_0^6$ . In the approximation of anomalously strong configuration interaction Hamiltonian (4) has additional parameters  $\Delta_{ci}$ , which correspond to the energy of a configuration with charge transfer, and implicitly has covalence parameters  $\gamma_{\sigma f}$  and  $\gamma_{\pi f}$ .

For  $\text{Cs}_2\text{NaYCl}_6:\text{Eu}^{3+}$  and  $\text{Cs}_2\text{NaEuCl}_6$  crystal systems the following crystal field parameters were obtained in the approximation of weak configuration interaction:  $B_0^4 = 1958 \text{ cm}^{-1}$ ,  $B_0^6 = 273 \text{ cm}^{-1}$  and  $B_0^4 = 2055 \text{ cm}^{-1}$ ,  $B_0^6 = 274 \text{ cm}^{-1}$ , respectively. The root-mean-square deviation between the calculated values and

the corresponding experimental data in this approximation is  $9.3 \text{ cm}^{-1}$  for  $\text{Cs}_2\text{NaYCl}_6:\text{Eu}^{3+}$  and  $11.0 \text{ cm}^{-1}$  for  $\text{Cs}_2\text{NaEuCl}_6$  (refer to Table I).

TABLE I

The comparison of experimental [14] and calculated Stark levels in the approximation of weak (1) and anomalously strong configuration interaction (4) for  $\text{Cs}_2\text{NaEuCl}_6$  and  $\text{Cs}_2\text{NaYCl}_6:\text{Eu}^{3+}$  crystal systems.  $E_e = E$  experimental,  $E_{ec1} = E_{\text{exp}} - E_{\text{calc1}}$ ,  $E_{ec2} = E_{\text{exp}} - E_{\text{calc2}}$ . All the units are in  $\text{cm}^{-1}$ .

$2S+1L_J$	$\text{Cs}_2\text{NaYCl}_6:\text{Eu}^{3+}$			$\text{Cs}_2\text{NaEuCl}_6$		
	$E_e$ [14]	$E_{ec1}$ (1)	$E_{ec2}$ (4)	$E_e$ [14]	$E_{ec1}$ (1)	$E_{ec2}$ (4)
${}^7F_0$	0	0.0	0.0	0	0.0	0.0
${}^7F_1$	359	0.0	0.0	360	0.0	0.0
${}^7F_2$	872 1086	7.1 -7.1	7.6 -7.6	875 1091	13.0 -13.0	12.5 -12.5
${}^7F_3$	1798 1900 1964	-2.1 -8.7 2.1	1.1 -10.4 1.1	1804 1904 1964	4.7 -5.5 -4.7	4.9 -7.2 -4.9
${}^7F_4$	2660 2975 3006 3036	0.1 4.9 0.9 -0.1	0.6 4.5 -4.0 -0.6	2665 2976 3008 3034	13.4 7.2 -3.3 -13.4	-3.7 -5.2 1.3 3.7
${}^7F_5$	3817 - 3877 -	-7.8 (3860.8) 7.8 (4133.3)	-6.1 (3858.8) 6.1 (4135.7)	3812 3851 3877 4150	-9.6 -5.7 11.7 9.6	-9.8 -5.2 3.1 9.8
${}^7F_6$	- - - - -	(4914.7) (4944.4) (5011.6) (5267.9) (5305.0) (5337.0)	(4908.9) (4939.2) (5006.7) (5278.3) (5315.6) (5347.6)	4835 4869 4945 5242 5254 5283	-2.0 1.0 8.4 30.6 5.4 2.0	-1.2 1.8 10.2 16.3 -8.8 1.2
${}^5D_0$	17206	0.0	0.0	17208	0.0	0.0
${}^5D_1$	18961	0.0	0.0	18960	0.0	0.0
${}^5D_2$	21381 21495	-22.8 22.8	-3.0 3.0	21383 21493	-19.1 19.1	0.7 -0.7
${}^5D_3$	24263 24296 -	-6.7 6.7 (24335.3)	1.9 -1.9 (24349.6)	24277 24310 -	-6.0 6.0 (24351.7)	4.8 -4.8 (24353.1)
$\sigma$		9.274	5.497		11.013	7.735

