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SPIN ORDERING UNDER PULSE FIELD

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An excited state spin ordering is observed in $NaNiAcac_3$·benzene in the course of pulsed adiabatic magnetization cooling with initial temperatures below 1K. The sharp peak of dM/dt signal corresponding to an antiferromagnetic transition is detected only in the increasing mode of the pulsed magnetic field.

1. Introduction

The idea of cooling by adiabatic magnetization (A. M.) is realized quite analogously to the case of the well known adiabatic demagnetization although there exist some technical difficulties to find out a suitable working substance and to produce a high magnetic field to cover the characteristic field H_c of the substance. In fact, the use of the ground state singlet (G.S.S.) paramagnet of $Cu(NO_3)_2 \cdot 2.5H_2O$ and a 5T superconducting magnet enabled us to observe the first successful cooling and the spin ordering [1] in high magnetic fields around the H_c (36.5kOe) where the lowest excited state and the ground state singlet cross as shown in Fig. 1.

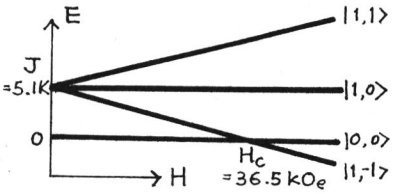

Fig. 1

At the same time, we noticed that the A.M. cooling might be possible using a pulsed magnetic field with the pulse duration shorter than the spin lattice relaxation time. The idea is verified experimentally again in $Cu(NO_3)_2 \cdot 2.5H_2O$ [2] and the result is shown in Fig. 2. The measured magnetization and the critical fields (H_{c1},H_{c2}) are in good agreement with those of the theoretical prediction [3]. By the above mentioned experiments, we recognized the convenience of the method in A.M. cooling.

Actually, we could proceed a lot of investigations on spin ordering in the G.S.S. system such as $CeCl_3 \cdot 7H_2O$, $Ni(NO_3)_2 \cdot xH_2O$ and $CsFeCl_3$, for which the detailed A.M. experiments are performed later with conventional static method. The most important and essential use of the pulse method would, however, for the study of the dynamical aspects in A.M. process.

Here we discuss a unique A.M. process which is possible to be produced only by the use of a pulsed magnetic field and results a transition to new type of ordered phase.

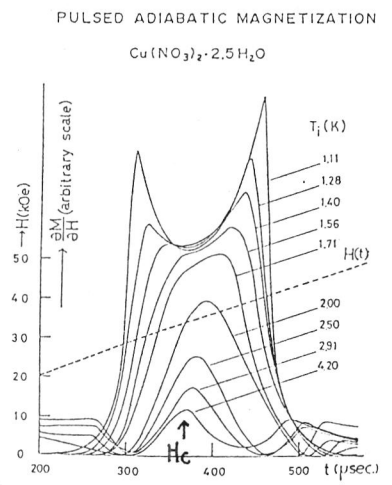

Fig. 2

2. Basic idea of excited state ordering

We consider a simple paramagnetic system in which a magnetic ion is described by the spin Hamiltonian

$$\mathcal{H} = -DS_z^2 + g\beta HS_z \quad (D>0,\ S=1)$$

where D/k is taken as several K for simplicity. The energy level splitting in a magnetic field is shown in Fig. 3. H_e and H_c is defined by $H_e=H_c/3$ and $H_c=D/g\beta$ respectively.

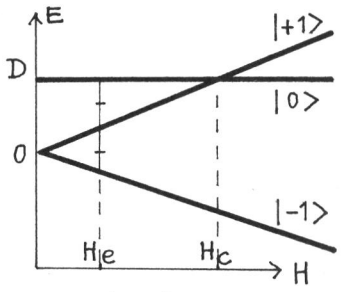

Fig. 3

What will happen at H_c when the system is cooled down to the temperature T_i well below D/k at zero magnetic field and then the pulsed magnetic field is applied? First we discuss the case that the spin-lattice relaxation time τ_{s1} is longer than the pulse duration T/2. Also, the thermal equilibrium within 3 level system is assumed to be attained only at H_e and H_c where the simultaneous spin flips take place easiest giving the time constant τ_{cr} and τ_3 respectively. We assume further that the pulse field passes H_e and H_c with the sweep time $2\cdot\langle H'_{int}\rangle\cdot(dH/dt)^{-1}$ faster than the relaxation times there, where $2\cdot\langle H'_{int}\rangle$ is the effective interaction field. In this case, the spin populations of each spin level is almost unchanged in the course of the field sweep. Therefore, if we look at the population difference between only $|0\rangle$ and $|+1\rangle$, and define a temperature T_{s2} among them, the T_{s2} would be most reduced at H_c. Here we define another spin-spin relaxation time τ_2 among 2 levels, $|0\rangle$ and $|+1\rangle$ in the present case, corresponding to the establishment of the T_{s2} and τ_2 is supposed to be shorter than the sweep time around H_c.

The cooling of the excited 2 level system could be monitored by the magnetization measurement. In the limiting case of $kT_i \ll D$, we may expect the spin ordering at $H=H_c$ which is described by the excited states $|+1\rangle$ and $|0\rangle$. The constant population at $|-1\rangle$ plays a role of 50% magnetic dilution effect.

In the following, we examine numerically the possibility of the experimental realization of the spin ordering at excited state.

