Crystal field, magnetic anisotropy, and excitations in rare-earth hexaborides

Gennadi Uimin
Landau Institute for Theoretical Physics, Chernogolovka, 142432 Moscow Region, Russia

Wolfram Brenig
Institut für Theoretische Physik, Technische Universität Braunschweig, 38106 Braunschweig, Germany

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We clarify the role of crystalline electric field (CEF) induced magnetic anisotropy in the ground state and spin-wave spectrum of cubic rare-earth materials with dominating isotropic magnetic exchange interactions. In particular we study the hexaboride NdB_6 which is shown to exhibit strong spin-quadrupolar coupling. The CEF scheme is analyzed and a noncollinear magnetization response is found. The spin orientation in the antiferromagnetically ordered ground state is identified. Moreover, the spin excitations are evaluated and in agreement with inelastic neutron scattering a suppression of one of the two magnetic modes in the strong-coupling regime is predicted.

INTRODUCTION

Over the past two decades cubic rare-earth hexaborides RB_6 (R, rare-earth element) with CaB_6-type crystal structure have been at the center of numerous studies of materials with crystalline-electric-field (CEF) driven, nontrivial ordering phenomena. Among these compounds, CeB_6 (e.g., Ref. 1) serves as a prototypical system which exhibits an impressively complex phase diagram. In this material the CEF of cubic symmetry selects the Γ_8 quartet to be the ground state of the Ce^{3+} ions ($\mathcal{J}=5/2$). The latter quartet is well separated from the next-highest Γ_7 doublet by an energy gap of the order of 540 K.[2] Thus, on a low-energy scale, the physics of CeB_6 is reasonably well described by projecting onto the Γ_8 subspace. Similar systems with Γ_8 ground states can be realized starting from the right side of the rare-earth series, i.e., invoking compounds of cubic symmetry with Yb^{3+} or Tm^{2+} ions, whose incomplete f shell contains 13 electron or one f hole. In accordance with Hund's rule and contrast to the Ce case, however, the Γ_8 basis has to be constructed from a $\mathcal{J}=7/2$ multiplet, breaking direct electron-hole symmetry thereby.

In this paper we will focus on the hexaboride NdB_6. Although investigated in detail experimentally by inelastic neutron scattering (INS),[3,4] the anisotropy of the magnetically ordered state below the temperature T_C of order $T_C \approx 8.6\,\mathrm{K}$ (Refs. 5 and 4) remains unclear as well as the existence of only a single magnetic mode as observed by INS. The aim of our work is to consider these open issues.

CRYSTALLINE ELECTRIC FIELD

The CEF level scheme of the Nd^{3+} multiplet (three f electrons, $\mathcal{J}=9/2$, $S=3/2$, and $L=6$) is consistent with the sequence $\Gamma_8^{(2)}(0\,\mathrm{K})$-$\Gamma_8^{(1)}(135\,\mathrm{K})$-$\Gamma_6(278\,\mathrm{K})$.[3] Similar to CeB_6 the energy gap separating the lowest quartet is large enough to restrict the Hilbert space to $\Gamma_8^{(2)}$ only. The basis states of this Γ_8 manifold are

$$\psi_{+\uparrow}=v_1|+9/2\rangle+v_2|+1/2\rangle+v_3|-7/2\rangle,$$

$$\psi_{-\uparrow}=w_1|+5/2\rangle+w_2|-3/2\rangle, \quad (1)$$

$$\psi_{+\downarrow}=v_1|-9/2\rangle+v_2|-1/2\rangle+v_3|+7/2\rangle,$$

$$\psi_{-\downarrow}=w_1|-5/2\rangle+w_2|+3/2\rangle. \quad (2)$$

The coefficients v_i and w_i are derived from the Stevens operator formulation of the CEF Hamiltonian[6] using the CEF parameters reported in the literature.[3] For NdB_6 one finds

