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Neutron Spin Echo Study of the Reentrant Spin Glass $Eu_xSr_{1-x}S$

by

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Abstract

Neutron spin echo (NSE) experiments were performed on polycrystalline samples of $Eu_xSr_{1-x}S$ for $x = 0.4$ and 0.54 in the temperature range of $1.2 < T < 10K$ and for $0.036 < Q < .18$ Å^{-1}. The $x = 0.4$ sample exhibits a paramagnetic (PM) to spin glass (SG) transition near $T_f \sim 2K$. In the $x = 0.54$ sample, large ferromagnetic correlations develop below 5K and a SG state appears at lower temperatures. In the NSE experiment, the spin-spin correlation function, $S(Q,t)$, is measured directly for times between $.03 < t < 5$ nsec. At low temperatures both materials exhibit a weak Q dependence in the dynamics and the spins are essentially frozen over the time range explored. On heating the $x = 0.4$ sample the spins start to fluctuate more rapidly, but no dramatic change occurs around T_f. On heating the $x = 0.54$ sample, $S(Q,t)$ decreases rapidly with time. Near the 5K, $S(Q,t)$ is exponential $(e^{-\Gamma t})$ with Γ being strongly Q dependent. Measurements of the depolarization of the scattered beam confirms the absence of true long range ferromagnetic order below T_c.

The presently unsolved spin glass problem has continued to attract a great deal of theoretical and experimental interest. A variation of the conventional spin glass (SG) system is the reentrant spin glass (RSG) which exhibits the following sequence of phase transitions. At high temperatures it is in a paramagnetic (PM) state. On cooling below T_c, extensive ferromagnetic (FM) correlations develop and it appears that a FM state is present. At even lower temperatures, the FM correlations break up and a SG phase is present. The most studied example of this type of magnetic material is $Eu_xSr_{1-x}S$.[1] whose phase diagram is shown in the inset of Fig. 1. The main advantage of this material is that only the first and second nearest neighbor interactions are important and they are of opposite sign. Upon dilution with a nonmagnetic impurity (Sr) the exchange constants remain unchanged and the frustration arises due to the change in the number of magnetic neighbors. The interesting region is near $x = 0.5$ where FM, PM and SG phases are present.

$Eu_{1-x}Sr_xS$ has been extensively studied in recent years by both bulk magnetization[1-3] measurements, and elastic[3,4] and inelastic neutron scattering.[5,6] In the latter measurements, the major response was resolution limited so little information is available about the dynamics of $Eu_{1-x}Sr_xS$. We present below a preliminary report on the results of a neutron spin echo (NSE) study of the spin dynamics of $Eu_xSr_{1-x}S$ for $x = 0.4$ and $x = 0.54$.

The NSE technique and its use in the study of spin glasses has been reviewed previously.[7,8] In this technique the spin correlations are measured directly in the time domain which is the Fourier transform of the spin correlation function (Q,ω), measured in conventional neutron spectroscopy:

$$S(Q,t) = \int_{-\infty}^{\infty} \frac{(Q,\omega)\cos\omega t\, d\omega}{S(Q)} \qquad (1)$$

Here, $S(Q) = \int_{-\infty}^{\infty} (Q,\omega)\, d\omega$. It can be seen above that $S(Q,t=0)=1$. The major advantage of the NSE is the ability to look at much longer times (higher energy resolution) without the loss of intensity inherent in conventional neutron techniques.

The experiments were performed on the IN11 instrument at ILL, Grenoble, France.[9] The incident wavelength of $\lambda = 6.07Å$ ($E_i = 2.2$ meV) with a wavelength spread of 23% was selected by a velocity selector. $S(Q,t)$ was measured for Q values between .036 $Å^{-1}$ and 0.18 $Å^{-1}$ for temperatures between $1.2 < T < 10.0$ K and for times $3 \times 10^{-11} < t < 5 \times 10^{-9}$ sec. The resolution of the instrument was determined by a separate NSE measurement on a CuMn(10%) spin glass ($Tg \sim 50K$) at $T = 1.3K$ where the freezing can be taken as complete, i.e. the magnetic scattering is perfectly elastic.

