$Eu_xSr_{1-x}S_ySe_{1-y}$: A Model System for Studying Competing Magnetic Interactions

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The exchange-interaction parameters J_1 and J_2 determine the type of magnetic ordering in $Eu_xSr_{1-x}S_ySe_{1-y}$. The ratio $J_2/J_1 = -1$, which holds at a concentration $y = 0.1$, appears as a critical point in the magnetic phase diagram. Close to this point mictomagnetism is observed in samples with Eu concentration high but lower than a critical value.

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In metallic alloy systems with competing antiferromagnetic and ferromagnetic interactions, magnetic short-range order, discussed in terms of spin-glass or mictomagnetism, often occurs at concentrations below the onset of long-range magnetic order.[1,2] Apart from the metallurgical problems,[3] the complexity of the exchange interactions in metals makes it very difficult to work out realistic theoretical models for describing these phenomena. Basically it is the discrepancy between theoretical models and real experimental systems which was the source of many controversial discussions concerning spin-glasses and mictomagnetism in the past. In this situation it is a challenge for experimentalists to find simpler model systems showing similar phenomena. The insulating spin-glass system $Eu_xSr_{1-x}S$ (Ref. 4) certainly is a step in this direction since there are only two exchange interactions present. This simplicity made the theoretical description of the anomalous magnetic properties so successful.[5] Nevertheless $Eu_xSr_{1-x}S$ is far from being completely understood if we consider its complicated dynamics[4] or the magnetic structure below the freezing temperature.[6] We want to present here new mictomagnetic compounds $Eu_xSr_{1-x}S_ySe_{1-y}$ which are, although somewhat more complicated chemically, simpler concerning their magnetic structure.

Details of the preparational method, experiments, and experimental results are given in two previous papers.[7,8] $Eu_xSr_{1-x}S_ySe_{1-y}$ forms a complete NaCl-type solid-solution system; the exchange interaction parameters J_1 and J_2 depend only on y and vary smoothly with the lattice parameter [see Fig. 3(a)].

Figure 1 summarizes the results of the ac susceptibility analysis in a magnetic field of 0.1 Oe at 87 s^{-1} measured on spherical samples. For S concentration $y \geq 0.2$ the samples concentrated in Eu are ferromagnetic; the susceptibility below the ferromagnetic Curie temperature is temperature independent and determined by the demagnetization factor only. Below a certain Eu concentration x the susceptibility decreases from this ferromagnetic saturation value and a rather broad peak with the amplitude and maximum temperature decreasing with x is observed (see Fig. 2 for a few examples). At the same concentration the sharp peak in the magnetic specific heat at the magnetic transition temperature disappears and a rather broad anomaly is observed instead (see Fig. 2 for a few examples). The specific-heat maximum temperature is somewhat higher than the temperature of the ac-susceptibility peak. These findings are typical for spin-glasses and mictomagnets and very similar to $Eu_xSr_{1-x}S$ below the critical concentration $x_c = 0.5$.[4,9] Note that the Eu concentration x where the anomalous type of magnetic ordering occurs first increases with the Se concentration (Fig. 1) and at $y = 0.1$ is observed in the concentrated-Eu compound $x = 1.0$ already!

The samples at the Se-rich side ($y < 0.1$) are

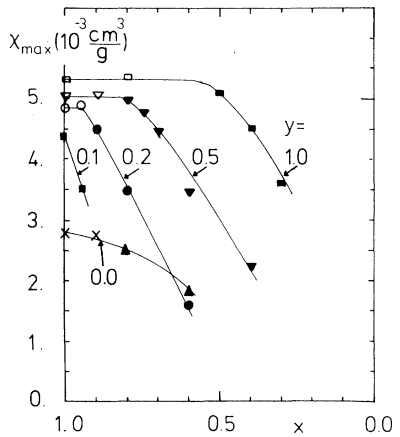

FIG. 1. Magnetic susceptibility of samples $Eu_xSr_{1-x}S_ySe_{1-y}$ as a function of x with y as a parameter. Open symbols: susceptibility below the ferromagnetic Curie temperature; full symbols: value at the susceptibility maximum for the spin-glass samples; crosses: susceptibility value at the Néel temperature.

