Spectroscopic Characterisation of Rare-Earth Impurities in Crystals

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A fully numerical scheme has been developed for the computation of the energy levels and wave functions of rare-earth impurities in a crystalline environment. Ligand fields due to impurities in sites of cubic, trigonal, tetragonal, monoclinic and orthorhombic symmetry in the diamond crystal lattice structure were considered. In actual calculations the crystal-field coefficients could be varied continuously, covering fields of arbitrary symmetry. By this feature, the numerical method distinguishes itself over the previously exercised analytical analyses, which are restricted to the specific isolated cases of fields of pure cubic or pure axial symmetry.

With the calculated energies and wave functions the positions of lines in optical spectra were analysed. Splitting of levels in a magnetic field, with lifting of all degeneracy, is quantitatively analysed by adding the Zeeman energy. Magnetic fields can be of arbitrary strength covering both the low-field regime with constant g tensors and the non-linear behaviour at high fields.

Application of the method will be demonstrated in detail taking the example of the Er^{3+} ion with $4f^{11}$ electron configuration. Crystal field effects, experimentally observed as structure in the photoluminescence of the transition from excited state $^4I_{13/2}$ to spin–orbit ground state $^4I_{15/2}$, were analysed. Specifically for the spectrum known as Si-Er-1, the crystal-field parameters were determined and the symmetry of the centre was concluded to be orthorhombic-I. With an additional magnetic field, g tensors describing the Zeeman effect were calculated and compared with the around 70 electron paramagnetic resonance spectra published previously. An empirical rule of constant trace is supported, for both Γ_6 and Γ_7 states, in arbitrary cubic fields with trigonal or tetragonal axial distortion. In several cases a convincing agreement is obtained with internal consistency between optical and magnetic resonance data.

In summary, a versatile tool is developed for the analysis of the available vast amount of data from optical spectroscopy (luminescence, magneto-optics, optical detection of magnetic resonance) and electron spin resonance in ground and excited states of rare-earth impurities in crystalline hosts.