$$v_1=0.1437, \quad v_2=-0.3615, \quad v_3=0.9212,$$

$$w_1=-0.9223, \quad w_2=0.3865. \quad (3)$$

The states in Eqs. (1) and (2) have been labeled such that the second index denotes a ''spin''-like projection, whereas the first index stands for two ''orbital''-like components which reflect the different shapes of the electron wave functions. This leads to a description of the quartet in terms of two Pauli matrices $\boldsymbol{\sigma}$ and $\boldsymbol{\tau}$:[8–10]

$$\sigma^z\psi_{\tau\pm}=\pm 1/2\psi_{\tau\pm}, \quad \sigma^{\pm}\psi_{\tau\mp}=\psi_{\tau\pm},$$

$$\tau^z\psi_{\pm\sigma}=\pm 1/2\psi_{\pm\sigma}, \quad \tau^{\pm}\psi_{\mp\sigma}=\psi_{\pm\sigma}.$$

Now the magnetic-moment operator can be represented in terms of $\boldsymbol{\sigma}$ and $\boldsymbol{\tau}$ by

$$M_\alpha=\mu_B(\xi+2\eta T_\alpha)\sigma_\alpha \quad (\alpha=x,y,z). \quad (4)$$

Here $\mathbf{T}$ is a vector with components

$$T_x=-\frac{1}{2}\tau_z+\frac{\sqrt{3}}{2}\tau_x, \quad T_y=-\frac{1}{2}\tau_z-\frac{\sqrt{3}}{2}\tau_x, \quad T_z=\tau_z, \quad (5)$$

which transforms according to the Γ_3 representation.

Evaluating the $\mathcal{J}=9/2$ angular momentum matrix elements in the Γ_8 basis (1) and comparing with Eq. (4) the values of ξ and η for NdB_6 are obtained as

$$\xi=-0.661, \quad \eta=-6.857. \quad (6)$$

This identifies NdB_6 as a system with strong coupling of the magnetic and quadrupolar degrees of freedom ($|\eta| \geqslant |\xi|$). Note that for Γ_8 states with one f electron (hole) ξ and η are universal and do *not* depend on the CEF splitting parameters. For Ce^{3+}, $\xi=2$ and $\eta=8/7$; for Yb^{3+} and Tm^{2+}, $\xi=-8/3$ and $\eta=-32/21$. Therefore CeB_6 and possible Yb and Tm candidates exhibit rather weak spin-quadrupolar coupling with a characteristic parameter $\eta/(2\xi)=2/7$.

EXCHANGE ANISOTROPY

In this section we clarify the spin orientation in the magnetically ordered ground state. Most likely, the dominant interaction in NdB_6 is of isotropic magnetic exchange type.[4] However, due to the Γ_8 ground state, a CEF induced magnetic anisotropy exists which *depends on the ratio* ξ/η. This can be understood by considering the single-ion Zeeman interaction, i.e., $-\Sigma_\alpha H_\alpha M_\alpha$ in an external magnetic field $\mathbf{H}$. Inserting M_α from Eq. (4) one obtains a 4×4 matrix which is easy to diagonalize with eigenvalues λ,

$$\lambda = \pm\sqrt{\xi^2+\eta^2 \pm |\eta| \sqrt{(3\eta^2/2-2\xi^2)-3F(\mathbf{n})(\eta^2/2-2\xi^2)}}, \tag{7}$$

measured in units of $g\mu_B H/2$. This clearly manifests a cubic anisotropy through the function $F(\mathbf{n})$:

$$F(\mathbf{n}) = n_x^4+n_y^4+n_z^4, \quad \mathbf{n}=\mathbf{H}/H. \tag{8}$$

The anisotropy results in a noncollinearity of the magnetic field and the magnetization for any general orientation of $\mathbf{H}$. Exceptions are the directions [111], [110], and [001] and their crystallographic equivalents. Energetically favorable states are related either to the cubic axes ([001] type), if $|\eta|<2|\xi|$, or the cubic diagonals ([111] type), if $|\eta|>2|\xi|$. The anisotropy caused by the CEF disappears, if $|\eta|=2|\xi|$. Therefore, we may conclude that Ce^{3+}, Yb^{3+}, and Tm^{2+} Γ_8 compounds tend to exhibit ''easy axis'' anisotropy [$\eta/(2\xi)=2/7$], whereas for Nd^{3+} in NdB_6 we have $\eta/(2\xi)\approx 5.19$ which results in ''easy diagonal'' anisotropy.