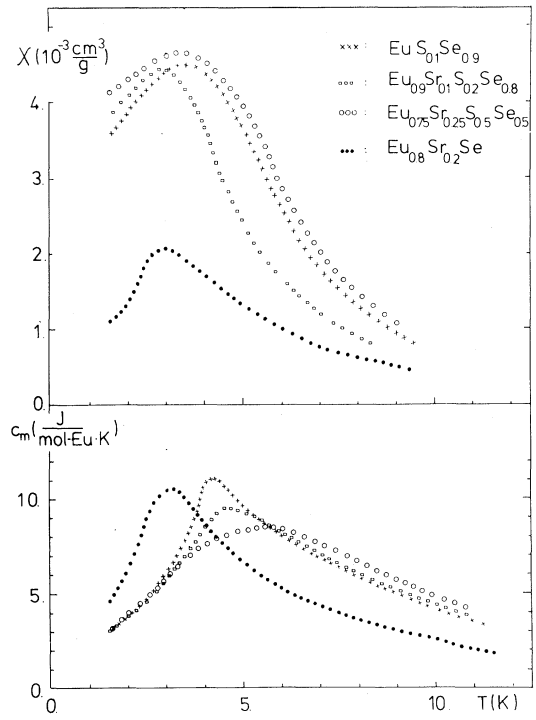

FIG. 2. (Lower) magnetic specific heat and (upper) ac susceptibility as function of temperature for samples $Eu_xSr_{1-x}S_ySe_{1-y}$.

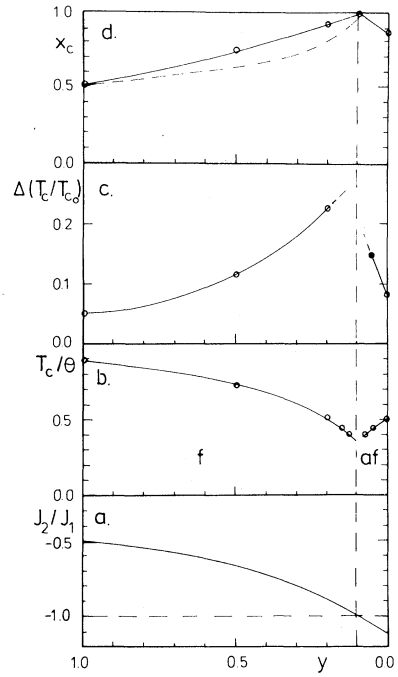

FIG. 3. Magnetic parameters as function of y in EuS_ySe_{1-y}. (a) Ratio of the exchange interaction parameters J_1 and J_2 as derived in Ref. 8. (b) Ferromagnetic Curie temperatures (f) and antiferromagnetic transition temperatures (af) divided by the paramagnetic Curie temperatures. (c) Decrease of the relative magnetic transition temperature by replacing 5% of Eu by Sr. (d) Critical concentration of long-range magnetic ordering. Dashed curve from theory by Binder *et al.* (Ref.5)

antiferromagnetic for $x = 1.0$.[8] On dilution of these samples with Sr, an anomalous type of magnetic specific heat and susceptibility very similar to what is found in the ferromagnetic Sr-diluted samples is measured at fairly high Eu concentrations (Fig. 2).

In Fig. 3 we summarize the essential results we have obtained from the analysis of the Eu_x-$Sr_{1-x}S_ySe_{1-y}$ system. The ratio of the exchange interaction parameters J_1 and J_2 [Fig. 3(a)] determines the other parameters. As J_1 approaches $-J_2$ the ratio of the magnetic transition temperature and the paramagnetic Curie temperature decreases as expected from theory[10,11] [Fig. 3(b)]. The long-range magnetic ordering becomes increasingly sensitive to the solution with Sr [Fig. 3(c)]. Note that the maximum shift of the magnetic ordering temperature in Fig. 3(c) is about five times the value expected from molecular-field theory. The critical concentration for long-range magnetic ordering x_c increases from both sides towards the critical point $J_1 = -J_2$ too [Fig. 3(d)]. The dependence of x_c on the J_2/J_1 ratio can be compared to computer simulation results of Binder *et al.*[5] and reasonable agreement is found [Fig. 3(d)].