Polycrystalline samples of $Eu_xSr_{1-x}S$ were studied with $x = 0.4$ and $x = 0.54$. The former exhibits a PM to SG transition at $T_c = 2.0K$ whereas the latter exhibits a double transition: a PM to FM transition near $T_c = 5.0K$ and a FM to SG transition at $T_{SG} = 2.0K$. The spatial correlations in these materials were previously studied with unpolarized neutrons. Figure 1 shows the temperture dependence of the scattered neutrons measured at $Q \sim .10\ Å^{-1}$ using unpolarized neutrons with a triple axis spectrometer set for zero energy transfer. For the $x = 0.4$ sample the intensity increases on decreasing the temperature and levels off at lower temperatures. For $x = 0.54$ two peaks are observed. The higher temperature peak represents a build up of FM correlations and the lower peak, whose position shifts with Q is connected with the break up of the FM correlations and the formation of a SG state.[4,10] There was no depolarization of the beam for the $x = 0.4$ sample but in the $x = 0.54$ sample, the beam partly depolarized near T_c. It became more depolarized on cooling further into the FM phase, but the beam became less depolarized in the SG state. Nevertheless, there was sufficient neutron polarization of the incident beam to perform the experiment at all temperatures.

Figure 2 shows the NSE results for the $x = 0.4$ sample. $S(Q,t)$ is plotted vs lnt since the data was taken for over two decades of time. For reference, the exponential decay of $S(Q,t) \sim e^{-\Gamma t}$ (a Lorentzian in frequency space) is shown with $\Gamma = 33$ μeV corresponding to time $\Gamma^{-1} = 2 \times 10^{-11}$ sec. In Fig. 2a, $S(Q,t)$ is shown for $T = 1.33 < T_g$ for two different Q values. The data is consistent with a straight line which corresponds to

$$S(Q,t) \sim A \ln t. \tag{2}$$

This can be seen to be much slower than the exponential rate ($\ell^{-\Gamma t}$) with a single relaxation rate. One could also fit the data with a distribution of decay rates

$$S(Q,t) = \int f(\Gamma)\, e^{-\Gamma t} d\Gamma \tag{3}$$

where $f(\Gamma)$ is the distribution function of decay rates. Another observation is that, within error, the data exhibits little Q dependence. In Fig. 2b, the temperature dependence of $S(Q,t)$ is shown for $Q = 0.072$ Å^{-1}. $S(Q,t)$ decays more rapidly with time as T increases. These results are very similar to the NSE measurements on CuMn[8].

The results on the $x = 0.54$ sample, which exhibits a reentrant spin glass behavior, is shown in Fig. 3. Fig. 3a shows $S(Q,t)$ at $T < T_g$ for two different Q values. The behavior looks similar to the $x = 0.4$ sample. In Fig. 3b, $S(Q,t)$ is shown for several temperatures. The most important observation is that the decay rate starts off as a straight line and then at higher temperatures looks like an exponential. Fig. 4 shows the Q dependence of $S(Q,t)$ at T = 5.5K. The behavior closely approximates an exponential behavior with Γ increasing as Q increases.

We presented above the first NSE study of a reentrant spin glass $Eu_xSr_{1-x}S$. At low temperatures the $x = 0.4$ and 0.54 sample exhibit behavior that is similar to the NSE observation on the typical SG, CuMn.[8] The decay of $S(Q,t)$ is much slower than $e^{-\Gamma t}$ and nearly Q independent. Ordered magnetic structures would exhibit an oscillatory behavior of $S(Q,t)$ due to spin waves. Also, relaxational effects in ordered magnets exhibit a strong Q dependence of relaxational processes. The Q independent behavior suggests a lack of kinematical slowing down.[8,11] which implies that spin non-conserving forces are present, e.g. spin-orbit or dipole interactions play an important role. The data is consistent with the behavior, $S(Q,t) \sim A \ln t$ or an exponential decay with a distribution of decay rates. As T increases, $S(Q,t)$ for $x = 0.4$ decays more rapidly but again very similar to typical spin glasses. For $x = 0.54$ a more interesting behavior occurs. On heating above $T_{SG} = 2°K$ into the FM regime, $S(Q,t)$ decays more rapidly but does not exhibit any oscillatory behavior implying spin waves within the energy window of .15-20 μeV that we explored. Furthermore, the fact that $S(Q,t)=1$ at small times implies that if there are higher energy spin waves excitations, their relative weight is small compared with that of the spin relaxation processes we observed. At high temperatures, $S(Q,t)$ changes into an exponential form of $e^{-\Gamma t}$ with Γ increasing with T and Q. At T = 5.5K the decay rate is consistent with the behavior $\Gamma = DQ^2$ where $D \sim 200$ μeV-Å^2.

