SITE-SELECTIVE TIME RESOLVED LASER SPECTROSCOPY AND DFT STUDY OF Nd^{3+} OPTICAL CENTERS IN BaF_2 Nd^{3+} DOPED CRYSTALS

E. E. Vinogradova, E. A. Vagapova, L.Dolgov, A. Pištšev, and Yu.V. Orlovskii

Institute of Physics, University of Tartu, W. Ostwaldi Street 1, Tartu 50411, Estonia

Presenting author: elena.vinogradova@ut.ee

Optical centers of Nd^{3+} ions formed in doped fluorite-type hosts (CaF_2, SrF_2, and BaF_2) as a result of the heterovalent substitution of Me^{2+} ions (Ca^{2+}, Sr^{2+}, and Ba^{2+}) by Nd^{3+} ions are of considerable interest for study for various application in modern optics and photonics [1-2]. While the Nd^{3+} optical centers of Nd^{3+} in CaF_2 and SrF_2 are well studied [3-4], there are practically no experimental data on the Nd^{3+} optical centers formed in the BaF_2 host.

To study the Nd^{3+} optical centers in a bulk Nd^{3+}: BaF_2 crystal at low temperature (T = 6.5K), we used the methods of site-selective time-resolved laser spectroscopy with high spectral resolution. Also, to determine the lifetimes of the $^4F_{3/2}(1)$ Stark level of the optical centers, we measured the fluorescence decay kinetics using gated photon counting.

As a result, four types of Nd^{3+} optical centers in a bulk BaF_2 crystal were found. One of them was previously described in the literature, and the other three were discovered for the first time. The known type of optical centers has a symmetry close to cubic and a long radiative lifetime of the $^4F_{3/2}(1)$ state.

The spectroscopic experimental data and DFT analysis showed the formation of three different configurations of the tetragonal single L center of Nd^{3+} in the BaF_2 host, in contrast to the CaF_2 host, where only one configuration of the L center is observed. These three tetragonal optical centers are characterized by lowering their point symmetry from C_{4v} to C_s and differ in the noticeable displacement of the interstitial charge compensator F^- along different crystal axes, either a, or b, or c in the local coordinates of the optical center. This shifts the energy of the $^4F_{3/2}(1)$ Stark level of Nd^{3+} ion differently and makes the radiative lifetimes of these three optical sites different.

References

[1] J.M. Baker, Rare-earth and other impurity ions in non-cubic sites, in: W. Hayes (Ed.) Crystals with the fluorite structure, Clarendon Press, London, 1974, pp. 341-414.
[2] Y.V. Orlovskii, E.A. Vagapova, V. Peet, E. Vinogradova, L. Dolgov, V. Boltrushko, V. Hizhnyakov, One- and two-exciton states of pair centers of Kramers Nd^{3+} ions in Nd-doped CaF_2 and SrF_2 crystals, and their possible use as qubits, Journal of Luminescence, 251 (2022) 119218.
[3] J. Corish, C.R.A. Catlow, P.W.M. Jacobs, S.H. Ong, Defect aggregation in anion-excess fluorites. Dopant monomers and dimers, Physical Review B, 25 (1982) 6425-6438.
[4] Y.V. Orlovskii, V.V. Fedorov, T.T. Basiev, M. Altwein, B. Leu, J. Heber, S. Mirov, Nonradiative relaxation and inhomogeneous splitting of aggregated optical centers in the Nd^{3+}-doped CaF_2 and SrF_2 crystals (FLN and decay study), Journal of Luminescence, 83-84 (1999) 361-366.

Euroopa Liit
Euroopa
Regionaalarengu Fond

Eesti
tuleviku heaks