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Theory of multipolar excitations and neutron scattering spectra of CeB_6

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Abstract
Multipolar excitations in the antiferroquadrupolar ordering phase of CeB_6 are studied theoretically. We develop the method of boson expansion of multipoles, and apply it to the Ruderman–Kittel–Kasuya–Yosida model, which has been introduced previously for CeB_6. Then the neutron scattering spectra are calculated within the dipole approximation and compared with experimental results obtained by Bouvet. The origin of the characteristic peak structures and their dependence on the magnetic field are discussed.

1. Introduction

The anomalous antiferroquadrupolar (AFQ) phase transition in the dense Kondo compound CeB_6 has been attracting renewed interest [1, 2]. Recent theoretical and experimental studies have succeeded in determining the symmetry of the AFQ order parameter, and clarifying the important role of a hidden octupolar moment [3–5]. In this work we investigate the excitations and the dynamics of the multipolar moments on the basis of the Ruderman–Kittel–Kasuya–Yosida (RKKY) model, which has been used previously to analyse the thermodynamics in the AFQ phase. This analysis enables us to give physical interpretations of the neutron scattering experiments carried out by Bouvet [6].

2. Boson expansion of multipolar fluctuations

As we have shown in previous work, one should consider three dipoles, five quadrupoles, and seven octupoles as the active degrees of freedom in the Γ_8-quartet basis. These 15 multipoles were defined in [7] as tensor operators on J, and such representations manifest their symmetry properties explicitly in the cubic lattice. In this work, however, we use a simple numbering of multipoles, which is denoted by X^α ($\alpha = 1, \ldots, 15$), to present general properties irrespective of the detailed form of the interaction.

Here we apply the Holstein–Primakoff (HP) method to study the multipolar dynamics [8, 9]; this is equivalent to the random-phase approximation given by Thalmeier *et al* [10] at zero temperature. Let us start by introducing the most general form of the interactions in the quartet basis:

$$H = \sum_{(ij)} \sum_{\alpha\beta} X_i^\alpha D_{ij}^{\alpha\beta} X_j^\beta - \sum_i \sum_\alpha h^\alpha X_i^\alpha, \tag{1}$$

where i and j represent site indices and $D_{ij}^{\alpha\beta}$ is the interaction strength. Since the mean-field solution is usually complicated, depending on the interactions and the magnetic fields, we describe here the general aspects of the formulation without making assumptions specific to CeB_6.

First a unitary transformation U depending on the site i is introduced via $|\Psi_n(i)\rangle = \sum_m U_{mn}(i)|\phi_m\rangle$, so Ψ_0 should be the mean-field ground state. Then the multipolar operators X^α can be expressed as linear combinations of the so-called Hubbard operators $S_{nn'}(i) = |\Psi_n(i)\rangle\langle\Psi_n'(i)|$; one can write $X^\alpha = \sum_{nn'} x_{nn'}^\alpha(i) S_{nn'}(i)$ with $x_{nn'}^\alpha$ being the matrix elements of X^α in the new basis.

In order to make model (1) tractable in a systematic approximation, we introduce a set of boson operators a_n and $a_n^\dagger$ for Ψ_n. In general, the Hubbard operators can be described in terms of the products of those boson operators as $S_{nn'} = a_n^\dagger a_{n'}$, provided that the Hilbert space should be restricted by the constraint for the total number of bosons $\sum_{n=0}^3 a_n^\dagger a_n = M$. Although $M = 1$ in the real system, we generalize the constraint and regard M^{-1} as an expansion parameter of the approximation. Then, eliminating a_0 and $a_0^\dagger$ one can set up the generalized HP transformation:

$$\begin{aligned} S_{00} &= M - \sum_{m=1}^{3} a_m^\dagger a_m, & S_{nn'} &= a_n^\dagger a_n', \\ S_{n0} &= \sqrt{M} a_n^\dagger + \mathrm{O}(M^{-1/2}), & S_{0n} &= \sqrt{M} a_n + \mathrm{O}(M^{-1/2}), \end{aligned} \tag{2}$$

with $n, n' \neq 0$. Using this transformation, one arrives at the expansion of multipolar operators in powers of M^{-1}:

$$X^\alpha = M x_{00}^\alpha + \sqrt{M} \sum_{n=1}^{3} (x_{n0}^\alpha a_n^\dagger + x_{0n}^\alpha a_n) + \sum_{n=1}^{3} \sum_{n'=1}^{3} (x_{nn'}^\alpha - x_{00}^\alpha \delta_{nn'}) a_n^\dagger a_{n'} + \cdots. \tag{3}$$

Here the first term represents the static order parameter while the second and third terms are the multipolar fluctuations associated with the excitations. The order parameter symmetries for various field directions have already been analysed in previous work [7]. Some useful symmetry properties in the fluctuation part will be discussed elsewhere [11].