Within a mean-field treatment of the exchange interaction

$$-\sum_{\mathbf{R},\mathbf{R}'} J_{\mathbf{R}\mathbf{R}'}\mathbf{S}_\mathbf{R}\cdot\mathbf{S}_{\mathbf{R}'}, \tag{9}$$

where $J_{\mathbf{R}\mathbf{R}'}$ is the exchange integral and $\mathbf{S}_\mathbf{R}$ the spin at site $\mathbf{R}$, the magnetic field in Eq. (7) and (8) has to be replaced by the Weiss field $J_0\langle\mathbf{S}\rangle/(g\mu_B)$ with $J_0=\Sigma_{\mathbf{R}'}J_{\mathbf{R}\mathbf{R}'}$ if ferromagnetic exchange is dominant. The Landé factor in NdB_6 is $g=8/11$. For bipartite antiferromagnetism (AFM), the Weiss field on sublattice A is proportional to $-J_0\langle\mathbf{S}_A\rangle+J_1\langle\mathbf{S}_B\rangle$ with $J_{0(1)}=(-)\Sigma_{\mathbf{R}'}J_{\mathbf{R}\mathbf{R}'}$ for $\mathbf{R}$ and $\mathbf{R}'$ on equal (opposite) sublattices. On sublattice B, one should replace $A\leftrightarrow B$.

Therefore, in conclusion, we expect [111] orientational ordering in the ground state of NdB_6 if isotropic exchange interactions are dominant.[11]

MAGNETIC EXCITATIONS

In this section we focus on the spin dynamics by considering the time-dependent magnetic susceptibility

$$\chi^S_{\alpha\beta}(\mathbf{k},t) = i\Theta(t)\langle[S_{\alpha\mathbf{k}}(t),S_{\beta-\mathbf{k}}]\rangle. \tag{10}$$

Lower Greek indices of χ and the spin operator refer to x, y, z and boldface vectors $\mathbf{k}$ denote the momentum. We use a spin operator rescaled by η^{-1}, i.e., $S_{\alpha\mathbf{k}}=M_{\alpha\mathbf{k}}/(g\mu_B\eta)$. Therefore the dependence of the magnetic spectrum on the CEF can be expressed solely in terms of the ratio ξ/η. To evaluate Eq. (10) we proceed via a mean-field analysis consistent with AFM ordering[5] on a bipartite lattice. Rather than employing the Pauli-matrix representation[8–10] of Eq. (10) we perform this analysis using a dyadic basis to express the spin operator within the Γ_8 manifold:[12]

$$S_{\alpha\mathbf{k}} = \frac{1}{\sqrt{2}} S_\alpha^{\mu\nu}(a_\mathbf{k}^{\mu\nu}+b_\mathbf{k}^{\mu\nu}),$$

$$a_\mathbf{k}^{\mu\nu} = \sqrt{\frac{2}{N}}\sum_\mathbf{R} e^{-i\mathbf{k}\cdot\mathbf{R}} a_\mathbf{R}^{\mu\nu}, \tag{11}$$

where a summation over repeated indices is implied for the remainder of this paper, $b_\mathbf{k}^{\mu\nu}$ is defined analogous to $a_\mathbf{k}^{\mu\nu}$ with, however, $\mathbf{R}\rightarrow\mathbf{R}'$, and

$$a_\mathbf{R}^{\mu\nu} = |\mu\mathbf{R}\rangle\langle\nu\mathbf{R}|, \quad b_{\mathbf{R}'}^{\mu\nu} = |\mu\mathbf{R}'\rangle\langle\nu\mathbf{R}'| \tag{12}$$

