

High speed algorithm for the calculation of magnetic and orbital excitations in rare earth based systems

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Abstract

An algorithm for the efficient and fast calculation of dispersive magnetic excitations in rare earth based systems has been developed. Crystal field anisotropy and anisotropy of the two ion interactions can be taken into account. It is based on the standard mean field–random phase approximation of the problem. In analogy to lattice dynamical calculations the central problem of finding the poles of the dynamical susceptibility $\chi(\mathbf{Q}, \omega)$ has been reduced to the diagonalisation of a dynamical matrix. This approach is applicable to complex magnetic structures, it is fast and easily generalised to include many single ion transitions and orbital interactions.

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1. Introduction

Many special models have been developed for the calculation of dispersive excitations in rare earth based systems (see [1] for an overview). An example is the spin wave model in ferromagnets, which is valid for negligible single ion anisotropy. For systems with significant crystal field anisotropy, pseudo spin models have been used, such as in the case of the cuprates [2]. Recently systems with large multipolar interactions have been investigated and isospin models have been developed to interpret the dispersive modes, such as in the case of CeB₆ [3]. All these special models are based on or equivalent to the mean field–random phase approach to the problem. In this article a very general and fast method for the calculation of dispersive excitations in rare earth based systems is presented. The model is applicable to arbitrary complex magnetic or orbital structures. Single ion anisotropy, isotropic and

anisotropic two ion interactions can be treated according to the following magnetic Hamiltonian \mathcal{H}_{mag} .

$$\mathcal{H}_{\text{mag}} = \sum_s B_l^m O_l^m(\mathbf{J}^s) - \frac{1}{2} \sum_{ss'\alpha\beta} J_{\alpha\beta}^s \mathcal{J}_{\alpha\beta}(ss') J_{\beta}^{s'} - \sum_s g_{Js} \mu_B \mathbf{J}^s \mathbf{H} \quad (1)$$

The first term describes the crystal field (Stevens operators O_l^m , see the table in the Appendix) [4], the second the magnetic two ion interaction, the third the Zeeman energy describing the effect of the application of an external magnetic field. The method is easily generalised to the case of multipolar interactions by identifying the components ($\alpha = a, b, c, \dots$) of the angular momentum operator \mathbf{J} with the first order Stevens parameters according to the table in the Appendix.

2. The mean field–random phase approximation

In order to define the computational problem this section is devoted to the description of the standard mean

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field–random phase approximation, which is commonly used to calculate dispersive magnetic excitations in rare earth based systems. Neutron spectroscopy is the most important experimental method for the determination of these excitations. The following form of the double differential inelastic neutron cross section of magnetic excitations has been given in [1]¹

$$\begin{aligned} \frac{d\sigma}{d\Omega dE'} &= N \frac{k'}{k} \left(\frac{\hbar\gamma e^2}{mc^2} \right)^2 \sum_{\mathbf{Q}} \delta_{\vec{k}, \mathbf{Q}} \sum_{\alpha\beta ss'} (\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta) \\ &\times \left\{ \frac{1}{2} g_J F(\mathbf{Q}) \right\}_s \left\{ \frac{1}{2} g_J F(\mathbf{Q}) \right\}_{s'} \\ &\times e^{-W_s(\mathbf{Q}) - W_{s'}(\mathbf{Q})} \frac{S_{ss'}^{\alpha\beta}(\mathbf{Q}, \omega)}{2\pi\hbar N_b} \end{aligned} \quad (2)$$

In Eq. (2) N denotes the number of magnetic atoms in the sample, \mathbf{k} and \mathbf{k}' the wavevector of the incoming and scattered neutron, respectively. The total magnetic cross section is $4\pi \left(\frac{\hbar\gamma e^2}{mc^2} \right)^2 = 3.65$ barn. $\hbar\omega = E - E'$ and $\vec{k} = \mathbf{k} - \mathbf{k}'$ denote the energy and momentum transfer. $\left\{ \frac{1}{2} g_J F(\mathbf{Q}) \right\}_s$ and $W_s(\mathbf{Q})$ are the magnetic form factor and the Debye–Waller factor of the atom number s in the magnetic unit cell. $\alpha, \beta = a, b, c$ denote the spatial coordinates and $s, s' = 1, \dots, N_b$ number the different atoms in the magnetic unit cell.