We can understand the observed dynamical behavior on the basis of the model we proposed earlier for $Eu_xSr_{1-x}S$.[4,10] On cooling from the PM phase, ferromagnetic correlations develop with long correlation lengths ($\xi > 400$ Å) but no long range order.[10] This is evidenced by the partial depolarization of the beam below T_c. The $\Gamma = DQ^2$ behavior observed in Fig. 4 is the collective behavior of the spins within the correlated regions. Below T_c, no oscillatory behavior is observed indicating the absence of spin waves. $S(Q,t)$ deviates from a simple exponential low and takes on a lnt behavior or an exponential behavior with distribution of relaxation rates. From diffraction measurements[10] it is known that below T_c, the FM correlations break up into smaller regions. The NSE results show that the fluctuations in these regions get slower and eventually freeze at low temperatures just like in a conventional spin glass as exemplified by x = 0.4 sample.

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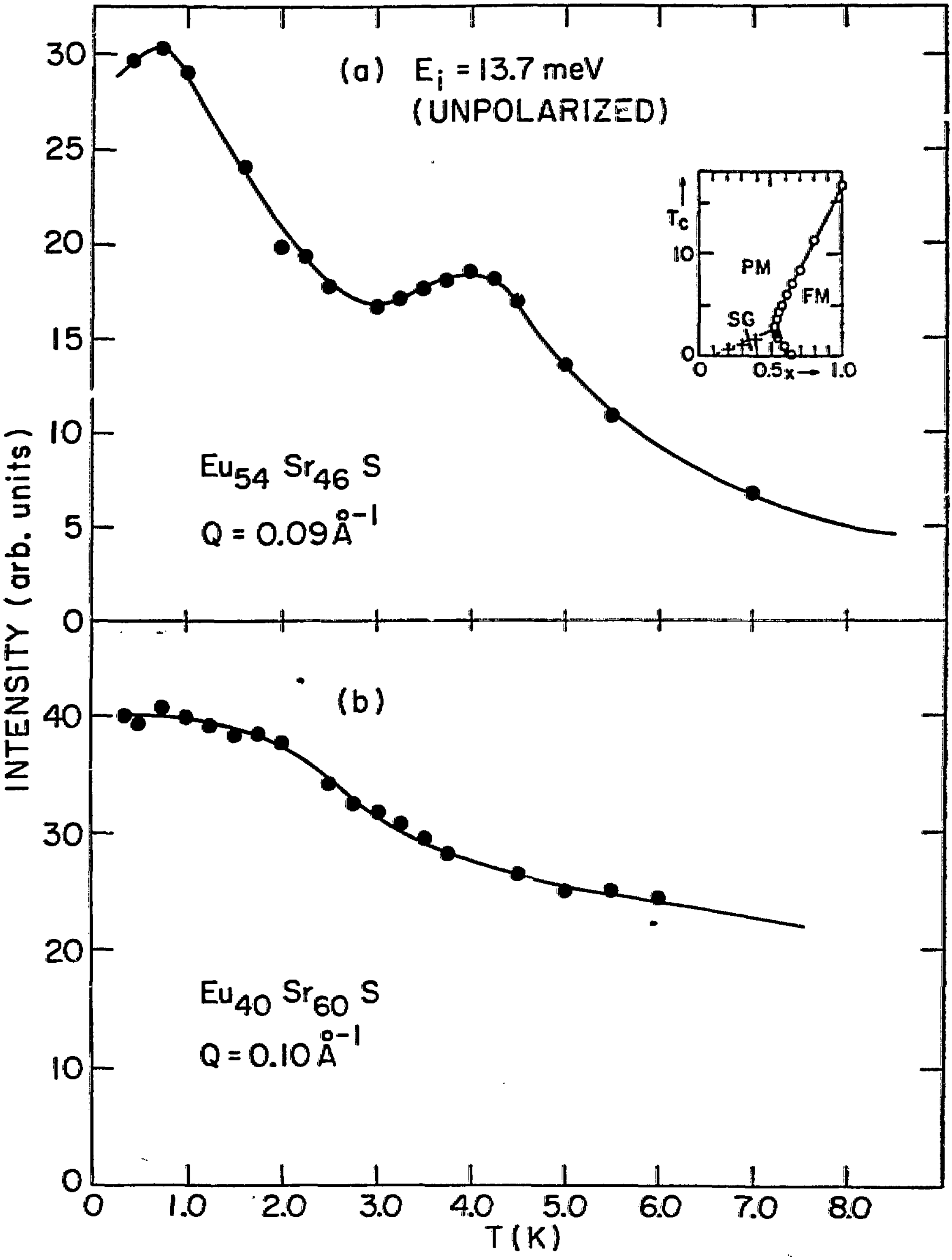
Figure Captions

