Mossbauer relaxation and thermodynamic properties of spin cluster-triad in EuMg_5

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MÖSSBAUER RELAXATION AND THERMODYNAMIC PROPERTIES OF SPIN CLUSTER-TRIAD IN $EuMg_5$

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The existence of cluster of three-antiferromagnetically coupled spin S=7/2 in the EuMg5 is found to give rise to peculiar behaviour in the thermodynamic as well as the Mössbauer relaxation. The A.C. susceptibility, specific heat and the high field magnetization as well as the ME spectra are reported and interpreted in terms of a model in which the intra-cluster interaction is treated exactly while the inter-cluster interaction is approximated by a molecular field.

1. INTRODUCTION

The intermetallic compound EuMg5 has quite interesting structural and magnetic properties. In fig.1, it can be seen that the structure is composed of cluster of three Eu ions(fig.1-a), which are surrounded in a cage of diamagnetic Mg atoms(fig.1-b). Furthermore,it can also be seen that these isolated clusters (triads) are arranged in planes that are stacked in the C-direction. This fairly uncommon crystal structure is found to give rise to peculiar behaviour as far as the thermodynamics and the hyperfine interactions are concerned. Although the system orders antiferromagnetically (ANFM) at T=7.9K, the following deviations from a normal three dimensional (3d) magnetic behaviour are seen. The susceptibility starts to deviate from a Curie-Weiss (CW) law at $T \simeq 3T_N$ already(fig.2). Also the specific heat (fig.3) and the high field magnetization (fig.4) show also unusual behaviours. In addition, line broadening (fig.5) is detected in the 151Eu Mössbauer effect (ME) spectra (fig.6) from $T \simeq 3T_N$ downwards.

The first attempt to explain the susceptibility behaviour was made by Leuken who introduced a model in which the intra-cluster interaction is treated exactly while the inter-cluster interaction is taken into account with a molecular field (MF) approximation. In the present investigation, the Leuken model will be extended and applied further to specific heat , a.c. susceptibility and the magnetization isotherms at T=4.2K. It will be shown that the triads are not isolated, but interact with one another within the same plane and also between adjacent planes. Further, a ME study was carried out to cover the temperature region where the deviation from the Curie-Weiss law is reported. The ME spectroscopy provides a local microscopic probe, which allows the study of the dynamics of the electronic spins in this region. It will be shown that the MF theory is unable to explain the observed ME spectra at T>7K. Instead , the spectra can rather be interpreted in terms of a slowing down of a relaxation process. The later may arise from rapid spin flips of the Eu++ spins within each triad.

2. EXPERIMENTAL PROCEDURE AND RESULTS.

The magnetization as a function of applied field(fig.4) was measured in pulsed magnetic fields up to 10T. A careful inspection of the data reveals a shallow

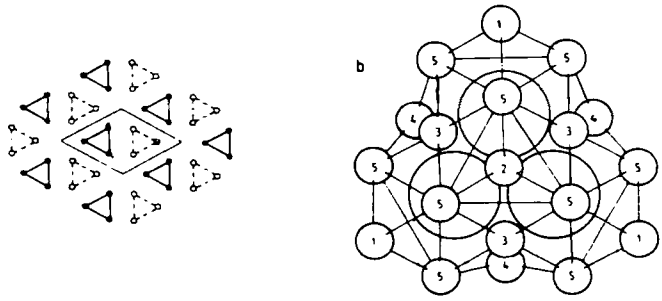

Fig.1-a The arrangement of the Eu-atoms in EuMg5;a projection on the a-b plane○:Eu in c=$\frac{1}{4}$ ● :Eu in$\frac{3}{4}$.The sides of the equilateral triangle is 4.32A.The nearest distance between two Eu atoms from planar neighbouring triangles is 6.09A, while that from different planes is greater than 11.32A Ref./2/.
Fig.1-b Eu triangles in a cage of Mg atoms;◯:Eu-atoms;○ :Mg-atoms.

maximum in the field derivative of the magnetization near 2.5T. This feature cannot be explained by MF theory alone, since the field derivative of the Brillouin function decreases monotonically with increasing the field. The ME spectra are shown in fig.6 below.