According to the fluctuation dissipation theorem, the scattering function S and thus excitation energies and intensities can be calculated from the absorptive part of the ω and \mathbf{Q} dependent susceptibility $\chi_{\alpha\beta}^{ss'}(\mathbf{Q}, \omega)$.

$$\begin{aligned} S_{ss'}^{\alpha\beta}(\mathbf{Q}, \omega) &= \frac{2\hbar}{1 - e^{-\hbar\omega/kT}} \chi_{\alpha\beta}^{s's}(\mathbf{Q}, \omega) \\ \chi_{\alpha\beta}^{s's}(\mathbf{Q}, \omega) &= \frac{1}{2i} [\chi_{\alpha\beta}^{ss'}(\mathbf{Q}, \omega) - \chi_{\beta\alpha}^{s's}(\mathbf{Q}, \omega)] \end{aligned} \quad (3)$$

The central problem is therefore the calculation of the dynamical susceptibility χ . Within the standard model of rare earth magnetism the approach to this problem is to make use of the mean field–random phase approximation: the susceptibility can be calculated from the single ion susceptibility $\chi_{\alpha\beta}^{\text{single ion-}s}(\omega)$ and the Fourier transform of the (in general anisotropic) two-ion interaction $\mathcal{J}_{\alpha\beta}^{ss'}(\mathbf{Q})$ by solving the following equation.

$$\delta_{ss'} \delta_{\alpha\beta} = \sum_{s''\delta} [\delta_{ss''} [\chi^{\text{single ion-}s}(\omega)]_{\alpha\delta}^{-1} - \mathcal{J}_{\alpha\delta}^{ss''}(\mathbf{Q})] \chi_{\delta\beta}^{s''s'}(\mathbf{Q}, \omega) \quad (4)$$

The single ion susceptibility in this equation is given by

$$\begin{aligned} \chi_{\alpha\beta}^{\text{single ion-}s}(\omega) &= \sum_{ij} \frac{\langle i | J_\alpha^s - \langle J_\alpha^s \rangle_{\mathbf{H}_{\text{eff}}^s, T} | j \rangle \langle j | J_\beta^s - \langle J_\beta^s \rangle_{\mathbf{H}_{\text{eff}}^s, T} | i \rangle}{E_j^s - E_i^s - \hbar\omega} (n_i - n_j) \end{aligned} \quad (5)$$

and the Fourier transform of the two-ion interaction $\mathcal{J}_{\alpha\beta}$ is defined as

$$\mathcal{J}_{\alpha\beta}^{ss'}(\mathbf{Q}) = \sum_{\mathbf{G}'} \mathcal{J}_{\alpha\beta}(\mathbf{G} + \mathbf{r}_s - (\mathbf{G}' + \mathbf{r}_{s'})) e^{-i\mathbf{Q}(\mathbf{G} + \mathbf{r}_s - (\mathbf{G}' + \mathbf{r}_{s'}))} \quad (6)$$

Here the \mathbf{G} are the lattice vectors of the Bravais Lattice and \mathbf{r}_s is the position of the atom s in the magnetic unit cell, $|i\rangle$ and $|j\rangle$ denote eigenstates of the ion s as calculated selfconsistently within mean field theory. \mathbf{J}^s is the angular momentum operator, n_i and n_j denote the Boltzmann occupation numbers of the energy levels E_i^s and E_j^s of this ion, respectively. Brackets $\langle \dots \rangle_{\mathbf{H}_{\text{eff}}^s, T}$ refer to a thermal expectation value at the temperature T and effective magnetic field $\mathbf{H}_{\text{eff}}^s$, which is determined selfconsistently by solving the mean field equations

$$H_{\text{eff},\alpha}^s = H_\alpha + \sum_{\mathbf{G}'s'} \frac{\mathcal{J}_{\alpha\beta}(\mathbf{r}_s - (\mathbf{G}' + \mathbf{r}_{s'}))}{g_{Js} \mu_B} \langle J_\beta^{s'} \rangle_{\mathbf{H}_{\text{eff}}^s, T} \quad (7)$$

In order to evaluate Eqs. (2)–(6) without producing a numerical divergence it is necessary to add to $\hbar\omega$ a small imaginary constant $\hbar\omega \rightarrow \hbar\omega + i\epsilon$ and insert this into Eq. (5). The calculation of the dynamical susceptibility is done by solving Eq. (4) for a series of different values of ω . Thus for each value of ω a matrix of dimension $3N_b \times 3N_b$ has to be inverted. This has to be done for different values of the scattering vector \mathbf{Q} . In order to minimise the computational effort and enable the treatment of large magnetic unit cells a different computational algorithm was developed, which requires only the solution of one generalised eigenvalue problem at each scattering vector \mathbf{Q} . Thus this method is similar to the standard approach to lattice dynamics. It is described in the next section.

