Study of Higher-order Terms in Spin Hamiltonian

*We have examined theoretically the role of the higher-order terms in spin Hamiltonians for transition ions (e.g. Cr, Ni, Fe) in various magnetic systems and overviewed experimental studies. The high magnetic fields (**B**) up to 100 T make the non-linear field-dependent terms: **B^2S^2, B^3S, B^5S** & **B^2I^2, B^3I, B^5I**, important.*

This study aims at providing theoretical and computational support for researchers working in the area of magnetic, optoelectronic, and laser materials. Two novel methods of characterization of such materials are the high-magnetic field (**B**) measurements (HMM) of magnetic properties as well as the high-magnetic field & high-frequency EMR (HMF-EMR) measurements of spectroscopic properties [1]. The HMF-EMR techniques offer several advantages over the conventional methods, e.g. unprecedented spectral resolution and detection of new phenomena and spectral features, not detectable in X- or Q-band EMR [1-5]. By the higher-order field-dependent (HOFD) terms in the generalized spin Hamiltonians (GSHs) for transition ions we mean the higher-rank terms in the electronic (**S**) and nuclear spin (**I**), i.e. the terms non-linear in **B** of the type**: B^2S^2** [5], **B^3S** [4] (see, Fig. 1), **B^5S** or **B^2I^2**, **B^3I**, **B^5I**. The high pulsed fields, up to 100 T achievable at present [2], make these terms significant in comparison with the linear Zeeman electronic (Ze) term **B.g.S**, even if the associated parameters are small. Till recently these terms were mostly omitted from experimental considerations. We have carried out feasibility study on the role of the HOFD terms in HMM and HMF-EMR experiments [6].

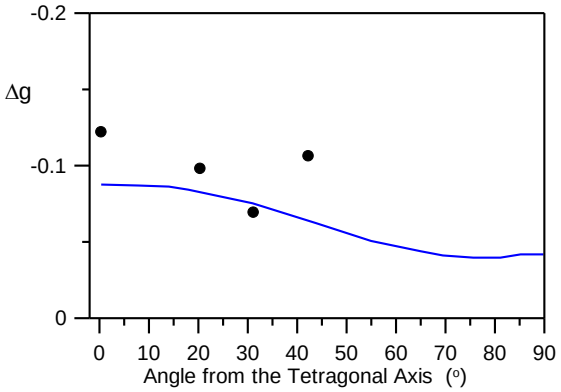

Fig. 1. First experimental evidence of the B^3S term in GSH observed in the g shift measured at 119 μm – adapted from [4].

The key issues involved in the SH approaches (see, Fig. 2) are: analysis of the mechanisms contributing to the HOFD parameters using microscopic SH (MSH); theoretical prediction of forms of various HOFD terms in GSH using the constructional SH approach; the role of the HOFD terms in the low- and high-field EMR; development of the computer simulation and fitting programs incorporating the HOFD terms; interpretation of the HMF-EMR spectra for the transition ions including HOFD parameters. Potential applications concern, e.g., Fe^{2+}:$CdPS_3$, Fe^{2+}/Fe^{3+}:$LiNbO_3$, GaAs:In:Cr, Mn_{12}-acetate, and Cr^{2+}/Cr^{3+}:forsterite. The preliminary research at IMR has provided a blueprint for future theoretical and experimental studies of the HOFD terms aimed at solving the pertinent theoretical and computational problems. The large scale project would have (i) direct applications for improving spectroscopic characterization of technologically important materials, and (ii) prime scientific importance for advancement of our knowledge. Collaboration of researchers from Japan and Poland is envisaged.

Constructional SH approach

Point symmetry group ⇒ **Group Theory** ⇒ ***Invariant*** combinations of the **spin** (**S**) & other operators (**B**, **I**):

⇒ ***Generalized SH (GSH)*** ∝ *(ZFS, Ze, HOFD terms)*

♦ Forms of SH predicted, including zero-field splitting (ZFS), Zeeman electronic (Ze), higher-order field-dependent (HOFD) terms, ***but*** no information on the parameter values

Derivational SH approach

$H_{physical}$ (FI+CF) ⇒ Perturbation Theory (**PT**) ⇒ *effective* SH:

⇒ ***Microscopic SH (MSH):*** ∝ *(physical parameters)*

♦ Forms of SH **and** values of ZFS & Ze parameters predicted

Fig. 2. Conceptual framework underlying the two SH approaches (for detailed explanations and references, see [6].

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Key Words

Higher-Order Field-dependent Terms, Generalized Spin Hamiltonians, Electron Magnetic Resonance

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