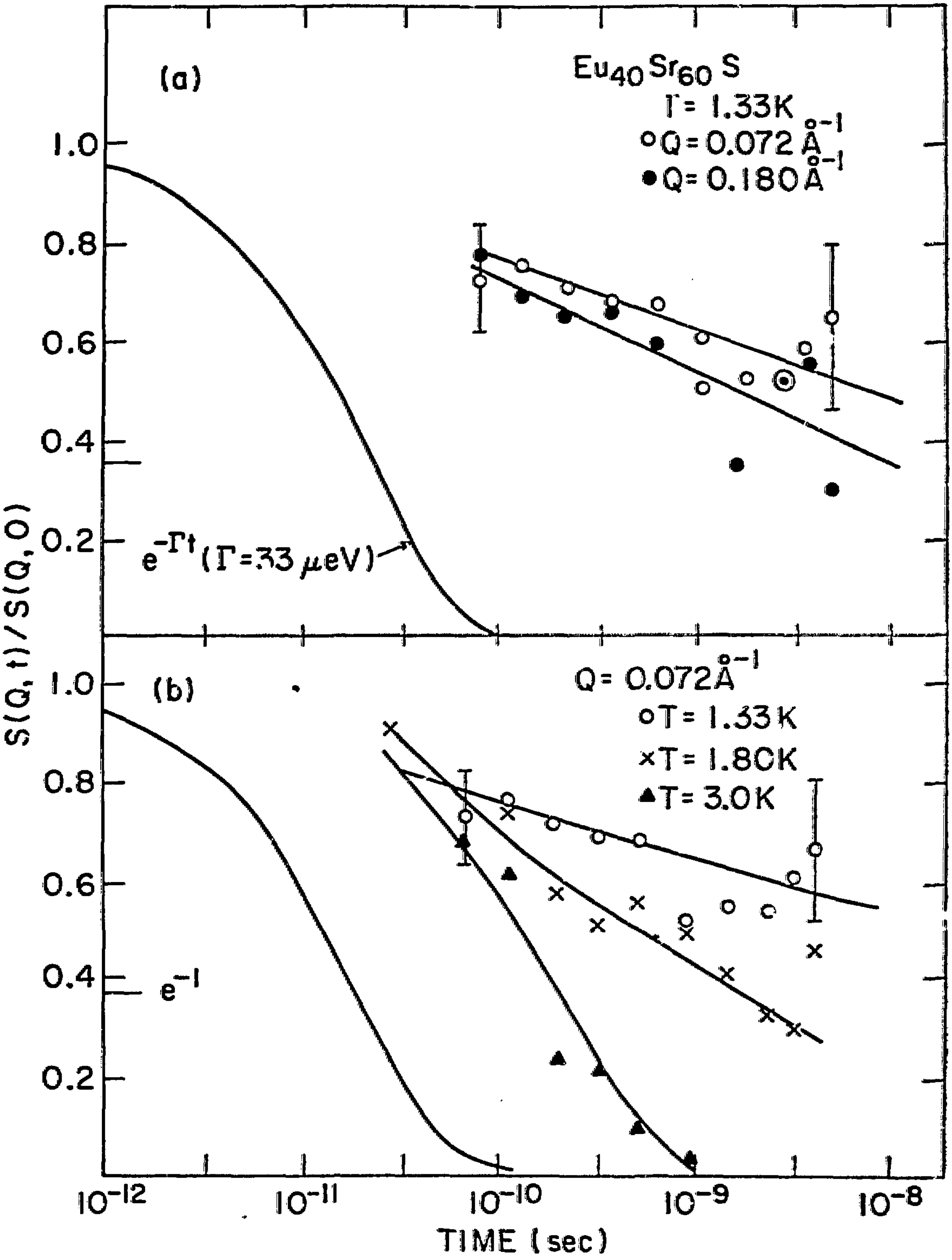
Fig. 1 Temperature dependence of scattered intensity of unpolarized neutrons for (a) $x = 0.54$ at $Q = .09$ Å^{-1} and (b) $x = 0.4$ at $Q = .10$ Å^{-1}. The inset shows the phase diagram of $Eu_xSr_{1-x}S$ from ref. 3.