3. Algorithm for fast calculation of excitation energies and intensities

The algorithm is presented in the following way: only one single ion transition with the excitation energy $\Delta^s = E_+ - E_-$ is considered for each ion. For instance, in case of a Kramer ground state doublet, which is split in the magnetically ordered state by the mean field, this single ion excitation energy is the distance between the upper and lower state of the doublet $|\pm\rangle$. The general case of more transitions can be easily formulated: it is only necessary to redefine s to index every transition of interest in the different ions of the magnetic unit cell. The algorithm has been implemented in this more general form in the public domain program package *McPhase* [5]. The user can select those single ion transitions, which should be considered in the computation.

For such a single ion transition the single ion susceptibility reads (compare Eq. (5))

$$\chi_{\alpha\beta}^{\text{single ion-}s}(\omega) = \frac{M_{\alpha\beta}^s}{\Delta^s - \hbar\omega} + \frac{M_{\beta\alpha}^s}{\Delta^s + \hbar\omega} \quad (8)$$

¹ <http://ntserv.fys.ku.dk/jens/book/book.htm>.

with the transition elements

$$M_{\alpha\beta}^s \equiv \langle -|J_\alpha^s - \langle J_\alpha^s \rangle_{\mathbf{H}_{\text{eff},T}^s}|+\rangle + |J_\beta^s - \langle J_\beta^s \rangle_{\mathbf{H}_{\text{eff},T}^s}|-\rangle \rangle (n_- - n_+) \quad (9)$$

Note that the 3×3 matrices $M_{\alpha\beta}^s$ always have one double degenerate eigenvalue = zero ($\sigma = 2, 3$) and one positive eigenvalue ($\sigma = 1$), which we denote as $\gamma^s = (\langle -|J_a|+\rangle \langle +|J_a|- \rangle + \langle -|J_b|+\rangle \langle +|J_b|- \rangle + \langle -|J_c|+\rangle \langle +|J_c|- \rangle) (n_- - n_+)$. The problem may be simplified by using the unitary transformation $\mathcal{U}_{\alpha\beta}^s (\mathcal{U}_{\alpha\beta}^{s\dagger} \mathcal{U}^s = 1)$, which diagonalises the $M_{\alpha\beta}^s$. Note that the matrix $M_{\alpha\beta}^s$ and consequently also $\mathcal{U}_{\alpha\beta}^s$ depend on temperature and magnetic field (both, external and molecular field according to Eq. (7)). The temperature dependence is due to the Boltzmann occupation numbers n_i and, in addition, to the temperature variation of the molecular field leading to a temperature variation of the single ion eigenstates $|i\rangle$ and energies E_i^s .

Using the transformation matrix $\mathcal{U}_{\alpha\beta}^s$ the single ion susceptibility (8) may be written as

$$\chi_{\alpha\beta}^{\text{single ion-}s}(\omega) = \sum_{\sigma=1}^3 \left(\frac{\mathcal{U}_{\alpha\sigma}^s \gamma^s \delta_{\sigma 1} U_{\sigma\beta}^{s\dagger}}{\Delta^s - \hbar\omega} + \frac{\mathcal{U}_{\alpha\sigma}^{s*} (-1) \gamma^s \delta_{\sigma 1} U_{\sigma\beta}^{s\dagger}}{-\Delta^s - \hbar\omega} \right) \quad (10)$$

The first term in Eq. (10) corresponds to energy loss of the neutron and only this term is considered in the following (the other term can also be considered – it is just necessary to add another transition (index s) to the problem with negative Δ_s , negative γ^s and the complex conjugate Matrix U).