are the dyades on sites $\mathbf{R}$ ($\mathbf{R}'$) of the magnetic A (B) sublattice. $|\mu\rangle$ are the eigenstates of the z component of the spin in the Γ_8 manifold $S_{\alpha=z}|\mu\rangle = s_\mu|\mu\rangle$. The spin should be *quantized along (against)* the [111] direction of the Weiss-field on the A (B) sublattice sites. $S_\alpha^{\mu\nu}$ are the matrix elements of the spin corresponding to the latter quantization direction. The dyadic transition operators $a_\mathbf{k}^{\mu\nu}$ and $b_\mathbf{k}^{\mu\nu}$ with $\mu,\nu=1,\ldots,4$ can be recast into a 32-component operator $A_\mathbf{k}^{\gamma=1,\ldots,32}=\{a_\mathbf{k}^{(1,1),\ldots,(4,4)}, b_\mathbf{k}^{(1,1),\ldots,(4,4)}\}$ with a corresponding 32×32 matrix susceptibility of the $A_\mathbf{k}^\gamma$ operators

$$\chi^{\mu\nu}(\mathbf{k},t) = i\Theta(t)\langle[A_\mathbf{k}^\mu(t),A_\mathbf{k}^{\nu\dagger}]\rangle. \tag{13}$$

The original magnetic susceptibility (10) can be obtained from this by projecting the dyades onto the magnetic moment

$$\chi_{\alpha\beta}(\mathbf{k},t) = \frac{1}{2}\chi^{\mu\nu}(\mathbf{k},t)C_{\beta\alpha}^{\nu\mu}, \tag{14}$$

where $C_{\beta\alpha}^{\nu\mu}=v_\beta^{\nu\star}v_\alpha^\mu$ with $v_{\alpha=x,y,z}^{\mu=1,\ldots,32}=\{S_\alpha^{(1,1),\ldots,(4,4)}, S_\alpha^{(1,1),\ldots,(4,4)}\}$ is a 32-component vector for each spin component α.

To proceed we evaluate the equation of motion (EQM) of the dyadic susceptibility:

$$i\partial_t\chi^{\mu\nu}(\mathbf{k},t) = -\delta(t)\langle[A_\mathbf{k}^\mu, A_\mathbf{k}^{\nu\dagger}]\rangle + i\Theta(t)\langle[[A_\mathbf{k}^\mu(t),H],A_\mathbf{k}^{\nu\dagger}]\rangle. \tag{15}$$

In this paper we concentrate on the spin dynamics for next-neighbor (NN) AFM exchange couplings J only. Therefore, setting $J\eta^2/g^2$ to unity the Hamiltonian in terms of the dyades reads

$$H = \sum_{\mathbf{R},\mathbf{l}} S_\alpha^{\mu\nu}S_\alpha^{\lambda\sigma}a_\mathbf{R}^{\mu\nu}b_{\mathbf{R}+\mathbf{l}}^{\lambda\sigma}, \tag{16}$$

where $\mathbf{l}$ runs over the NN sites of $\mathbf{R}$. The real-space representation of the commutator on the right-hand side of the EQM is evaluated using the algebra of the dyades, yielding

$$[a_{\mathbf{R}}^{\mu\nu},H]=\sum_{\mathbf{l}}\,(S_{\alpha}^{\nu\omega}a_{\mathbf{R}}^{\mu\omega}-S_{\alpha}^{\omega\mu}a_{\mathbf{R}}^{\omega\nu})S_{\alpha}^{\lambda\sigma}b_{\mathbf{R}+\mathbf{l}}^{\lambda\sigma}. \tag{17}$$

An analogous expression results on the B sublattice. On the mean-field level the EQMs are closed by factorizing all quadratic terms in Eq. (17) according to the scheme $a_{\mathbf{R}}^{\mu\nu}b_{\mathbf{R}'}^{\lambda\sigma}=\langle a_{\mathbf{R}}^{\mu\nu}\rangle b_{\mathbf{R}'}^{\lambda\sigma}+a_{\mathbf{R}}^{\mu\nu}\langle b_{\mathbf{R}'}^{\lambda\sigma}\rangle$. Moreover, "up" ("down") [111] polarization on the A (B) sublattice is enforced by setting

$$\langle a_{\mathbf{R}}^{\mu\nu}\rangle=\delta^{\mu1}\delta^{\nu1},\quad \langle b_{\mathbf{R}'}^{\mu\nu}\rangle=\delta^{\mu4}\delta^{\nu4}. \tag{18}$$