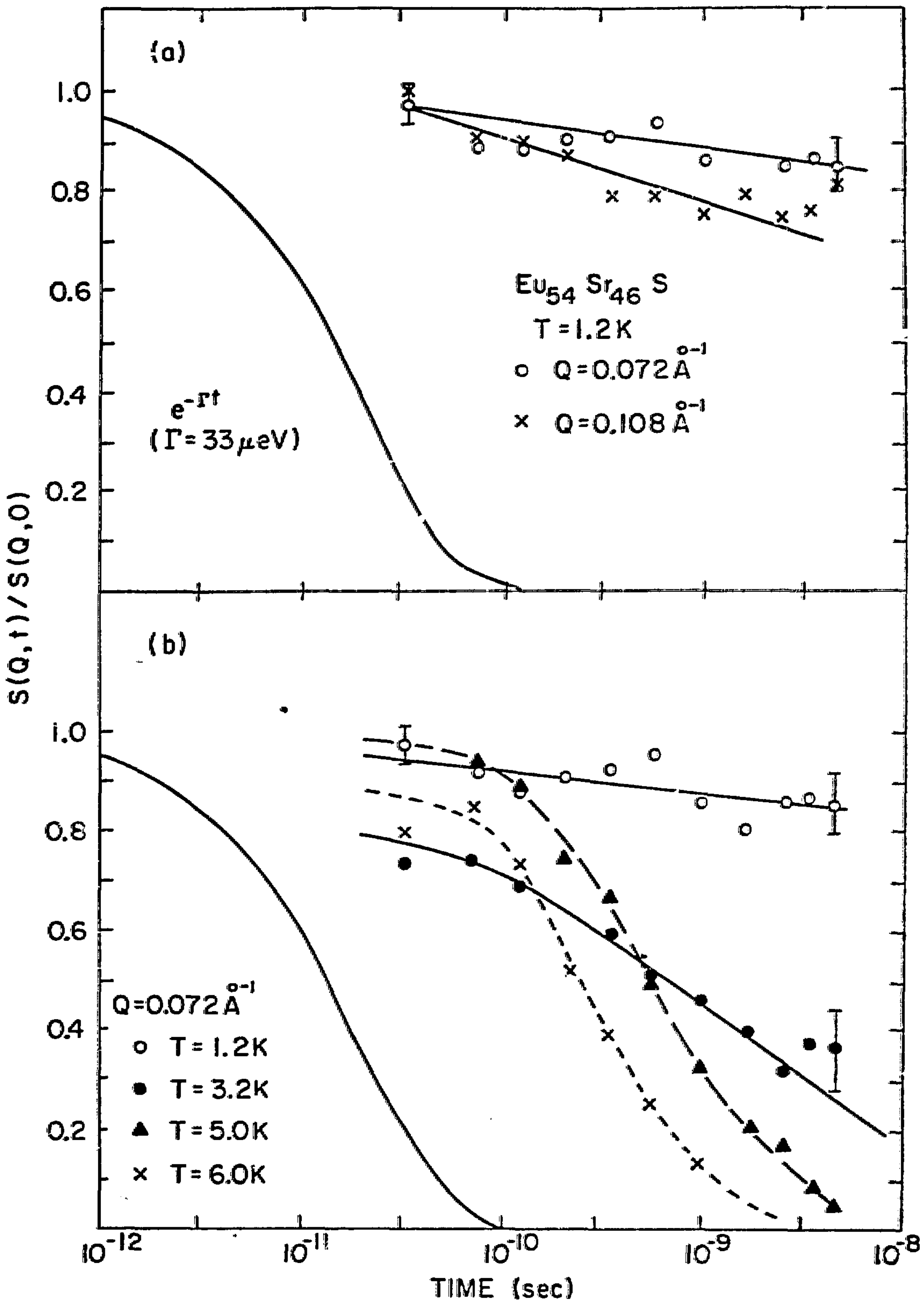
Fig. 2 Time correlation function $S(Q,t)/S(Q,0)$ for $Eu_{40}Sr_{60}S$. (a) Q dependence at $T = 1.33K$, below $T_g = 2.0$. (b) T dependence at $Q = .072$ Å^{-1}.