$$\chi_{\alpha\beta}^{\text{single ion-}s}(\omega) \sim \sum_{\sigma=1}^3 \frac{\mathcal{U}_{\alpha\sigma}^s \gamma^s \delta_{\sigma 1} U_{\sigma\beta}^{s\dagger}}{\Delta^s - \hbar\omega} \quad (11)$$

Using the expression (11) for the single ion susceptibility we multiply Eq. (4) with $\chi_{\alpha\beta}^{\text{single ion-}s}(\omega)$ and transform both sides of the resulting equation with the unitary transformation $\mathcal{U}^{s\dagger}[\dots]\mathcal{U}^s$. We get

$$\frac{\gamma^s \delta_{\sigma 1} \delta_{\sigma\sigma'} \delta_{ss'}}{\Delta^s - \hbar\omega} = \sum_{s''} \sum_{\delta=1}^3 \left[\delta_{ss''} \mathcal{U}_{\sigma\delta}^{s''\dagger} - \sum_{\alpha''=1}^3 \frac{\gamma^s \delta_{\sigma 1} \mathcal{U}_{\sigma\alpha''}^{s\dagger}}{\Delta^s - \hbar\omega} \mathcal{J}_{\alpha''\delta}^{ss''}(\mathbf{Q}) \right] \times \sum_{\beta=1}^3 \chi_{\delta\beta}^{s''s'}(\mathbf{Q}, \omega) \mathcal{U}_{\beta\sigma'}^s \quad (12)$$

Now the unitary transform of χ is introduced by

$$\Psi_{\sigma''\sigma'}^{s''s'}(\mathbf{Q}, \omega) \equiv \sum_{\delta\beta} \frac{\mathcal{U}_{\sigma''\delta}^{s''\dagger} \chi_{\delta\beta}^{s''s'}(\mathbf{Q}, \omega) \mathcal{U}_{\beta\sigma'}^s}{\sqrt{\gamma^{s''}} \sqrt{\gamma^{s'}}} \quad (13)$$

Transforming the exchange interaction \mathcal{J} in a similar way, i.e.

$$\mathcal{L}_{\sigma\sigma''}^{ss''}(\mathbf{Q}) \equiv \sum_{\delta\beta} \mathcal{U}_{\sigma\delta}^{s\dagger} \mathcal{J}_{\delta\beta}^{ss''}(\mathbf{Q}) \mathcal{U}_{\beta\sigma''}^s \quad (14)$$

Eq. (12) may be rewritten

$$\frac{\delta_{\sigma 1} \delta_{\sigma\sigma'} \delta_{ss'}}{\Delta^s - \hbar\omega} = \sum_{s''} \sum_{\sigma''=1}^3 \left[\delta_{ss''} \delta_{\sigma\sigma''} \frac{\sqrt{\gamma^{s''}}}{\sqrt{\gamma^{s'}}} - \frac{\delta_{\sigma 1}}{\Delta^s - \hbar\omega} \sqrt{\gamma^s} \mathcal{L}_{\sigma\sigma''}^{ss''}(\mathbf{Q}) \sqrt{\gamma^{s''}} \right] \times \Psi_{\sigma''\sigma'}^{s''s'}(\mathbf{Q}, \omega) \quad (15)$$

Note that in this expression (15) the ratio $\frac{\sqrt{\gamma^{s''}}}{\sqrt{\gamma^{s'}}$ is equal to +1 if γ^s is positive and –1 if γ^s is negative. We define therefore the matrix $A_{ss''} = \delta_{ss''} \frac{\sqrt{\gamma^{s''}}}{\sqrt{\gamma^{s'}}$. We show now that only the $\sigma''\sigma' = 11$ component of Ψ may be nonzero: First, consider Eq. (15) for $\sigma = 2, 3$. It follows directly, that $\Psi_{2\sigma'}^{s''s'}(\mathbf{Q}, \omega) = 0$ and $\Psi_{3\sigma'}^{s''s'}(\mathbf{Q}, \omega) = 0$. Second, we use this result and consider Eq. (15) for $\sigma' = 2, 3$:

$$0 = \sum_{s''} \left[A_{ss''} \delta_{\sigma 1} - \frac{\delta_{\sigma 1}}{\Delta^s - \hbar\omega} \sqrt{\gamma^s} \mathcal{L}_{\sigma 1}^{ss''}(\mathbf{Q}) \sqrt{\gamma^{s''}} \right] \Psi_{1\sigma'}^{s''s'}(\mathbf{Q}, \omega) \quad (16)$$