In momentum space the linearization results in

$$\begin{aligned}[a_{\mathbf{k}}^{\mu\nu},H]&=zS_{\alpha}^{44}(S_{\alpha}^{\nu\sigma}\delta^{\mu\lambda}-S_{\alpha}^{\lambda\mu}\delta^{\nu\sigma})a_{\mathbf{k}}^{\lambda\sigma}\\&\quad+z\gamma_{\mathbf{k}}(\delta^{1\mu}S_{\alpha}^{\nu1}-S_{\alpha}^{1\mu}\delta^{\nu1})S_{\alpha}^{\lambda\sigma}b_{\mathbf{k}}^{\lambda\sigma}\\&=z(L_{\mathbf{k}11}^{\mu\nu\lambda\sigma}a_{\mathbf{k}}^{\lambda\sigma}+L_{\mathbf{k}12}^{\mu\nu\lambda\sigma}b_{\mathbf{k}}^{\lambda\sigma}),\end{aligned} \tag{19}$$

where z is the coordination number and $z\gamma_{\mathbf{k}}=\Sigma_{\mathbf{l}}e^{i\mathbf{k}\cdot\mathbf{l}}$. A similar equation arises for $[b_{\mathbf{k}}^{\mu\nu},H]$ introducing two additional 16×16 matrices $L_{\mathbf{k}22}^{\mu\nu\lambda\sigma}$ and $L_{\mathbf{k}21}^{\mu\nu\lambda\sigma}$. Switching to frequency space the EQMs can be solved as

$$\chi_{\alpha\beta}^{S}(\mathbf{k},\omega)=-\mathrm{Tr}[(\omega\mathbf{1}-z\mathbf{L}_{\mathbf{k}})^{-1}\boldsymbol{\chi}_0]^{\mu\nu}[\mathbf{C}_{\beta\alpha}^{T}]^{\mu\nu}, \tag{20}$$

where boldface symbols refer to matrix notation in a 32×32 space. $\mathbf{L}_{\mathbf{k}}$ is set by $L_{\mathbf{k},ij}^{\mu\nu\lambda\sigma}$ with $i,j=1,2$ labeling four 16×16 subblocks. Similarly $\boldsymbol{\chi}_0$ consists of four subblocks $\chi_{0,ij}^{\mu\nu\lambda\sigma}$ with $\chi_{0,i\neq j}^{\mu\nu\lambda\sigma}=0$ and $\chi_{0,11(22)}^{\mu\nu\lambda\sigma}=\delta^{\nu\sigma}\delta^{\mu1(4)}\delta^{\lambda1(4)}-\delta^{\lambda\mu}\delta^{\sigma1(4)}\delta^{\nu1(4)}$.

Equation (20) allows for substantial simplifications. First, all diagonal dyades, i.e., $a(b)_{\mathbf{k}}^{\mu\mu}$, commute with H. Second, the linearized form of Eq. (17) for the nondiagonal dyades, i.e., for $a(b)_{\mathbf{k}}^{\mu\nu}$ with $\mu\neq\nu$, is diagonal with respect to $\mu\nu$ *and* remains local for nearly all pairs $\mu\nu$. This follows from the identity

$$S_{\alpha}^{11(44)}S_{\alpha}^{\mu\nu}=0. \tag{21}$$

The only set of dyades which couple dispersively via the EQMs is

$$B_{\mathbf{k}}^{\gamma=1,\ldots,4}=\{a_{\mathbf{k}}^{(1,2)},a_{\mathbf{k}}^{(3,1)},b_{\mathbf{k}}^{(3,4)},b_{\mathbf{k}}^{(4,2)}\}, \tag{22}$$

