Novel Magnetic Ordering in Neptunium Dioxide : A Mystery for Half a Century is Addressed using the Nuclear Magnetic Resonance Technique

The phase transition in Neptunium dioxide (NpO_2) was originally discovered in 1953. Since this discovery, however, it has remained a most-lasting mystery in the physics of actinide compounds. In recent works, the possibility that this phase transition can be described as one of a new class of phase transition associated with the orbital degrees of freedom has been discussed. With this in mind, we have tackled this problem anew using the nuclear magnetic resonance (NMR) technique.

Actinide compound are very exciting, because they have most of the interesting effects that are being studied in condensed matter physics nowadays. They are very complex systems to understand, and they are the least understood. One of the reasons that these systems have not been studied much is because of their radioactivity. Therefore, they require special facilities and special places. There are only a few places in the world where these compounds are studied, and International Research Center for Nuclear Materials Science, Institute for Materials Research Science is one of them.

A complexity of actinide compound arises from the fact that the orbital moment of f-electron is never quenched by the crystal field, which differentiates the f-electron systems from the d-electron systems. Furthermore, because of the strong spin-orbit interaction in a f-electron shell, their spin and orbital degrees of freedoms are tightly coupled with each other. In such the case, the relevant degrees of freedom for one ion are its magnetic and electric multipole moments, that is, charge (rank-0), dipole (rank-1), quadrupole (rank-2), and octupole (rank-3) etc. The interactions between these multipole moments bring complex phase diagram to actinide compounds, even if the f-electrons stay in the localized limit.

The phase transition in NpO_2 was originally discovered through specific heat and magnetic susceptibility measurements, which appeared to suggest a transition to a magnetic dipolar ordering state at T_0 =26 K. However, in the 1980s, such a dipole order scenario has been unambiguously ruled out by both neutron elastic scattering and Mösbauer spectroscopy measurements. In 2000, the possibility of ordering of higher rank of magnetic multipole, i.e. octupole was proposed theoretically. Soon after that, resonant x-ray scattering measurements suggested the occurrence of longitudinal triple-q type antiferro-octupolar (AFO) order below T_0 . This AFO ordered state has also been suggested by recent microscopic calculations based on the j-j coupling scheme by K.Kubo and T. Hotta (Fig.1) [1].

In order to elucidate the microscopic properties of this exotic ordered state, we have initiated the first NMR studies in NpO_2 [2,3]. Figure 2 shows the field angular (θ) dependence of the ^{17}O NMR line splitting, which illustrates the angular variation of hyperfine (HF) fields at ^{17}O nuclei [3]. In the presence of longitudinal triple-q type order, the ^{17}O NMR line splits into two inequivalent sites, *i.e.,* the $O^{(1)}$ and $O^{(3)}$ sites [2]. The nearly flat curve for the $O^{(1)}$ site corresponds to the isotropic nature of its HF field. On the other hand, the two other curves for the $O^{(3)}$ sites show a strong θ-dependence due to the anisotropic nature of their HF fields. Using the invariant form of the HF field contributions derived by O.Sakai *et al.* (TMU) by assuming the longitudinal triple-q AFO ordered state for NpO_2, we can deduce the angular dependence of HF field at O sites. As shown in Fig. 2, we have found that the angular dependences of HF field are well reproduced by the model calculation for all three O sites. This excellent agreement obtained with just three scaling parameters provides microscopic evidence for the proposed longitudinal triple-q AFO model for NpO_2.

Compared with dipolar ordering, multipolar ordering is hard to investigate by conventional techniques. In the present work, we demonstrate that microscopic investigation of multipole ordering is possible by means of NMR through the HF interactions.

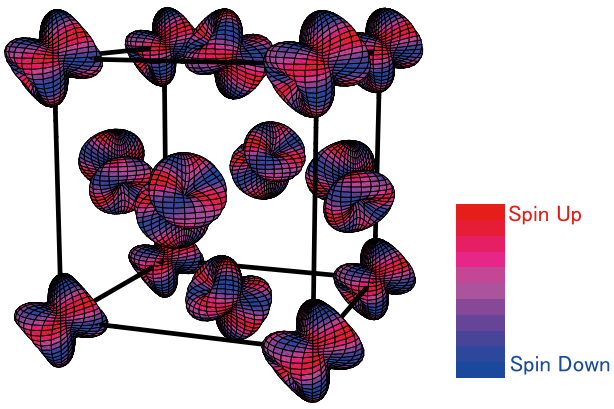

Fig.1. Magnetic-octupolar ordering state of NpO_2 calculated by K. Kubo and T. Hotta [1]. Red and Blue colors on the surface indicate the weight of up- and down-spin states.

References

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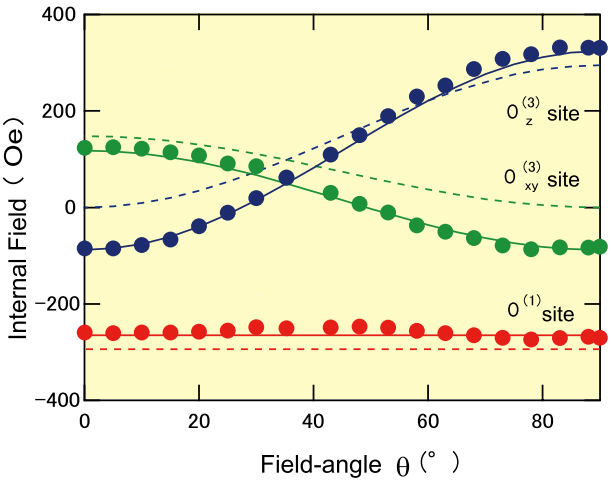

Fig.2. Field-angle dependence of the internal field at O sites in NpO_2. The solid and dotted lines are the results of model calculations with and without the contribution from the field-induced magnetic octupolar ordering, respectively [3].