Substituting (3) into (1), the boson expansion of the Hamiltonian can be obtained formally as

$$H = M^2 \sum_{n=0}^{\infty} M^{-n/2} H_n, \tag{4}$$

where H_n represents the n-boson Hamiltonian with the normal ordering of the operators. In particular, analysing the bilinear term H_2, we study the excitation spectra of the RKKY model for CeB_6 in the next section.

3. Neutron scattering spectra

The neutron cross-section is given within the dipole approximation as

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}E\,\mathrm{d}\Omega} \propto I(\boldsymbol{k}\omega) = \sum_{\alpha\beta} (\delta_{\alpha\beta} - \kappa_\alpha \kappa_\beta) S^{\alpha\beta}(\boldsymbol{k}, \omega), \tag{5}$$

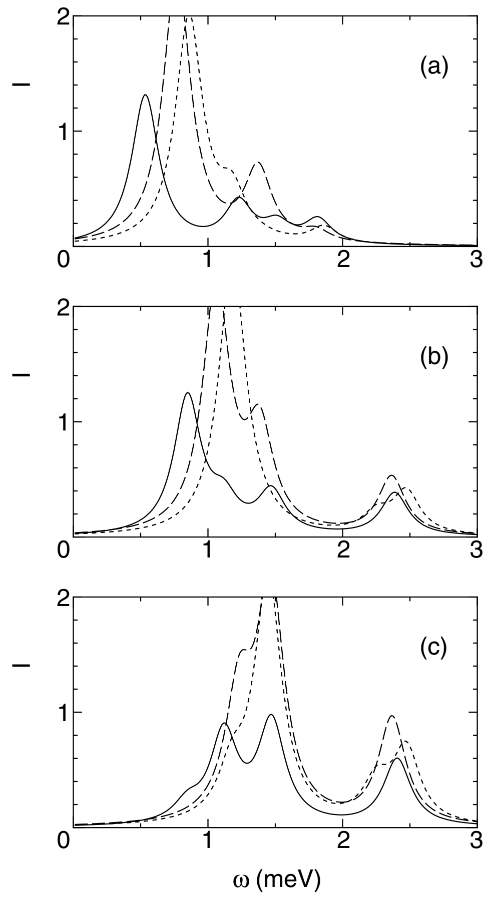

Figure 1. Neutron scattering spectra as function of energy for (a) $\boldsymbol{k} = (0, 0, 0)$, (b) $\boldsymbol{k} = (1/4, 1/4, 0)$, and (c) $\boldsymbol{k} = (1/2, 1/2, 0)$. The solid, dotted, and broken curves are the results for the (001), (111), and (110) fields respectively. The parameters in the RKKY model are $\epsilon_Q = 0.5$, $\epsilon_O = 0.5$, and $h = 2.0$. (See [12] for details of the model.)

where α and β run over x, y and z respectively, and $\kappa_\alpha = k_\alpha/|\boldsymbol{k}|$. The dynamical structure function $S^{\alpha\beta}$ at zero temperature is related to the dipole–dipole Green function, which is calculated from the boson Hamiltonian. Physically, the spectra should have peak structures around the excitation energies whose intensities represent the magnitudes of the dipolar polarizations of the modes.

Here let us discuss the theoretical results for the scattering spectra. The model analysed here is the same as that used in [12]. In figure 1 we show the spectra in finite fields for the momenta $\boldsymbol{k} = (k, k, 0)$, using the linewidth $\Gamma = 0.16$ meV. The solid, dotted, and broken curves are the results for the (001), (111), and (110) fields respectively. For various momenta and field directions, the results are characterized commonly by a large peak around $\omega \sim 1$ meV and a small peak above 2 meV. The small peak is identified as arising from the modes with simultaneous quadrupolar and octupolar flips, which tend to be isolated in the high-energy region, reflecting the large octupolar interaction. This characteristic feature of the spectra has been observed experimentally by Bouvet for various parameters [6].

It should be noted that the anisotropy of the spectra for the field directions is not small even in the low-field region. This is partly due to the anisotropy of the structure function $S^{\alpha\beta}$ originating from the order parameters and the Zeeman term. The other cause is that the components of the structure function contributing to the spectra depend on the field directions through (5), reflecting the change of the relative angle between the momentum and the field direction. As a result, one finds that the peak structure at low energy becomes sharper in the (111) and (110) fields than in the (001) field. Following the dispersions of excitation energies, each peak shows a shift of the position depending on the momentum, together with the characteristic change of the intensity; such momentum dependences of the spectra are also consistent with experiments [6].

4. Summary

In summary, we have studied excitations and neutron scattering spectra of the AFQ phase of CeB_6, by means of the method of boson expansion of multipoles. It has been found that the spectra have a large peak at low energy and a small peak at high energy, reflecting the large octupolar interaction. Those characteristics of the spectra and their dependence on momenta are shown to agree well with experiments carried out by Bouvet. The detailed description of the model and the analysis in this work will be given in a forthcoming paper [11].

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