and the corresponding Hermitian conjugate set $B_{\mathbf{k}}^{\gamma=1,\ldots,4\dagger}$. From the preceding discussion it is conceivable that the complete spin dynamics can be expressed in terms of the physically *relevant dyades* $B_{\mathbf{k}}^{\gamma=1,\ldots,4(\dagger)}$ only. In fact, after some elementary rearrangements of the matrix EQM (20), the longitudinal spin susceptibility, which, due to cubic symmetry, is identical to the three-trace $\chi_{\alpha\alpha}^{S}(\mathbf{k},\omega)$, simplifies to

$$\chi_{\alpha\alpha}^{S}(\mathbf{k},\omega)=-\mathrm{Tr}[D^{-1}N], \tag{23}$$

where the dynamical matrix D and the static susceptibility-matrix N are identical to $[(\omega/z)\mathbf{1}-\mathbf{L}_{\mathbf{k}}]$ and $\boldsymbol{\chi}_0\mathbf{C}_{\alpha\alpha}/z$ *restricted* to within the four-dimensional subspace spanned by Eq. (22). The complex conjugate dyades $B_{\mathbf{k}}^{\mu\dagger}$ introduce an overall prefactor of 2 only. After some algebra we find that D and N are determined by five parameters a, b, c, d, and e through

$$D=\begin{bmatrix}w-a & 0 & -c\gamma_{\mathbf{k}} & -e\gamma_{\mathbf{k}}\\ 0 & w-b & -e\gamma_{\mathbf{k}} & -d\gamma_{\mathbf{k}}\\ c\gamma_{\mathbf{k}} & -e\gamma_{\mathbf{k}} & w+a & 0\\ -e\gamma_{\mathbf{k}} & d\gamma_{\mathbf{k}} & 0 & w+b\end{bmatrix},$$

$$N=\frac{1}{z}\begin{bmatrix}c & -e & c & e\\ e & -d & e & d\\ -c & e & -c & -e\\ e & -d & e & d\end{bmatrix}, \tag{24}$$

with $w=\omega/z$ and

$$a=S_{\alpha}^{44}(S_{\alpha}^{22}-S_{\alpha}^{11}),\quad b=S_{\alpha}^{44}(S_{\alpha}^{11}-S_{\alpha}^{33}),$$

$$c=S_{\alpha}^{21}S_{\alpha}^{34},\quad d=-S_{\alpha}^{13}S_{\alpha}^{42},\quad e=-S_{\alpha}^{13}S_{\alpha}^{34}=\sqrt{cd}. \tag{25}$$

With this the longitudinal spin susceptibility of Eq. (23) is obtained readily as

$$\chi_{\alpha\alpha}^{S}(\mathbf{k},\omega)=\frac{Z(\mathbf{k},w)/z}{(w^2-w_1^2)(w^2-w_2^2)}, \tag{26}$$

where the weight factor $Z(\mathbf{k},w)$ given by

$$\begin{aligned}Z(\mathbf{k},w)&=2[ac-bd-(c-d)^2\gamma_{\mathbf{k}}]w^2\\&\quad+(ad-bc)[ab+(ad-bc)\gamma_{\mathbf{k}}],\end{aligned} \tag{27}$$

and the excitation energies $\pm w_{1,2}(\mathbf{k})$ are being set by the roots of the biquadratic equation

$$w^4+w^2[(c-d)^2\gamma_{\mathbf{k}}-(a^2+b^2)]+a^2b^2-(ad-bc)^2\gamma_{\mathbf{k}}=0. \tag{28}$$