The above results show that the anomalous spin-glass state found in $Eu_xSr_{1-x}S$ at $x = 0.5$ shifts towards higher concentrations x when $J_1 \to -J_2$ and finally, for $J_1 = -J_2$, occurs in the sample concentrated in Eu already.

The type of magnetic ordering which should develop at $J_1 = -J_2$ can be predicted easily. Regard the fcc lattice as composed of Eu and (S, Se) planes packed along the (111) direction. Within the (111) Eu planes there is only ferromagnetic exchange, namely, $6J_1$ per Eu ion. Between two adjacent Eu(111) planes, separated by one anion plane, there are $3J_1$ and $3J_2$ per Eu acting, and thus this interplanar coupling is effectively 0 for $J_1 = -J_2$. We conclude that this weakening of the interplanar coupling destroys the long-range magnetic order in our system; it breaks up into two-dimensional ferromagnetic planes coupled to each other by weak dipolar forces and some fluctuating residual exchange field.

It is obvious from this reasoning that the "clus-

ter blocking model" for spin-glasses[12] is an appropriate description for our anomalous magnetic ordering in $EuS_{0.1}Se_{0.9}$. The ferromagnetic, strongly exchange-coupled Eu(111) planes can be regarded as the clusters which are blocked at low temperatures by weak interplanar interactions.

For $J_1 \neq -J_2$ ($y \neq 0.1$) there is an effective exchange coupling between the Eu(111) planes, and the compounds concentrated in Eu show long-range magnetic ordering (antiferromagnetic for $J_2 < -J_1$, ferromagnetic for $J_2 > -J_1$). But nevertheless the interplanar coupling is rather weak for J_2 close to $-J_1$ and it is expected to be most heavily disturbed on introduction of fluctuations of the exchange interactions by replacement of Eu by diamagnetic Sr. Thus we think the magnetic structure of the spin-glass state for the Sr-diluted samples with $J_1 \neq -J_2$ is similar to the one described above.

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Long-Range Surface-Plasma Waves on Very Thin Metal Films

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The dispersion equation of injected surface-plasma waves that propagate on thin metal films has been solved as a function of the film thickness, and splitting of the modes into two branches is observed. For one branch the imaginary part of the propagation constant goes to zero as the thickness of the metal decreases. Reflectivity calculations agree with this result, which predicts that one can obtain propagation distances that are more than 1 order of magnitude larger than observed before.

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The lifetime or decay constant of a surface-plasma wave (SPW) that propagates on a metal surface is an important physical quantity because it is a strong function of the properties of the metal surface, and can therefore serve as a sensitive probe for characterizing surface conditions such as roughness and composition.[1-4] There is only one SPW mode for thick metal films, and the upper limit of its propagation range is determined by the complex refractive index of the metal and the loss introduced by the medium bounding the metal.[5]

The existence of two thermally excited SPW modes, one symmetric and one antisymmetric, that propagate on an unsupported thin metal film (the two media bounding the metal film are identical) has been discussed theoretically and verified experimentally by using electron scattering techniques.[6,7] The theory that predicted the existence of these two modes did not treat the problem of their lifetime and optical-wavelength dependence, and since the resolution and signal-to-noise ratio of the experiments were low, the linewidth of these modes could not be measured.

A comprehensive analysis of the dispersion of SPW that propagate on various combinations of thin metal films sandwiched between thin dielectric films revealed the existence and splitting of the SPW modes as the metal thickness decreases.[8,9] The theory dealt only with the dependence of the real wave vector on the real part of the frequency, and did not treat the properties of the imaginary part of the frequency, which is associated with the lifetime of the thermal SPW.