3. Experimentals

Ni^{2+} spins in $NaNiAcac_3\cdot$ benzene are described by the Hamiltonian appeared in section 2, where the external magnetic field is applied parallel to the crystal c axis H//c, D=3.15K, g=2.20 and H_c is 21.1kOe. The initial cooling of $kT_i < D/2$ will be attained easily by the use of a conventional He^3 cryostat.

According to the paramagnetic relaxation experiment [4], τ_{s1} is estimated to be about 0.1msec at 4.2K. The 2 spin cross relaxation time τ_{cr} at H_e is temperature dependent and 6 μsec at 1.5K. From the magnetic field dependence of τ_{cr}, we may take the interaction field as $\langle H'_{int}\rangle = \langle H_{int}\rangle/2=150$ Oe, taking the 50% dilution into account.

Now, we need a fast pulsed magnetic field with a sweep rate of $dH/dt \gtrsim 10^9$ Oe/sec to perform the A.M. cooling experiment stated above.

Fig. 4 shows the cryostat and the pulse coil used in the present experiment. The pulse coil produces 40kOe in liq.He^3 with a quarter period T/4 of 10 μsec by a capacitor ($4\sim14$ μF) discharge using a thyratron switch. This results the maximum sweep rate of $dH/dt=5\cdot10^9$ Oe/sec.

The pick up voltage proportional to dM/dt is stored directly in a 100MHz transient recorder and/or a 100MHz memoriscope and recorded by a XY-writer.

The effect of misalignment between the magnetic field and the crystal axis could be studied using an adjustable mechanism by which the tilting of the field axis with respect to the fixed crystal is possible up to ± 3° with fine adjustment within ± 0.5°.

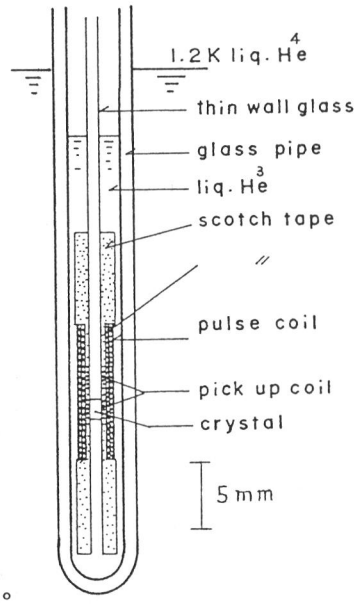

Fig. 4

4. Results

The dM/dt vs H curves around $H=H_c$ are obtained, varying initial temperature T_i from 4.2K to 0.46K. Noteworthy is that the development of the sharp peak is observed at about 1.1kOe lower field than H_c for $T_i < 1.5$K, only in the increasing mode of the pulsed magnetic field. This peak is interpreted to be appeared at the lower critical field H_{c1} of the supposed phase boundary of excited state spin ordering.

To verify above interpretation, the tilting angle (θ) dependence of dM/dt signal is also studied in the case of $T_i=0.63$K. Simple estimation of the maximum angle that allows the phase transition is given by

$$\theta_{max} = 180 \cdot 2g\beta \cdot \langle H'_{int} \rangle / [\pi D \sqrt{2}] \cong 0.6^\circ$$

Experimental results show the abrupt disappearance for $\theta > 0.7^\circ$ as is predected.

In the decreasing mode of $dH/dt < 0$, however, no anomalous peak is observed but only the dM/dt maxima at H_c is observed.

Details of the preliminary experimental data are already appeared in the references [5] and [6].

5. Discussions

The magnetic ordering at ground state in zero magnetic field is expected to take place at around $kT_c = g\beta \cdot \langle H_{int} \rangle = 44$mK. The susceptibility measurement at H=0 was performed by present authors [7] and the transition from paramagnetic to ferrimagnetic state was observed at T=32mK in agreement with the expectation. The ferrimagnetic property is most clearly demonstrated in the stepwize magnetization change from ferrimagnetic to paramagnetic state giving the maximum critical field H_c of 320 Oe at T=0K. This result shows the characteristics of the Ising antiferromagnetic triangular lattice of the crystal.

Thus, we simply expect the ordered state is of antiferromagnetic nature and the ordered temperature is roughly $T_c = g\beta \cdot \langle H_{int} \rangle / 2k = 22$mK for the case of the excited state ordering, taking into account of the 50% magnetic dilution effect by factor 1/2. Then, the phase boundary for a simple antiferromagnetic case should have the field span of $H_{c2} - H_{c1} = 6 \cdot \langle H_{int} \rangle / 2 = 900$ Oe, which is close to the measured value of 1.1kOe. But the upper critical field H_{c2} seems to be not appeared in the experimental results.

The tentative interpretation of the observed hysteresis of dM/dt signal is given by assuming that the spin-spin relaxation time τ_3^{-1} among 3 spin level is very fast and nearly comparable with $\langle H_{int} \rangle \cdot (dH/dt)^{-1}$ at H_c and cause a thermal mixing there.

We are now trying to produce a pulse field with faster sweep rate than, for example, 10^{10} Oe/sec to verify the above interpretation.

References

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INSIDE VIEW OF KANSAI SEMINAR HOUSE (2)

View from the main
conference hall.

View of inner garden
from the main building.