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AN ELECTRON SPIN RESONANCE STUDY OF SOME INSULATING EUROPIUM CHALCOGENIDE SPIN GLASSES

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Abstract:

ESR in insulating spin glasses is of interest because skin effect and relaxation via conduction electrons are absent. The high temperature line widths have been correctly ascribed to exchange narrowed dipolar broadening (7). Recently Levy and Raghavan have used the Kubo-Tomita formalism to account for the concentration and frequency dependences and the Mori-Kawasaki formalism to account for the temperature dependence of line widths (5). In this report, additional data on $Eu_xSr_{1-x}S$, $Eu_xBa_{1-x}S$ and the mictomagnetic EuS_ySe_{1-y} compounds are presented. Detailed comparison of the line widths with the computed values from the above theory is also made.

1. Introduction:

In the Europium Chalcogenides each Eu^{2+} ion experiences ferromagnetic exchange interactions, J_1, from 12 nearest neighbours and antiferromagnetic exchange interactions, J_2, from 6 next nearest neighbours. As the Eu-X distance increases from EuO to EuTe, J_1 decreases rapidly while J_2 increases gradually. As a result, EuO and EuS are ferromagnetic while EuSe and EuTe are antiferromagnetic in the ordered state. In EuSe $|J_2|$ is only slightly greater than J_1 (2).

The competing magnetic exchange interactions also make it possible to observe 'frustration' effects by dilution of the magnetic ions. Thus the magnetic phase diagram of $Eu_xSr_{1-x}S$ consists of four concentration ranges (3). A similar behaviour is also observed for $Eu_xBa_{1-x}S$ (4).

2. Exchange narrowing of dipolar broadening:

The ESR line shape near the spin glass transition is complicated and, as pointed out by Levy et.al.(5) determined by a 4-spin correlation function in the presence of many body interactions. An analysis of the 'high temperature' line shape is an essential first step since it presents a simpler physical situation. Here the line broadening due to the dipolar interaction (6) and the narrowing due to exchange (7) have to be considered. Depending on the relative magnitudes of the resonance frequency, ω_0, the dipolar frequency, ω_d, and exchange frequency, ω_e, three limiting cases arise as follows:

i. $\omega_0 \gg \omega_d \gg \omega_e$: static dipolar broadening,
line width $= \omega_d$ (1)

ii. $\omega_o \gg \omega_e \gg \omega_d$: line width $= \omega_d^2/\omega_e$ (2)

iii. $\omega_e \gg \omega_o \gg \omega_d$: line width $\simeq 10/3\,(\omega_d^2/\omega_e)$ (3)

Here the factor 10/3 comes from the contributions from satellite transitions at $\omega = \omega_o$, $2\omega_o$ and $3\omega_o$.

In the general case the line width $= c\,\omega_d^2/\omega_e$ where (4)

$c = 1 + (5/3)\exp(-\omega_o^2/\pi\omega_e^2) + (2/3)\exp(-\frac{4\omega_o^2}{\pi\omega_e^2})$ (8). (5)

Here $\omega_d^2 = 3/5\,(g^2\mu_B^2/\hbar^2)\,S(S+1)\langle\sum_j 1/r_{ij}^6\rangle_c$ (6). (6)

where S = 7/2 and $\langle\sum_j 1/r_{ij}^6\rangle_c$ denotes configurational average.

$\omega_e^2 = 4/3\,\langle\sum_j J_{ij}^2\rangle_c\,S(S+1)/\hbar^2$ (9) (7)

$\omega_o = g\mu_B H_o/\hbar$ (8)

For an fcc lattice, $\langle\sum_j 1/r_{ij}^6\rangle_c = 115.2/a^6$ (10) (9)

where a is the lattice constant. For $Eu_xSr_{1-x}S$ and $Eu_xBa_{1-x}S$ the expression on the right hand side has to be multiplied by x assuming a random distribution of magnetic and nonmagnetic ions. Similarly,

$\langle\sum_j J_{ij}^2\rangle_c = 6(2J_1^2 + J_2^2)x$ (10)

3. ESR line width data:

The samples used in these measurements were prepared and characterised at the Ruhr Universitat, Bochum, West Germany. From the considerations of section 2 it can be seen that the high temperature linewidth should be proportional to $x^{1/2}$. Such a dependence has already been observed for $Eu_xSr_{1-x}S$ by Monod et al.(11). In figure 1 the straight lines 'a' and 'b' show our data on $Eu_xSr_{1-x}S$ and $Eu_xBa_{1-x}S$ at room temperature and $Eu_xBa_{1-x}S$ at 77K respectively. At low concentrations one would expect a crossover from $\sqrt{x}$ to x dependence due to a change from limiting case (ii) discussed in section 2 to case (i). For x>o.4, it is seen that the linewidth data satisfy a relation of the form 5: $a + bx^{1/2}$. An increase in linewidth in the case of $Eu_xBa_{1-x}S$ may arise from the phonon modulation of the exchange interaction which is sensitively dependent on the lattice constant and hence on the ionic radius of the nonmagnetic ion (12).