For $\sigma = 2, 3$ this equation gives no further information, but for $\sigma = 1$ it means, that $\Psi_{12}^{s''s'}(\mathbf{Q}, \omega)$ and $\Psi_{13}^{s''s'}(\mathbf{Q}, \omega)$ must be either zero or eigenvectors of the matrix $d_{ss''} = \frac{1}{\Delta^s - \hbar\omega} \sqrt{\gamma^s} \mathcal{L}_{11}^{ss''}(\mathbf{Q}) \sqrt{\gamma^{s''}}$. As the second case cannot be true for arbitrary ω and \mathbf{Q} , we may write Ψ as

$$\Psi_{\sigma''\sigma'}^{s''s'}(\mathbf{Q}, \omega) = \delta_{\sigma''\sigma'} \delta_{\sigma 1} \Psi_{\sigma''\sigma'}^{s''s'}(\mathbf{Q}, \omega) \quad (17)$$

Setting $\sigma = \sigma' = 1$ in Eq. (15) leads to the RPA equation from which $\Psi_{11}^{s''s'}(\mathbf{Q}, \omega)$ can be determined:

$$\delta_{ss''} = \sum_{s''} \left[A_{ss''} (\Delta^s - \hbar\omega) - \sqrt{\gamma^s} \mathcal{L}_{11}^{ss''}(\mathbf{Q}) \sqrt{\gamma^{s''}} \right] \Psi_{11}^{s''s'}(\mathbf{Q}, \omega) \quad (18)$$

Defining the hermitian matrix $A_{ss''} = A_{ss''} \Delta^s - \sqrt{\gamma^s} \mathcal{L}_{11}^{ss''}(\mathbf{Q}) \sqrt{\gamma^{s''}}$ the following generalised eigenvalue problem may be solved

$$\sum_{s'} A_{ss'} \mathbf{t}^{s'} = \hbar\omega \sum_{s'} A_{ss'} \mathbf{t}^{s'} \quad (19)$$

Note that in the form given above the matrix A is not positive definite, but A is positive definite, this has to be taken into account when using the standard algorithm for generalised eigenvalue problems. The solution of the eigenvalue problem (19) yields the eigenvectors $\mathcal{T} = (\mathbf{t}_1, \mathbf{t}_2, \dots, \mathbf{t}_r, \dots)$ and eigenvalues $\hbar\omega_r$ (which may be written as the eigenvalue matrix $\Omega_{rr'} = \delta_{rr'} \hbar\omega_r$). The eigenvalues $\hbar\omega_r$ represent the excitation energies of the system. Thus Eq. (19) can be viewed as the analogon to the diagonalisation of the dynamical matrix in the case of lattice dynamical problems.

The eigenvector matrix \mathcal{T} provides a unitary transformation of Eq. (18) (note the relations $\mathcal{T} \mathcal{T}^\dagger A = \mathcal{T}^\dagger A \mathcal{T} = 1$ and $A \mathcal{T} \Omega \mathcal{T}^\dagger A = A$)

$$\delta_{ss''} = \sum_{s''r'r's''s''} A_{ss''} \mathcal{T}_{s''r}(\hbar\omega_r - \hbar\omega) \delta_{rr'} \mathcal{T}_{r's''}^\dagger A_{s''s''} \Psi^{s''s'}(\mathbf{Q}, \omega) \quad (20)$$

This equation may be easily inverted to provide the following expression for $\Psi^{ss'}(\mathbf{Q}, \omega)$:

$$\Psi^{ss'}(\mathbf{Q}, \omega) = \sum_{r'r'} \mathcal{T}_{sr}(\hbar\omega_r - \hbar\omega)^{-1} \delta_{r'r'} \mathcal{T}_{r's}^\dagger \quad (21)$$

