Stark splitting effects for Er^{3+} in Er_2O_3

P. Rynkun[1], G. Gaigalas[1,2], D. Kato[2,3], L. Radžiūtė[1], and P. Jönsson[4]

[1]*Institute of Theoretical Physics and Astronomy, Vilnius University, Vilnius, Lithuania*
[2]*National Institute for Fusion Science, Oroshi-cho, Toki, Japan*
[3]*Department Fusion Science, SOKENDAI (The Graduate University for Advanced Studies), Oroshi-cho, Toki, Japan*
[4]*Group for Materials Science and Applied Mathematics, Malmö University, Malmö, Sweden*

Presenting Author: pavel.rynkun@tfai.vu.lt

In the present work a fully *ab-initio* method was used to calculate Stark splitting effects of Er^{3+} in Er_2O_3. Crystal field effects were included by treating external ions as point charges at fixed positions. Charges and positions of the external ions are parameters (which depend on the compound) in the calculations. The coordinates of the external ions were obtained by molecular statics calculations based on density functional theories performed with the VASP code [1, 2]. Using first order perturbation theory, splitting of the degenerate atomic energy levels in the crystal electric field (Stark effect) can be calculated. To perform such calculations the GRASP2K [3] relativistic atomic structure program was extended to include programs for calculating the crystal field operator matrix elements and diagonalizing the full atomic Hamiltonian matrix [4].

Stark splitting of the ground state $4f^{11}\ ^4I^o_{15/2}$ and the excited state $4f^{11}\ ^4F^o_{9/2}$ of Er^{3+} in the Er_2O_3 crystal were computed from relativistic configuration interaction wave functions including Breit and QED effects. Different strategies to include electron correlation effects were tested. Also the effects of different number of neighbor ions (6 ions or 2 159 ions) and the influence of J-mixing were evaluated. The results are summarized in Table 1. It is clear that number of neighbor ions has a big impact and changes results dramatically. J-mixing influence to the Stark splitting is small. To calculate the transitions of Stark levels between states of Er^{3+} different strategies to include crystal field should be used.

MR S		DHF SD		DHF+5d		DHF+5d + 5f + 5g		Exp.
6 ions	2 159 ions	6 ions	2 159 ions	6 ions	2 159 ions	6 ions	2 159 ions	[5]
Stark levels of ground state $[Xe]4f^{11}\ ^4I^o_{15/2}$								
0	0	0	0	0	0	0	0	0
31.27	28.33	30.01	26.01	31.44	28.28	34.94	31.66	38
84.18	73.71	79.65	69.28	84.38	73.59	93.95	81.37	75
145.14	117.88	134.66	108.10	145.53	117.89	163.87	132.49	88
214.52	158.34	189.92	143.22	215.99	159.30	248.19	177.06	159
330.37	166.00	290.98	153.56	331.76	166.60	381.11	191.40	265
498.38	308.03	448.09	285.23	499.91	308.16	567.84	344.24	490
578.06	325.09	513.81	299.64	580.46	325.80	664.05	365.82	505
Stark levels of excited state $[Xe]4f^{11}\ ^4F^o_{9/2}$								
0	0	0	0	0	0	0	0	0
271.00	141.24	219.22	118.56	243.16	129.78	265.95	142.09	74
445.70	206.41	356.06	169.19	397.88	186.22	435.26	203.66	149
532.38	229.42	416.64	184.94	468.70	205.10	511.73	224.24	218
689.10	322.47	551.42	265.25	616.67	293.88	674.52	322.12	306

Table 1: *Comparison of computed energy (Stark) levels (in cm^{-1}) of Er^{3+} in Er_2O_3 without J-mixing induced by the crystal field. MR S: configuration state functions (CSFs) generated by single excitations from all core shells in to 5d shell. DHF SD: CSFs generated by single double (SD) excitations from all shells in to 4f shell. DHF+5d: CSFs generated by selected SD excitations from all shells in to 4f and 5d shells. DHF+5d + 5f + 5g: CSFs generated by selected SD excitations from all shells in to 4f, 5d, 5f, and 5g shells.*

References

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