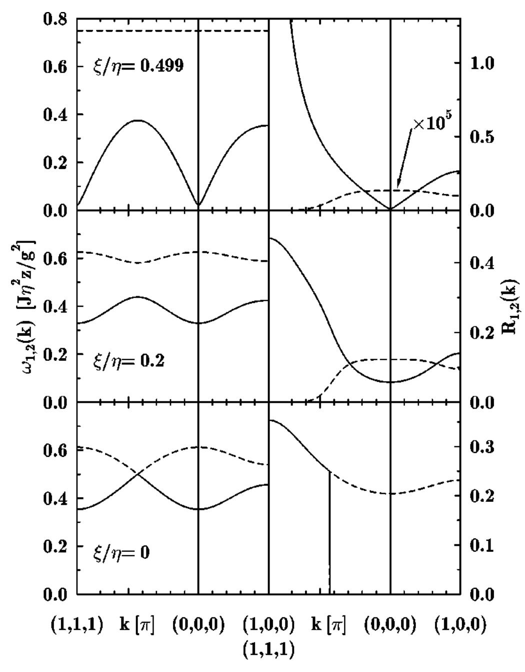

FIG. 1. Dispersion and weight of spin excitations.

In Fig. 1 the dispersion as well as the weight $R_{1,2}(\mathbf{k}) = \chi^S_{\alpha\alpha}(\mathbf{k},\omega)(\omega-\omega_{1,2}(\mathbf{k})|_{\omega=\omega_{1,2}(\mathbf{k})}$ of the two positive-frequency modes is depicted along a path in the Brillouin zone (BZ) ranging from $\mathbf{k}=(1,1,1)$ to (0,0,0) to (1,0,0) for various values of the anisotropy ratio ξ/η. This figure clarifies the concluding issue aimed at in this paper, i.e., the observation of only a *single* excitation mode in NdB_6. Based on the eigenvalues (7) *two* excitations of comparable energy are expected in the Weiss field of the AFM state at $\xi/\eta \ll 1$. However, Fig. 1 shows that only a single mode carries significant weight at small ξ/η. Furthermore, in agreement with the spectrum of a single-ion pseudospin $\mathcal{J}=3/2$, the system exhibits a single-mode spin-wave-like excitation at the isotropic point $2\xi=\eta$. Only for intermediate anisotropy do both modes show sizable weight at any given point in the BZ.

CONCLUSION

In summary we have considered rare-earth (RE) compounds of cubic symmetry with a Γ_8-quartet ground state of the RE ions. Particular emphasis has been put on the hexaboride NdB_6. Analyzing the CEF splitting we have identified NdB_6 to be a genuine example of a system with strongly coupled magnetic and quadrupolar degrees of freedom.

We have studied the CEF induced intrinsic magnetic anisotropy superimposed onto an isotropic exchange interaction revealing that NdB_6 should display magnetic anisotropy of a different type, i.e., ''easy diagonal,'' as compared to Ce or Yb compounds which show ''easy axis'' anisotropy.

The magnetic anisotropy leads to a noncollinear **M** vs **H** behavior and it is tempting to speculate that angular-dependent magnetization measurements on the corresponding RE cubic compounds, as well as diluted systems, e.g., $La_{1-x}Ce_xB_6$, should be able to detect this behavior.

We have evaluated the magnetic excitations in the AFM state of an ''easy diagonal'' type using a dyadic operator approach. For systems with strong spin-quadrupolar coupling this method is superior[7] to less controlled pseudoparticle descriptions which are applicable to the weak-coupling system CeB_6 and are based on the conventional $\boldsymbol{\sigma}$-$\boldsymbol{\tau}$ Pauli-matrix representation (4). In accordance with the number of independent Pauli matrices ($\boldsymbol{\sigma}$ and τ), we find two branches of spin excitations. However, the spectral weights in the two magnetic channels are very different in a strongly coupled spin-quadrupolar system. In fact, in the $\xi=0$ limit one channel disappears completely. This is reminiscent of the INS data on NdB_6 (Ref. 4) which display only one branch of spin excitations. Although derived by a linearization of the EQMs we believe that our results are quite robust against nonlinear corrections since the spin-wave spectrum in the nonisotropic case is gapful. This should diminish the relevance of quantum fluctuations.

Finally, regarding a direct comparison to experimental data we note that NdB_6 displays a [0,0,1/2] wave vector of the AFM modulation. This requires the inclusion of longer-range exchange interactions which have been neglected in this paper. These will be studied elsewhere.[7]

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