Definition of covalence parameters in trigonal systems on optical spectrum

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Abstract—For the Pr\(^{3+}\) doped trigonal systems it is shown, that the application of a new crystal field Hamiltonian allows not only essentially to improve description of Stark multiplet splitting, but also to define the covalence parameters and odd symmetry crystal field parameters on optical spectrum. In the new Hamiltonian the influence of excited configurations with a charge transfer and opposite parity configurations is in more details taken into account.

Single crystals of YAl\(_3\)(BO\(_3\))\(_4\) doped with Pr\(^{3+}\) ions are comprehensively explored experimentally as perspective laser crystals. They have high hardness, non-deliquescence, high UV transparency, and absence of colour centres. 28 experimental energy levels from 91 possible can be found in [1]. The description of an energy spectrum in the one-configuration approximation which has been carried out in this paper, has shown, that the calculated multiplet splittings are badly agreed with experimental one. About poor adequacy of an one-configuration approximation for description of Stark structure of multiplets in trigonal systems was reported also in work [2]. It is possible to explain by strong influence of excited configurations, which is taken into account in an one-configuration approximation not full enough.

In this paper for account of influence of excited configurations we have applied the crystal field Hamiltonian offered in [3]

\[ H_{cf} = E_{\gamma J}^{\gamma J'} B^\gamma J + \sum_{\gamma} \sum_{J} \left( B^\gamma J \left( \frac{\Delta d}{\Delta_c} \right) \frac{G^\gamma J}{\Delta_c} \right) \delta_{\gamma J,\gamma J'} C^\gamma J \]  

Here \( B^\gamma J \) are the ordinary crystal field parameters, \( \Delta d \) is energy of an excited configuration of opposite parity \( 4f^{3-1}5d \); \( \Delta_c \) is energy of an excited configuration with charge transfer, \( G^\gamma J \) are the parameters related to the interconfiguration interaction, \( E_{\gamma J} \) and \( E_{\gamma J'} \) are the energies of \( \gamma J \) and \( \gamma J' \) multiplets.

The application of Hamiltonian (1) allows to reduce a root-mean-square deviation by 50% than in an one-configurational approximation. Besides \( B^\gamma J \) (Total) parameters have simple dependence on \( E_{\gamma J} \) and \( E_{\gamma J'} \) energy. This circumstance allows enough to uniquely determinate parameters \( B^\gamma J \), odd crystal field parameters \( B^\gamma J (d) \) and covalence parameters \( \gamma_{\alpha f}, \gamma'_{\alpha f} \).