NMR studies of molecular nano-magnets

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Recently, much attention has been paid to molecular nanomagnets with 3*d* transition metal (TM) elements such as $[Mn_{12}O_{12}(CH_3COO)_{16}(H_2O)_4]2CH_3COOH\cdot 4H_2O$ (for short, Mn12), $[Fe_8(N_3C_6H_{15})_6O_2(OH)_{12}][Br_8\cdot 9H_2O]$ (Fe8), *etc*., because novel quantum spin tunneling phenomena appear at low temperature. The ground state of those molecules forms a high total spin state, (for instance, S=10 for Mn12 and for Fe8) due to strong antiferromagnetic exchange coupling among the TM spins. The ground state of high spin state is split by crystal field effects, giving super paramagnetic behavior combined with quantum phenomena. In order to shed light on the particular spin states of the molecular nanomagnets from a microscopic point of view, we have carried out NMR measurements for various nuclei of Mn, Fe, H and D with applying magnetic field parallel and also perpendicular to the magnetic easy axis in a wide temperature range. [1-6]

We have measured proton spin-lattice relaxation rates (T_1^{-1}) as functions of the external magnetic field in Mn12 and Fe8. When the external magnetic field is applied along the easy-axis (parallel field), spin-lattice relaxation rates, $1/T_1$ decreases monotonically with increasing field. The external field dependence of $1/T_1$ is well explained by a model in terms of thermal fluctuations of the magnetization due to spin-phonon interaction at high temperature range above 1.5K. [6] On the other hand, when the magnetic field is applied perpendicular to the easy-axis (transverse field), we have observed a peak of $1/T_1$, which can not be explained by the model in terms of spin-phonon interaction. We will report details of the peculiar temperature- and external magnetic field-dependences of the spin dynamics in Fe8, and will discuss the origin of the peak of $1/T_1$ in connection with tunneling dynamics under application of the transverse field.

It should be emphasized that the nuclear spin fluctuations can play an important

role for their ground state dynamics though hyperfine coupling as pointed out by recent experimental and theoretical studies. We have performed NMR studies for the Fe8 compounds with enriched isotope of ^{57}Fe at low temperature down to 40mK. $1/T_1$ of ^{57}Fe and ^{1}H is temperature-independent below 400mK. Together with extended results of ^{1}H-and ^{57}Fe-NMR in ^{57}Fe-enriched Fe8 cluster, we will discuss the isotope substitution effects on spin dynamics in the Fe8 clusters.

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