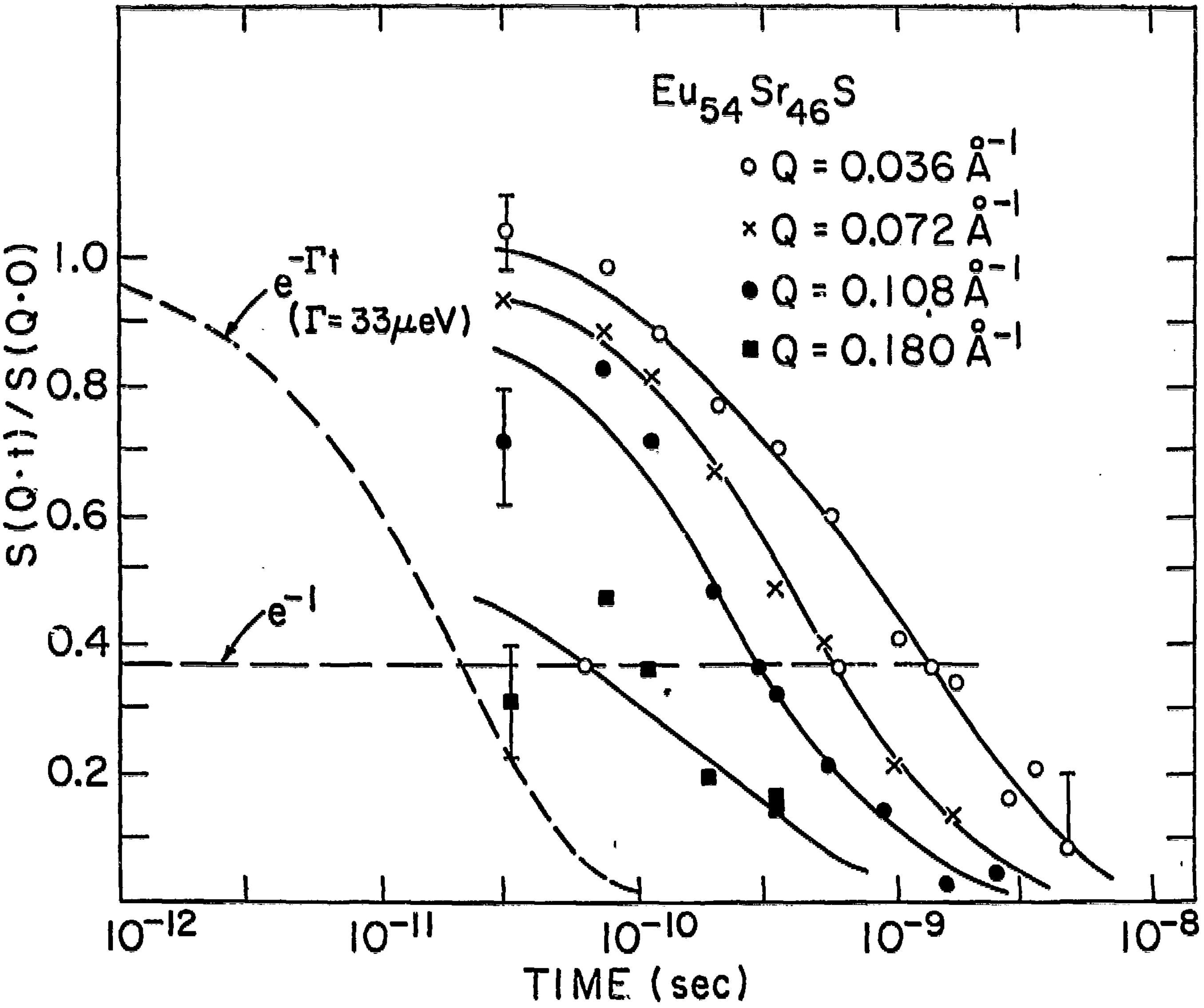
Fig. 3 Time correlation function $S(Q,t)/S(Q,0)$ for $Eu_{54}Sr_{46}S$. (a) Q dependence at $T = 1.2K$ below $T_g = 2.0K$ (b) T dependence for $Q = 0.72$ Å^{-1}.

