Anisotropic Form Factors of Neutron Scattering by Magnetic Octupole in CeB$_6$

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Abstract. The form factor of elastic neutron scattering is calculated for the quadrupole order phase of CeB$_6$ in magnetic fields. It is shown that the dependence of the form factor on the direction of neutron momentum transfer is very small for the Bragg reflections, whereas the scattering due to field-induced octupoles gives rise to a significant anisotropy of the form factor for the super-lattice reflections. These results are discussed in quantitative comparison with recent experimental results. The similarity and dissimilarity with the results for phase IV in Ce$_{1-x}$La$_x$B$_6$ is also discussed.

1. Introduction

Recently, the multipole effects in $f$ electron systems have been widely studied in relation with their anomalous ordered phases. A well-known example having competing interactions of high-rank multipoles is CeB$_6$, which exhibits an antiferro-quadrupole (AFQ) order phase at low temperatures. It is established that some unusual properties of the AFQ phase in magnetic fields originate from large influences of field-induced magnetic octupoles.[1, 2] A number of proposals on other materials after clarifying the AFQ problem in CeB$_6$ have been found in recent review articles such as [3, 4]. Among them, it is quite remarkable that the realizations of pure octupole phases at zero magnetic field are promising for Ce$_{1-x}$La$_x$B$_6$ ($x < 0.8$) and NpO$_2$.[5, 6, 7]

In general, however, the direct observation of high-rank multipoles and the quantitative estimation of their magnitudes remain to be a difficult problem. In this paper, we report theoretical results on the magnetic form factor of elastic neutron scattering for the AFQ phase of CeB$_6$ in magnetic fields. It is shown that the form factor as a function of neutron momentum transfer becomes very anisotropic for the super-lattice reflections, under an influence of the field-induced octupoles. The comparison with the recent experimental results is discussed.

2. General Aspects of Magnetic Form Factor

In neutron scattering by a periodic array of magnetic ions, the differential cross section for the momentum transfer $\boldsymbol{\kappa} = \boldsymbol{k}' - \boldsymbol{k}$ of neutrons is expressed as [8]

$$\frac{d^2\sigma}{d\Omega dE} \propto \sum_{n,n'} P_n |\langle n' | Q_{\kappa}^\dagger (\boldsymbol{\kappa}) | n \rangle|^2 \delta(h\omega + E_n - E_{n'}).$$ (1)
Here, \( n(n') \) denotes an initial (final) state of electrons with energies \( E_{n(n')} \), and the probability for the initial state is represented by \( P_n \). In this paper, we study the elastic scattering with \( n' = n \) for the ground state of magnetic ions. It is assumed that the ground state consists of fixed-\( J \) multiplets with \( J \) being the total angular momentum of \( f \) electrons.

The full informations on the interaction between neutrons and magnetic ions are involved in the total scattering operator \( Q_i^\dagger \) which corresponds to a lattice sum of local operators \( Q_i^\dagger \) with the site index \( i \). In order to clarify the physical meaning, however, it is more useful to consider local unprojected operators \( Q_i \) defined in \( Q_i^\dagger = \kappa \times (Q_i \times \kappa) \). In particular, it is well known that \( Q_i \sim Q_i^D = \frac{2}{3} F(\kappa) J_i \) in the long wave-length region \( \kappa \sim 0 \), where \( J \) represents the dipole operator of \( f \) electrons with \( g \) being the Landé \( g \) factor. Since \( F \) is a decreasing function of \( \kappa = |\kappa| \), the dominant contribution usually comes from scattering by the dipoles. In order to study multipole orders, however, one should go beyond such a dipole approximation. Since all the operators in the \( J \) basis must be reduced to a set of tensor operators, the correction to the dipole approximation is generally given by a linear combination of odd-rank multipoles. Therefore, omitting the site index \( i \), the \( m \) component of the unprojected scattering operator \( Q \) can be expressed as

\[
Q_m(\kappa) = Q_m^D(\kappa) + \sum_n C_{mn}(\kappa) X_n,
\]

where \( X_n \) denotes a component of higher-rank magnetic multipole operators and \( C_{mn}(\kappa) \) are the expansion coefficients.

As we have shown in ref. [9], this mixture of multipoles can be predicted in terms of the symmetry of the scattering process. A basis of the symmetry analysis is the fact that the cross section must be invariant under transformations that do not change either crystal and momentum transfer. In the case of cubic CeB\(_6\), the group theory results in \( Q = 0 \) at the superlattice spots for the following octupole orders: \( T_2^3 \) and \( T_{xyz} \) for \( \kappa || (0,0,1) \), \( T_2^3 + T_2^3 + T_2^3 \) for \( \kappa || (1,1,1) \) and \( T_2^3 \) for \( \kappa || (1,1,0) \). Note that the symbols for the multipole operators classified by the symmetry are defined in terms of the angular momentum operator of \( J = 5/2 \) in ref. [1]. In addition, one can find that \( Q \) should be in parallel to \( \kappa \) in some cases, such as \( T_2^3 \) for \( \kappa || (0,0,1) \). Since the relevant quantity in the scattering is the component of \( Q \) projected onto the \( \kappa \) plane, the scattering is forbidden in all of the above cases.