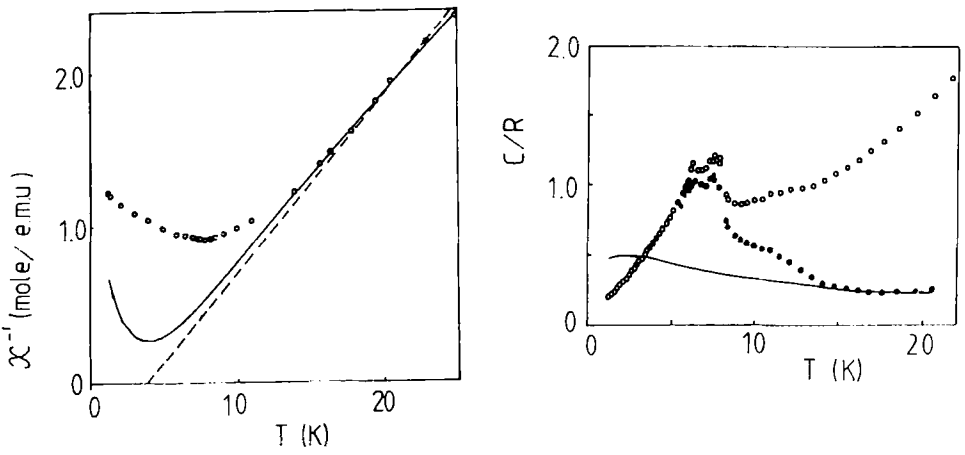

Fig.2(left) Reciprocal molar susceptibility vs temperature.The part of the solid line for T≳10K is a theoretical fit to the data while that below is a calclation using the obtained parameters.The dashed line is the CW behaviour , ($u_{eff}=8.3u_B$).

Fig.3(right) The total specific heat,its magnetic specific heat and the theoretical fit (see text).

At higher temperatures these spectra consist of a single resonance absorbtion line with a slight asymmetry due to a small unresolved quadrupole splitting-which rendered the analysis of the Isomer shift and the line width difficult for $T \gg 3T_N$ Ref/3/. However, for $T \gtrsim 10K$, the ME spectra could be least-squares fitted by using the Lorentzian approximation, with the further assumptions of equivalent sites and a small approximately constant quadrupole splitting. The temperature dependence of the resonance line width (fig.5) shows a broadening starting well above the T_N. Such an anomaly is generally not present in a 3-d magnetic system. Near T=8K, shoulders develop around the spectra-a feature which also cannot be explained by MF theory. In analysing the relaxation effects, we have used the uniaxial relaxation model of Wickmann. Below 7K, the hyperfine field and the relaxation rate were obtained by fitting, whereas above 7K, the hyperfine field was evaluated from graphical interpolation between low temperature ($T < 7K$) and high temperature ($T > 10K$) determination.

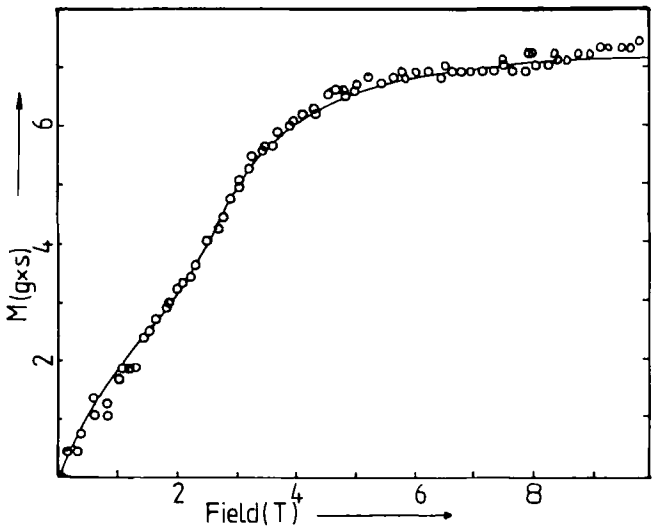

Fig.4 Magnetization as a function of the applied field at T=4.2K The solid line is a theoretical fit to the data.

