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High precision wavefunctions for hyperfine states of low symmetry materials suitable for quantum information processing

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ABSTRACT

We report on a method of employing spin Hamiltonian data to enhance the accuracy of crystal-field fits for rare-earth impurity sites of low symmetry in crystalline hosts. As an initial test of this method, we apply it to orthorhombic sites in Li^+ and Na^+ ion charge compensated Sm^{3+} centers in codoped CaF_2 and SrF_2 crystals. This yields a good agreement between theory and experiment, in addition to accurately modelling crystal strains due to the differing ionic radii of Li^+ and Na^+ .

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1. Introduction

There are two formalisms commonly employed to parameterize experimental energy levels in rare-earth spectroscopy. The first approach consists of determining the effective Hamiltonian for the $4f^N$ configuration, often referred to as a crystal-field Hamiltonian, which contains terms for the free-ion, the crystal-field, the Zeeman, the hyperfine, and the nuclear quadrupole interactions. By diagonalizing this Hamiltonian the entire energy level spectrum of a system can be characterized. However, for host crystals of very low symmetry it is difficult to achieve a crystal-field fit that accurately reproduces Zeeman and/or hyperfine splittings [1,2].

The second formalism consists of defining an effective Hamiltonian, referred to as a spin Hamiltonian, in a basis spanned by spin and nuclear angular momentum eigenstates. By substantially reducing the number of parameters to be fit, and additionally by allowing for an arbitrary orientation between the principal axes of interaction terms, this formalism can be used to achieve an accurate parameterization of materials having point group symmetries as low as C_1 for the dopant ion. However, the relationship between spin

Hamiltonians of different crystal-field levels must be determined by experiment, which diminishes the predictive power of this method.

This has resulted in the crystal-field Hamiltonian being primarily employed for modeling dopants in hosts that have a high point group symmetry. Spin Hamiltonians in contrast are often used for describing low site symmetry systems. Here we propose a method of employing the information encoded in spin Hamiltonians to produce accurate crystal-field fits for dopants at impurity sites of low symmetry.

A number of low site symmetry hosts have seen widespread application in optical quantum memory and quantum repeater experiments [3–8]. Specifically, their exceptionally narrow homogeneous linewidths, and correspondingly long coherence times, have made host materials such as Y_2SiO_5 and YAlO_3 the primary choice for optical quantum device experiments [9,10]. However, their low symmetry has to date rendered them impervious to a complete crystal-field analysis. Towards the resolution of this limitation, we present a method that aims to accurately fit the crystal-field parameters for low point group symmetry sites.

To test the efficacy of this method we apply it to Li^+ and Na^+ ion charge compensated Sm^{3+} orthorhombic [11,12] centers in codoped CaF_2 and SrF_2 crystals. The many crystal-field levels of the low lying ^6H and ^6F terms [13], and since C_{2v} point group symmetry ensures that the spin-Hamiltonian principal \hat{z} -axis is coincident with the corresponding crystal-field axis, make these materials well suited for the investigations presented here.

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2. Theoretical description

In this section we outline both the crystal-field and spin Hamiltonian formalisms before describing the method used to enhance our crystal-field fit using spin Hamiltonian data.

The effective Hamiltonian describing the energy levels of rare-earth ions has the form

$$H = H_{\text{FI}} + H_{\text{CF}} + H_Z + H_{\text{HF}}, \quad (1)$$

where H_{FI} corresponds to the free-ion term, H_{CF} is the crystal-field Hamiltonian, H_Z is the Zeeman interaction, and H_{HF} is the hyperfine interaction.

We start with a crystal field Hamiltonian for a cubic center, which has the following form:

$$\begin{aligned} H_{\text{cubic}} = & -\frac{1}{4}B_4^4 \left[C_0^{(4)} - \sqrt{10}(C_2^{(4)} + C_{-2}^{(4)}) \right. \\ & - \frac{3}{7}\sqrt{\frac{35}{2}}(C_4^{(4)} + C_{-4}^{(4)}) \left. \right] - \frac{13}{8}B_6^6 \left[C_0^{(6)} \right. \\ & + \frac{\sqrt{105}}{26}(C_2^{(6)} + C_{-2}^{(6)}) - \frac{5}{13}\sqrt{\frac{7}{2}}(C_4^{(6)} + C_{-4}^{(6)}) \\ & \left. + \frac{\sqrt{231}}{26}(C_6^{(6)} + C_{-6}^{(6)}) \right]. \quad (2) \end{aligned}$$

The B_q^k are the crystal field parameters for cubic symmetry sites, expressed in Wybourne's normalization [14], and $C_q^{(k)}$ are spherical tensors, the matrix elements of which are evaluated in the f^N configuration.

To model the C_{2v} symmetry sites we add the following terms to the crystal field Hamiltonian in order to account for a distortion from cubic symmetry

$$\begin{aligned} H_{\text{CF}} = & H_{\text{cubic}} + B_0^2 C_0^{(2)} + B_2^2 (C_2^{(2)} + C_{-2}^{(2)}) + B_0^4 C_0^{(4)} \\ & + B_2^4 (C_2^{(4)} + C_{-2}^{(4)}), \quad (3) \end{aligned}$$

with B_q^k being the crystal-field parameters. The \hat{z} -axis in Eqs. (2) and (3) is chosen such that it intersects the Sm^{3+} ion, passes through the edge formed by the F^- ion cube, and then intersects the Na^+/Li^+ site. The Zeeman term H_Z describes the effect that an external magnetic field, \mathbf{H} , has on rare-earth systems. The final term in Eq. (1), H_{HF} , describes the coupling of nuclear spin with the spin of the $4f^N$ electrons.