### 3. Calculated Results

The component of the AFQ order parameter of CeB\(_6\) is believed to be \( O_{xy} \) for \( H || (001) \), \( O_{yz} + O_{zx} \) for \( H || (111) \), \( O_{yz} + O_{xx} \) for \( H || (110) \), all of which induces strongly an octupole \( T_{xyz} \). [2, 3, 4, 10] We consider these orders in the framework of the quartet \( \Gamma_8 \) as the crystal field ground state of \( 4f \) electrons. The ordering wave vector is \( k = (1/2, 1/2, 1/2) \) in the simple-cubic lattice of the Ce ions. Note that the AFQ ground state inevitably leaves a Kramers degeneracy, which should be lifted by an infinitesimal field strength in the model analysis. We have used for the present calculation such a state with lifting the degeneracy, which can be comparable to the experimental results for 5~10T at around 1K. See ref. [2] for the details of multipolar interactions and the mean-field theory.

First, the numerical results for the Bragg reflections in the AFQ phase in \( H || (001) \) are presented as continuous functions of the momentum transfer of neutrons in Fig. 1, which are compared with the results for the normal phase in the same figure. It is shown that the momentum dependence of the form factor in the AFQ phase is of a quite conventional form with little anisotropy, which is almost equivalent to that in the normal phase. In fact, the observation of the form factor has been reported in the literatures and is consistent with this calculation.[11, 12] Note that since the available experiments were performed at higher temperatures, the thermal fluctuation should be taken into account for the quantitative comparison.
The calculated results for the superlattice spots in the AFQ phase in $H \parallel (001)$ are plotted in Fig. 2. It is apparent that the form factor increases with $\kappa$ in the region of small momentum transfer and takes a maximum around $\sin \theta/\lambda \sim 0.6 \text{Å}$, with the intensity $2Q^\perp \sim 0.17 \mu_B$. The qualitative features are consistent with previous calculations.[13, 14] We should also remark the anisotropy for the direction of momentum transfer. It is clear that $Q^\perp$ for a series of $\kappa \sim (m,m,1)$ have large values compared with others. In particular, we confirmed that the form factor for $(m,m,m)$ vanishes, irrespective of $m$. This is nothing but the symmetry restriction discussed in §2, which characterizes the scattering by $T_{xyz}$. We have also calculated the form factor in other field directions and have found characteristic momentum dependences and anisotropies reflecting dipole and octupole ordering induced by the fields, details of which will be published elsewhere.

Quite recently, a neutron scattering experiment for phase II in $H \parallel (001)$ has been carried out by Kuwahara et al.[15] It shows that the form factor is enhanced as $\kappa$ increases. The maximum appears around $\sin \theta/\lambda \sim 0.5 \text{Å}$ with the value slightly less than $0.2 \mu_B$. In addition, the distinct anisotropy is observed, which indicates that intensities are large for $\kappa$ close to the direction $(1,1,0)$ and almost vanishing for the direction $(1,1,1)$. These features are even quantitatively consistent with the theoretical results. The agreements clearly show that the octupole $T_{xyz}$ has an almost saturation value in a weak magnetic field ($\sim 5T$), giving a strong support to the multipole RKKY model in ref. [2].

Finally, it is useful to compare the above results with the form factor in phase IV, which appears in La doped CeB$_6$. As shown in ref. [9], momentum dependence of the form factor and the absolute value of the intensities calculated for phase IV at zero field are quite similar to the present results for phase II. Note, however, that the anisotropy for the momentum transfer is completely different. It is shown that the largest intensity in phase IV appears for $\kappa \parallel (1,1,1)$.

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**Figure 1.** Magnetic form factor for the Bragg spots as a function of neutron momentum transfer $\kappa/4\pi = \sin \theta/\lambda$. The blue and red solid lines are the results for the AFQ phase in $H \parallel (001)$. The dashed lines are the corresponding results for the normal phase which is scaled by the saturation value of the magnetization $M_z = 11/7 \mu_B$.

**Figure 2.** Magnetic form factor for the super-lattice reflections as a function of neutron momentum transfer. The dots are the results at the super-lattice spots for the AFQ phase in $H \parallel (001)$. The indices $(l,m,n)$ for the dots represent $\kappa = (l/2,m/2,n/2)$. The solid lines are the results for the limit of high symmetry directions.
whereas in phase II, high intensities are obtained for \( \kappa \) close to the direction \((1, 1, 0)\). Such a difference in the anisotropy of the form factor in the same material clearly reflects the realizations of different octupole orders in phases II and IV.

4. Summary
The magnetic form factor for elastic neutron scattering due to field-induced octupolar order in the AFQ phase of CeB\(_6\) was studied theoretically. First, we reviewed the result of the symmetry properties on the basis of the general framework of neutron scattering due to octupoles. Then, we presented the numerical results of the form factor whose dependence on the neutron momentum transfers exhibits a significant anisotropy for the super-lattice reflections. It was reported that the intensities and the anisotropy of the calculated results account well for the recent experimental observations by Kuwahara et al.. The similarity and difference in the form factors for phases II and IV was also discussed. The present study provides a typical example of an identification of the symmetry and the magnitude of hidden octupole moments by neutron scattering. This method is in principle applicable to any other materials showing the multipole ordering.

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