3. ANALYSIS AND DISCUSSION.

The triangular arrangement of the Eu-atoms and the associated distances suggest that a cluster might be applicable, in which the intra-cluster interaction is described exactly by a Heisenberg type exchange interaction (J). Furthermore, if the inter-cluster interaction are weak, they can be taken into account reasonably well in terms of the molecular field approximation. The Hamiltonian and the resulting thermodynamical properties can be found in Ref./4/. We have attempted to fit our experimental data of the a.c. susceptibility (fig.2), the specific heat (fig.3) and the magnetization isotherm (fig.4) to such a model. The obtained fitting parameters are tabulated in table-1. From our analysis, it follows the intra-cluster interaction is antiferromagnetic. It is well known that an antiferromagnetically coupled spin triad leads to frustration effects and to different although energetically degenerate ground states. This will lead to temperature-dependent flipping of the spins. The ME measures the hyperfine field at the site of the nucleus and the direction of the latter is related to the electronic spin of the individual Eu-ions. Depending on the fluctuation rates relative to the characteristic Eu-nuclear Larmor frequencies, one may expect: fully split spectra at slow rates, sharp nonbroadened

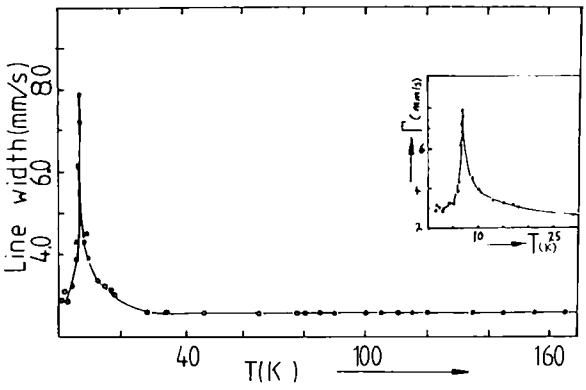

Fig.5(above) Experimental line width vs temperature as obtained from fitting the spectra with lorentzian lines.

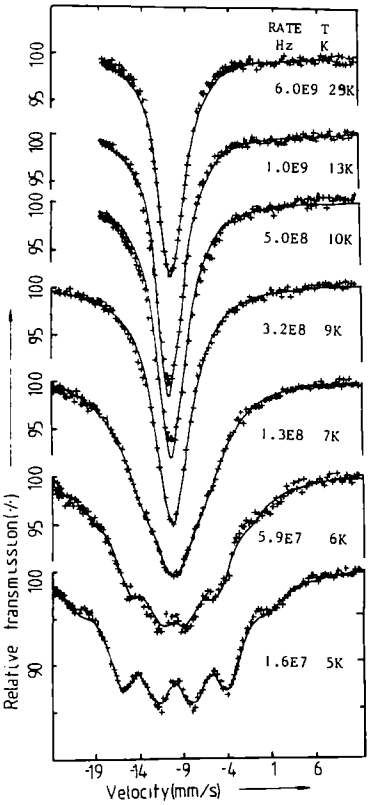

Fig.6(right) 151Eu ME spectra of EuMg5 at different temperatures,fitted with Wickmann's uniaxial relaxation model/5/.

lines at fast rates and highly broadened lines at intermediate rates. This is what is being reflected in the ME spectra of this compound. In fig.6 above the best-computer-fit spectra with the involved relaxation rates are shown.Below 7K the effective hyperfine field could be fitted to a Brillouin function with S=7/2 ($H(T=0)=19.6T$, $T=7.9K$). This reflects the long range interaction character as is shown by the specific heat and the susceptibility data.

Table 1
Results obtained from fitting the, a.c. susceptibility,specific heat and magnetization to the theoretical model(see text).

measurement	J/K(±0.05)	Lambda(mole/emu) ±1.0
Susceptibility	-1.34	5.3
Specific heat (Debye temperature=90K)	-1.19	
Magnetization(T=4.2K)	-1.03	3.3

4. CONCLUSION.

We have shown that the magnetic triad behaviour is reflected in the measured thermodynamical properties and in the ME spectra. At high temperatures, the exchange coupling in the triad is negligable compared to $k_B T$ and the magnetic properties are those of the free Eu-ions. As the temperature is decreased, the higher energy levels of the cluster are depopulated and the magnetic behaviour show a gradual smooth deviation from the free Eu-ions. When the temperature is reduced further, the exchange coupling between the triads establish the 3-d ANFM ordering at T=7.9+0.15 K. The observed ME line broadening for temperatures well above T_N is explained in terms of a slowing down of a relaxation process which is due to the frustration encountered within the antiferromagnetically coupled spin triads. Further analysis and details will be published elsewhere.

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