Fig. 4 Q dependence of the time correlation function $S(Q,t)/S(Q,0)$ for $Eu_{54}Sr_{46}S$ measured at $T = 5.5K$.


(a) E_i = 13.7 meV
(UNPOLARIZED)
$Eu_{54}Sr_{46}S$
Q = 0.09 Å^{-1}
T_c
10
PM
FM
SG
0
0
0.5 x →
1.0
(b)
$Eu_{40}Sr_{60}S$
Q = 0.10 Å^{-1}
INTENSITY (arb. units)
30
25
20
15
10
5
0
40
30
20
10
0
0
1.0
2.0
3.0
4.0
5.0
6.0
7.0
8.0
T (K)


(a)
$Eu_{40}Sr_{60}S$
T= 1.33K
○Q=0.072 Å^{-1}
●Q= 0.180 Å^{-1}
$e^{-\Gamma t}$ (Γ=33 μeV)
(b)
Q= 0.072Å^{-1}
○T= 1.33K
×T= 1.80K
▲T= 3.0K
e^{-1}
S(Q,t)/S(Q,0)
1.0
0.8
0.6
0.4
0.2
10^{-12}
10^{-11}
10^{-10}
10^{-9}
10^{-8}
TIME (sec)


(a)
1.0
0.8
0.6
0.4
0.2
$Eu_{54}Sr_{46}S$
T = 1.2 K
○ Q = 0.072 Å⁻¹
× Q = 0.108 Å⁻¹
$e^{-\Gamma t}$
(Γ = 33 μeV)
S(Q, t) / S(Q, 0)
(b)
1.0
0.8
0.6
0.4
0.2
Q = 0.072 Å⁻¹
○ T = 1.2 K
● T = 3.2 K
▲ T = 5.0 K
× T = 6.0 K
10^{-12}
10^{-11}
10^{-10}
10^{-9}
10^{-8}
TIME (sec)



$Eu_{54}Sr_{46}S$
○ Q = 0.036 Å^{-1}
× Q = 0.072 Å^{-1}
● Q = 0.108 Å^{-1}
■ Q = 0.180 Å^{-1}
$e^{-\Gamma t}$
(Γ = 33 μeV)
e^{-1}
S(Q·t)/S(Q·0)
1.0
0.8
0.6
0.4
0.2
10^{-12}
10^{-11}
10^{-10}
10^{-9}
10^{-8}
TIME (sec)