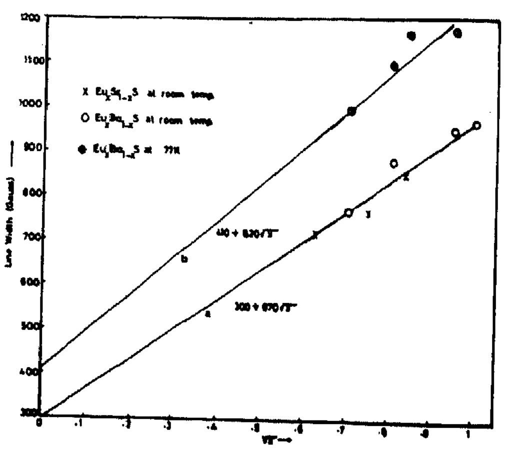

Fig.1. Line width vs. $\sqrt{x}$

The frequency dependence of the line width would also provide a sensitive test of the model. A detailed study of the frequency dependence of EuO, EuS, EuSe and EuTe was carried out by Jansen(8). From his measurements over the frequency range from X band (9.1 GHz) to V band (67.6 GHz), he obtained values for the exchange fields in these chalcogenides. Assuming that the exchange field is proportional to $x^{1/2}$ as above, we determined the corresponding values in $Eu_xSr_{1-x}S$ and $Eu_xBa_{1-x}S$. The factor c in equation (4) may then be determined at Q band relative to the X band for different values of x. A comparison of these values with the ratios of the measured line widths at the two bands in our case as well as from the work of Monod et al. is presented in Table 1. It can be seen that while there is an overall agreement, the agreement becomes poorer at small values of x in general.

Table 1.

x	c (observed)	Exchange field (KOe)	c (calculated from eq. 5)
0.1	3.11	10.7	2.2
0.2	2.71	15.1	2.62
0.4	2.64	21.1	2.92
0.5	2.70	23.8	3.00
0.65	3.18	27.1	3.07
0.7	3.22	28.1	3.09

(c observed was determined assuming that at X band the value was 3.33 and then multiplying it by the ratio of the ESR linewidths at Q and X bands respectively)

The measured ESR line widths at X band at room temperature for EuS_ySe_{1-y} were then used to make a comparison with the values calculated on the above model using the values for J_1 and J_2 for different y given by Westerholt and Bach. It was found that there was no agreement. A diferent method of calculating the line widths from an interpolation of the values of the exchange fields for EuS and EuSe obtained by Jansen was then tried. This is found to lead to a better agreement as can be seen from the data presented in Table 2.

Table 2

y	a (Å)	$(\langle\sum_j J_{ij}^2\rangle_c)^{1/2}$	Calculated line width from eq. 7 (Gauss)	Exchange field from data of ref.8 (KOe)	Calculated line width using data of ref.8 (Gauss)	Measured linewidth (Gauss) (±10 G)
1	5.96	0.8083	--	33.48	--	970
0.5	6.08	0.6288	1106	29.83	966	970
0.3	6.12	0.5853	1154	28.40	976	1000
0.03	6.19	0.4285	1458	26.44	979	930
0	6.19	0.4285	1458	26.22	986	950

4. Conclusions

1. The high temperature ESR line widths in both $Eu_xSr_{1-x}S$ and $Eu_xBa_{1-x}S$ are found to be proportional to $x^{1/2}$ in accordance with the model of exchange narrowed dipolar broadening. However there are deviations at low concentrations due to the small exchange interactions.

2. While the linewidth is temperature independent in $Eu_xSr_{1-x}S$ it increases for $Eu_xBa_{1-x}S$ at 77K, possibly because of phonon modulation of the exchange interactions.

3. The variation of the linewidth in EuS_ySe_{1-y} could also be accounted for if, instead of J_1 and J_2 values, the values of the exchange frequencies obtained for EuS and EuSe from the frequency dependence of the ESR linewidth were used for the calculation.

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