The standard procedure to avoid divergencies is to substitute $\hbar\omega$ with $\hbar\omega + i\epsilon$ and take the limit for $\epsilon \rightarrow 0^+$. Using Diracs formula

$$\lim_{\epsilon \rightarrow 0^+} \frac{1}{\hbar\omega_r - \hbar\omega - i\epsilon} = \mathcal{P} \frac{1}{\hbar\omega_r - \hbar\omega} + i\pi\delta(\hbar\omega_r - \hbar\omega) \quad (22)$$

we may calculate Ψ'' which is the unitary transform of χ'' (compare Eq. (13)):

$$\begin{aligned} \Psi_{\sigma''\sigma'}^{s's'}(\mathbf{Q}, \omega) &= \sum_{\beta\delta} \frac{\mathcal{U}_{\sigma''\delta}^{s'\dagger} \chi_{\delta\beta}^{s's'}(\mathbf{Q}, \omega) \mathcal{U}_{\beta\sigma'}^s}{\sqrt{\gamma^{s\star}} \sqrt{\gamma^{s'}}} \\ &= \frac{1}{2i} [\Psi^{ss'}(\mathbf{Q}, \omega) - \Psi^{\star s's}(\mathbf{Q}, \omega)] \delta_{\sigma''\sigma'} \delta_{\sigma''1} \end{aligned} \quad (23)$$

Inserting Eqs. (21) and (22) into (23) and transforming the result back using \mathcal{U}^s and $\mathcal{U}^{s'\dagger}$ yields the final result for the dynamical susceptibility:

$$\begin{aligned} \chi_{\alpha\beta}^{s's'}(\mathbf{Q}, \omega) &= \sqrt{\gamma^{s\star}} \sqrt{\gamma^{s'}} \pi \sum_r \mathcal{U}_{\alpha 1}^s \mathcal{T}_{sr}(\mathbf{Q}) \\ &\quad \times \delta(\hbar\omega_r(\mathbf{Q}) - \hbar\omega) \mathcal{T}_{rs}^\dagger(\mathbf{Q}) \mathcal{U}_{1\beta}^{s'\dagger} \end{aligned} \quad (24)$$

In order to get the neutron cross section this result has to be inserted in Eqs. (2) and (3).

The procedure for the calculation of excitation energies $\hbar\omega_r$ and neutron intensities outlined above is very fast, because it involves only a diagonalisation (determination of the matrix \mathcal{T}) for every scattering vector of interest. It is not necessary to calculate the energy dependence of the dynamical susceptibility saving a lot of computation time. The price to be paid for this advantage is that in this level of approximation it is not possible to discuss more complex response functions than given by Eq. (5). These would be necessary to describe for example a nonzero intrinsic line-width of the modes.

A very interesting fact is, that the procedure outlined in this section may easily be generalised to multipolar interactions just by letting the spacial indices (e.g. α) run not only from a to c but to the number of multipolar operators considered. Care must then be taken to take only the components $\alpha, \beta = a, b, c$ of $\chi_{\alpha\beta}^{s's'}(\mathbf{Q}, \omega)$ when inserting Eq. (24) into (2) and (3), because neutrons are only probing the matter by virtue of their spin moment.

4. Conclusion

A general and fast method for the calculation of dispersive excitations in rare earth based materials has been developed. Neutron spectra can easily be calculated in complex magnetic/orbital structures. Crystal field effects are fully included. The algorithm presented above has been imple-

mented and tested in the *McPhase* program package [5]² by comparison with the conventional type of analysis based on the computation of the full energy dependence of the dynamical susceptibility as described at the end of section 2.

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Appendix. Stevens operators

A complete list of operators up to $l = 6$ as introduced by Stevens [4,6] is given in the following table. In the algorithm developed in this work and used in the program package *McPhase* [5] these operators are identified with J_α ($\alpha = a, b, c, d, e, \dots$). This notation is convenient for the analysis of multipolar interactions.

$$X = J(J + 1)$$

$$O_{00} = 1$$

$$J_a = O_{11}^s = \frac{-i}{2} [J_+ - J_-] = J_y$$

$$J_b = O_{10} = J_z$$

$$J_c = O_{11} = \frac{1}{2} [J_+ + J_-] = J_x$$

$$J_d = O_{22}^s = \frac{-i}{2} [J_+^2 - J_-^2] = J_x J_y + J_y J_x = 2P_{xy}$$

$$J_e = O_{21}^s = \frac{-i}{4} [J_z(J_+ - J_-) + (J_+ - J_-)J_z] = \frac{1}{2} [J_y J_z + J_z J_y] = P_{yz}$$