Having defined the complete Hamiltonian, the energy level spectrum can be calculated by diagonalizing H in the $|l_S l_M J M_J M_I\rangle$ basis. Here the state label γ corresponds to all additional degrees of freedom of the desired electronic configuration.

The spin Hamiltonian approach simplifies the treatment of the Zeeman and hyperfine interactions by working in a basis in which the free-ion and crystal-field terms of H are diagonal. This allows for the Zeeman and hyperfine interactions to be parameterized by effective spin half states and nuclear angular momentum states. The most general spin Hamiltonian has the form [15,16]

$$H_{\text{SH}} = \mu_B \mathbf{H} \cdot \mathbf{g} \cdot \mathbf{S} + \mathbf{I} \cdot \mathbf{A} \cdot \mathbf{S} + \mathcal{H}_Q + \mathcal{H}_Z, \quad (4)$$

where μ_B is the Bohr magneton, \mathbf{H} is the applied magnetic field, and \mathbf{g} is the g -value matrix. The term containing \mathbf{A} , in addition to the terms \mathcal{H}_Q and \mathcal{H}_Z , are responsible for the hyperfine, the nuclear quadrupole, and the nuclear Zeeman interactions, respectively. \mathbf{S} denotes the total spin angular momentum, and \mathbf{I} is the total nuclear angular momentum. For further details on spin Hamiltonians, the reader is referred to reference [15].

In order to fit the parameters of Eq. (1) to both energy level and spin Hamiltonian data we must calculate the theoretical spin Hamiltonian for a given set of free-ion and crystal-field

parameters. This is achieved by employing the well established method of effective operators [17,18].

The spin Hamiltonian for rare-earth doped crystals is an effective Hamiltonian in a basis spanned by spin half eigenstates. In order for H_{SH} to have the same eigenvalues as the complete Hamiltonian, we apply the transformation that diagonalizes $H_{\text{FI}} + H_{\text{CF}}$ to the operators of interactions described by H_{SH} . This readily allows for the principal components of spin Hamiltonian parameter tensors to be calculated from a given set of crystal-field parameters.

3. Experimental techniques

CaF_2 or SrF_2 crystals doped with either LiF or NaF, and codoped with SmF_3 , were grown in an inert (argon gas) atmosphere by the vertical Bridgman–Stockbarger technique using an Arthur D. Little 38 kW radio-frequency furnace. Laue X-ray diffraction was used to produce [100] oriented samples of around 5 mm³ in size.

Laser excitation and fluorescence spectra were obtained using a 5 W Coherent Innova 70 argon ion laser to optically pump a Spectra Physics 375B dye laser. Rhodamine 560 dye dissolved in 100% ethyl-glycol was used for excitation of the Sm^{3+} transitions. The samples were cooled to 10 K using a CTI-Cryogenics model 22C cold cycle refrigerator unit. Fluorescence was dispersed using a Spex Industries 500 M single monochromator. For fluorescence wavelengths in the range 18,000 cm⁻¹ to 12,000 cm⁻¹, a Hamamatsu R9249 photomultiplier which was thermoelectrically cooled to -25 °C, was used to detect the light. For near infrared light, a liquid nitrogen cooled germanium detector was used with phase sensitive detection provided by an Ortholoc model 9502 lock-in amplifier.

4. Results and discussion

The $4f^5$ configuration is appropriate for trivalent samarium which consists of 1001 two-fold degenerate electronic states. Of the available multiplets absorbing in the visible region, fluorescence is observed only from $^4G_{5/2}$ the barycenter of which is near 18,000 cm⁻¹. The ground multiplet is $^6H_{5/2}$ and fluorescence to all multiplets of the 6H_J and 6F_J terms could be observed with the exception of $^6F_{11/2}$.

The standard notation of a letter plus a numerical subscript is used here for labeling the crystal-field levels of the various LSJ multiplets. The ground multiplet is labeled Z, with the ground state being Z₁, and the first excited multiplet $^6H_{7/2}$ labeled by Y. The levels of the $^4G_{5/2}$ multiplet are labeled A₁, A₂ and A₃. The levels of the close lying $^6F_{1/2}$, $^6F_{3/2}$ and $^6H_{13/2}$ multiplets are heavily mixed through crystal-field J -mixing and hence are given a single label of S, following the conventions adopted in our previous work [13,19,20].

As the $\text{Sm}^{3+} \ ^6H_{5/2} \rightarrow \ ^4G_{5/2}$ transitions are nominally spin forbidden and because $\text{CaF}_2:\text{RE}^{3+}$ and $\text{SrF}_2:\text{RE}^{3+}$ are multi-site crystals, site selective laser excitation is the appropriate technique for studying the optical energy level structure of individual rare-earth ion centers. Two crystals of $\text{CaF}_2:0.075\%\text{Sm}^{3+}:1.09\%\text{Na}^+$ and $\text{CaF}_2:0.026\%\text{Sm}^{3+}:0.99\%\text{Li}^+$, as well as two crystals of $\text{SrF}_2:0.01\%\text{Sm}^{3+}:0.86\%\text{Na}^+$ and $\text{SrF}_2:0.037\%\text{Sm}^{3+}:1.17\%\text{Li}^+$ were chosen as optimally doped, from a selection of crystals grown. These Sm^{3+} concentrations are in a range appropriate to obtain predominantly single ion centers in the absence of codoping with either Li^+ or Na^+ [13], although in CaF_2 crystals preferentially formed clusters will be present at low levels.

Fig. 1 shows the broadband and site selective excitation spectra of a representative sample of $\text{CaF}_2:0.075\%\text{Sm}^{3+}:1.09\%\text{Na}^+$ as measured at 10 K. Only two centers are present, observed at roughly equal intensities. These are the cubic center studied in detail previously

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