We did not manage to achieve a considerable improvement in describing the Stark structure in  $\text{Cs}_2\text{NaYCl}_6:\text{Eu}^{3+}$  and  $\text{Cs}_2\text{NaEuCl}_6$  crystal systems with the help of Hamiltonians (1)–(3), therefore calculations were carried out in the approximation of anomalously strong configuration interaction with the help of Hamiltonian (4). The application of Hamiltonian (4) has allowed to improve considerably the description of the Stark structure of  $\text{Eu}^{3+}$  ion. The root-mean-square deviation is  $5.5 \text{ cm}^{-1}$  for  $\text{Cs}_2\text{NaYCl}_6:\text{Eu}^{3+}$  and  $7.7 \text{ cm}^{-1}$  for  $\text{Cs}_2\text{NaEuCl}_6$ . This means the improvement of the description by 41% and 30%, respectively, in comparison with the approximation of weak configuration interaction.

In Table II the varied parameters obtained as the result of the description of the Stark structure in the approximation of anomalously strong configuration interaction are given. Crystal field parameters  $B_0^4$  and  $B_0^6$ , obtained in the approximation of anomalously strong configuration interaction, slightly differ from the parameters obtained with the help of Hamiltonian (1). It demonstrates that the new operator forms of Hamiltonian (4) describe the effects that are not reflected in Hamiltonian (1).

The optimal values of energies  $\Delta_{c1}$  and  $\Delta_{c2}$  are in good compliance with the relevant energies obtained in describing the intensities of the absorption bands of  $\text{Eu}^{3+}$  ion [31, 32].

The covalence parameters are also obtained during the description of the Stark structure of multiplets, these parameters are usually calculated during painstaking experiments (Table II).

TABLE II

Parameters of the Hamiltonian of the crystal field (4), calculated in the approximation of anomalously strong configuration interaction.

	$\text{Cs}_2\text{NaYCl}_6:\text{Eu}^{3+}$		$\text{Cs}_2\text{NaEuCl}_6$	
		[%]		[%]
$B_0^4$ [ $\text{cm}^{-1}$ ]	1771.5±12.7	0.715	1838.1±18.2	0.992
$B_0^6$ [ $\text{cm}^{-1}$ ]	314.1±16.0	5.088	332.4±18.6	5.604
$\gamma_{\sigma f} \times 10^4$	-370.5±0.2	0.057	-360.9±0.4	0.110
$\gamma_{\pi f} \times 10^4$	427.7±0.4	0.083	489.0±0.2	0.032
$\Delta_{c1}$ [ $\text{cm}^{-1}$ ]	17274.1±0.5	0.003	17173.6±0.1	0.001
$\Delta_{c2}$ [ $\text{cm}^{-1}$ ]	27378.3±0.2	0.001	26875.3±0.4	0.002

For  $\text{Cs}_2\text{NaYCl}_6:\text{Eu}^{3+}$  and  $\text{Cs}_2\text{NaEuCl}_6$  crystal systems we calculated the errors of the parameters of the crystal field Hamiltonian (4), obtained in the approximation of anomalously strong configuration interaction (Table II), on the basis of the above mentioned procedure (7)–(13). The small relative errors of the Hamiltonian parameters (4) demonstrate the high, almost resonant, sensitivity of the energy of the Stark components to the variation of these parameters. If to take into account that  $\sigma_{\text{exp}}$  contains many local minima in relation to the varied parameters, then the search for the optimum values of the parameters  $B_0^4$ ,  $B_0^6$ ,  $\gamma_{\sigma f}$ ,  $\gamma_{\pi f}$ ,  $\Delta_{c1}$ ,  $\Delta_{c2}$  is a difficult problem even for modern computers.

#### 4. Conclusions

The description of the Stark structure of  $\text{Eu}^{3+}$  ion in  $\text{Cs}_2\text{NaYCl}_6:\text{Eu}^{3+}$  and  $\text{Cs}_2\text{NaEuCl}_6$  crystal systems is carried out in the approximation of weak and anomalously strong configuration interaction. If the consideration of the configuration interaction is more correct, the root-mean-square deviation essentially decreases and there is a possibility to define covalence parameters from optical spectra. It essentially broadens the possibilities of optical spectroscopy in the determination of the electronic structure of optical centers.

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