$$J_f = O_{20} = [3J_z^2 - X]$$

$$J_g = O_{21} = \frac{1}{4} [J_z(J_+ + J_-) + (J_+ + J_-)J_z] = \frac{1}{2} [J_x J_z + J_z J_x] = P_{xz}$$

$$J_h = O_{22} = \frac{1}{2} [J_+^2 + J_-^2] = J_x^2 - J_y^2$$

$$J_i = O_{33}^s = \frac{-i}{2} [J_+^3 - J_-^3]$$

$$\dots = O_{32}^s = \frac{-i}{4} [(J_+^2 - J_-^2)J_z + J_z(J_+^2 - J_-^2)]$$

$$\begin{aligned} O_{31}^s &= \frac{-i}{4} [(J_+ - J_-)(5J_z^2 - X - 1/2) \\ &\quad + (J_+ - J_-)(5J_z^2 - X - 1/2)(J_+ - J_-)] \end{aligned}$$

$$O_{30} = [5J_z^3 - (3X - 1)J_z]$$

$$\begin{aligned} O_{31} &= \frac{1}{4} [(J_+ + J_-)(10J_z^2 - 2X - 1) \\ &\quad + (5J_z^2 - X - 1/2)(J_+ + J_-)] \end{aligned}$$

$$O_{32} = \frac{1}{4} [(J_+^2 + J_-^2)J_z + J_z(J_+^2 + J_-^2)]$$

$$O_{33} = \frac{1}{2} [J_+^3 + J_-^3]$$

² www.mcphase.de.

$$\begin{aligned}
O_{44}^s &= \frac{-i}{2} [(J_+^4 - J_-^4)] \\
O_{43}^s &= \frac{-i}{4} [(J_+^3 - J_-^3)J_z + J_z(J_+^3 - J_-^3)] \\
O_{42}^s &= \frac{-i}{4} [(J_+^2 - J_-^2)(7J_z^2 - X - 5) + (7J_z^2 - X - 5)(J_+^2 - J_-^2)] \\
O_{41}^s &= \frac{-i}{4} [(J_+ - J_-)(7J_z^3 - (3X + 1)J_z) + (7J_z^3 - (3X + 1)J_z)(J_+ - J_-)] \\
O_{40} &= [35J_z^4 - (30X - 25)J_z^2 + 3X^2 - 6X] \\
O_{41} &= \frac{1}{4} [(J_+ + J_-)(7J_z^3 - (3X + 1)J_z) \\
&\quad + (7J_z^3 - (3X + 1)J_z)(J_+ + J_-)] \\
O_{42} &= \frac{1}{4} [(J_+^2 + J_-^2)(7J_z^2 - X - 5) + (7J_z^2 - X - 5)(J_+^2 + J_-^2)] \\
O_{43} &= \frac{1}{4} [(J_+^3 + J_-^3)J_z + J_z(J_+^3 + J_-^3)] \\
O_{44} &= \frac{1}{2} [(J_+^4 + J_-^4)] \\
\\
O_{55}^s &= \frac{-i}{2} [J_+^5 - J_-^5] \\
O_{54}^s &= \frac{-i}{4} [(J_+^4 - J_-^4)J_z + J_z(J_+^4 - J_-^4)] \\
O_{53}^s &= \frac{-i}{4} [(J_+^3 - J_-^3)(9J_z^2 - X - 33/2) + (9J_z^2 - X - 33/2)(J_+^3 - J_-^3)] \\
O_{52}^s &= \frac{-i}{4} [(J_+^2 - J_-^2)(3J_z^3 - (X + 6)J_z) + (3J_z^3 - (X + 6)J_z)(J_+^2 - J_-^2)] \\
O_{51}^s &= \frac{-i}{4} [(J_+ - J_-)\{21J_z^4 - 14J_z^2X + X^2 - X + 3/2\} + \{\dots\}(J_+ - J_-)] \\
O_{50} &= [63J_z^5 - (70X - 105)J_z^3 + (15X^2 - 50X + 12)J_z] \\
O_{51} &= \frac{1}{4} [(J_+ + J_-)(21J_z^4 - 14J_z^2X + X^2 - X + 3/2) \\
&\quad + (21J_z^4 - 14J_z^2X + X^2 - X + 3/2)(J_+ + J_-)] \\
O_{52} &= \frac{1}{4} [(J_+^2 + J_-^2)(3J_z^3 - (X + 6)J_z) + (3J_z^3 - (X + 6)J_z)(J_+^2 + J_-^2)] \\
O_{53} &= \frac{1}{4} [(J_+^3 + J_-^3)(9J_z^2 - X - 33/2) + (9J_z^2 - X - 33/2)(J_+^3 + J_-^3)] \\
O_{54} &= \frac{1}{4} [(J_+^4 + J_-^4)J_z + J_z(J_+^4 + J_-^4)] \\
O_{55} &= \frac{1}{2} [J_+^5 + J_-^5] \\
\\
O_{66}^s &= \frac{-i}{2} [J_+^6 - J_-^6] \\
O_{65}^s &= \frac{-i}{4} [(J_+^5 - J_-^5)J_z + J_z(J_+^5 - J_-^5)] \\
O_{64}^s &= \frac{-i}{4} [(J_+^4 - J_-^4)(11J_z^2 - X - 38) + (11J_z^2 - X - 38)(J_+^4 - J_-^4)]
\end{aligned}$$

$$\begin{aligned}
O_{63}^s &= \frac{-i}{4} [(J_+^3 - J_-^3)(11J_z^3 - (3X + 59)J_z) \\
&\quad + (11J_z^3 - (3X + 59)J_z)(J_+^3 - J_-^3)] \\
O_{62}^s &= \frac{-i}{4} [(J_+^2 - J_-^2)\{33J_z^4 - (18X + 123)J_z^2 + X^2 + 10X + 102\} \\
&\quad + \{\dots\}J_+^2 - J_-^2] \\
O_{61}^s &= \frac{-i}{4} [(J_+ - J_-)\{33J_z^5 - (30X - 15)J_z^3 + (5X^2 - 10X + 12)J_z\} \\
&\quad + \{\dots\}(J_+ - J_-)] \\
O_{60} &= [231J_z^6 - (315X - 735)J_z^4 + (105X^2 - 525X + 294)J_z^2 - 5X^3 \\
&\quad + 40X^2 - 60X] \\
O_{61} &= \frac{1}{4} [(J_+ + J_-)\{33J_z^5 - (30X - 15)J_z^3 + (5X^2 - 10X + 12)J_z\} \\
&\quad + \{\dots\}(J_+ + J_-)] \\
O_{62} &= \frac{1}{4} [(J_+^2 + J_-^2)\{33J_z^4 - (18X + 123)J_z^2 + X^2 + 10X + 102\} \\
&\quad + \{\dots\}(J_+^2 + J_-^2)] \\
O_{63} &= \frac{1}{4} [(J_+^3 + J_-^3)(11J_z^3 - (3X + 59)J_z) \\
&\quad + (11J_z^3 - (3X + 59)J_z)(J_+^3 + J_-^3)] \\
O_{64} &= \frac{1}{4} [(J_+^4 + J_-^4)(11J_z^2 - X - 38) + (11J_z^2 - X - 38)(J_+^4 + J_-^4)] \\
O_{65} &= \frac{1}{4} [(J_+^5 + J_-^5)J_z + J_z(J_+^5 + J_-^5)] \\
O_{66} &= \frac{1}{2} [J_+^6 + J_-^6]
\end{aligned}$$

References

- [1] J. Jensen, A.R. Mackintosh, *Rare Earth Magnetism*, Clarendon Press, Oxford, 1991.
- [2] W. Henggeler, T. Chattopadhyay, P. Thalmeier, P. Vorderwisch, A. Furrer, *Europhys. Lett.* 34 (7) (1996) 537–542.
- [3] R. Shiina, H. Shiba, P. Thalmeier, A. Takahashi, O. Sakai, *J. Phys. Soc. Jpn.* 72 (5) (2003) 1216–1225.
- [4] M.T. Hutchings, in: F. Seitz, D. Thurnbull (Eds.), *Handbook of Magnetic Materials*, vol. 16, Academic Press, New York and London, 1964, pp. 227–273.
- [5] M. Rotter, *J. Magn. Mater.* 272–276 (2004) e481–e482.
- [6] K.W.H. Stevens, *Proc. Phys. Soc. A* 65 (1952) 209.