Cluster dynamics in Au-15%Fe alloy: neutron spin echo studies

1990 J. Phys.: Condens. Matter 2 7873

(http://iopscience.iop.org/0953-8984/2/38/014)

J. Phys.: Condens. Matter 2 (1990) 7873–7876. Printed in the UK

LETTER TO THE EDITOR

Cluster dynamics in Au–15%Fe alloy: neutron spin echo studies

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Received 21 June 1990

Abstract. Neutron spin echo (NSE) measurements have been carried out on the **Au**–15%Fe alloy within the temperature range $1.5 \leqslant T \leqslant 39$ K and for a Q-value of 0.04 Å^{-1}. The results show that the cluster relaxations have a Kohlrausch 'stretched' exponential of form $\exp(-(t/\tau)^{\beta})$, like the dynamics of the glassy relaxation. The macroscopic relaxation time τ deduced from the Kohlrausch relaxation grows as T is reduced to T_f in a manner similar to an Arrhenius-type thermally activated dynamics.

In some earlier work (Sarkissian 1990) I showed that interesting developments in the magnetic character of the **Au**–15%Fe alloy with changes in temperature can be obtained in neutron spin depolarization studies. In this alloy, the depolarization observed below the temperature T^* (90 K) is caused by the dominant magnetic effects of the non-divergently large finite clusters. The depolarization remains large up to temperatures close to 50 K, where it drops rapidly to values close to zero, because the large clusters break up into smaller clusters (containing a few tens of spins), which then freeze into spin-glass-like behaviour below $T_f = 25$ K.

In the present letter, we report the results of a study of the dynamic behaviour associated with the freezing process of clusters in the **Au**–15%Fe alloy obtained by NSE spectroscopy. In this method, the real-time dynamic scattering cross-section $S(Q, t)$, where Q is the associated neutron momentum transfer and t is the time, is measured as a function of t using the Larmor precession technique.

The measurements were performed using the IN11 NSE spectrometer at the high-flux reactor of the Institut Laue–Langevin, Grenoble, France. Neutrons of wavelength 5.2 Å were selected by a helical velocity selector with FWHM resolution of $\simeq 18\%$ in wavelength. Measurements were carried out on the same specimen as was used in early neutron spin depolarization studies (Sarkissian 1990). In this study, the values of the cross-section for the static structure factor $S(Q, 0)$ were deduced from three-dimensional polarization analysis data The analysis proceeded in the same manner as in previous NSE studies (Mezei and Murani 1979), and a description will not be given here.

The $S(Q, t)$ data normalized to $S(Q, 0)$ obtained in time domains of 4×10^{-11} to 4×10^{-9} s over a wide temperature range for Q-values of 0.04 Å^{-1} are shown in figure 1. The features of our measurements are as follows.

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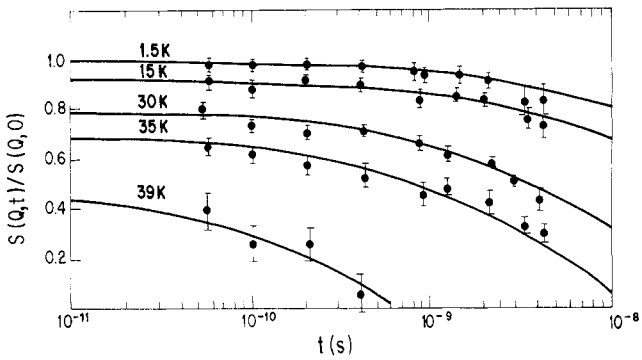

Figure 1. The time correlation function $S(Q,t)/S(Q,0)$ for the **Au**–15%Fe alloy at $Q = 0.04$ Å^{-1}. The full curves are fits to (1) described in the text.

Table 1. Fitting parameters for various temperatures, described in the text.

T (K)	φ_F	τ (s)
39	0.55	$(1.2 \mp 0.4) \times 10^{-9}$
35	0.30	$(1.0 \mp 0.2) \times 10^{-8}$
30	0.20	$(2.0 \mp 1.0) \times 10^{-8}$
15	0.08	$(8.0 \mp 2.0) \times 10^{-8}$
1.5	≈ 0	$(1.4 \mp 0.2) \times 10^{-7}$

(i) It is very significant that the initial values (at 4×10^{-11} s) of $S(Q,t)/S(Q,0)$ are smaller than unity for temperatures above 1.5 K. Extrapolation of the data obtained between 4×10^{-11} and 4×10^{-9} s to $t = 0$ suggests the existence of a fast initial dynamic component of spectral weight φ_F which decreases rapidly to zero as T reduces to zero. This component presumably relaxes on a microscopic timescale $\ll 10^{-11}$ s, which is too short to be detected with the experimental time domain limit of the present experiment.

(ii) There are pronounced changes in $S(Q,t)/S(Q,0)$ as T is decreased through T_f due to a slow-time-decay component of a spectral weight φ_S. We found that this component can be described by the Kohlrausch 'stretched' exponential

$$\varphi_S = S(Q,t)/S(Q,0) - \varphi_F = \exp(-(t/\tau)^{\beta}) \tag{1}$$

with three adjustable parameters, the spectral weight of the fast component φ_F, the time-stretched exponent β, and the macroscopic relaxation time τ. The exponent β is independent of temperature throughout the spin freezing process, whereas τ and φ_F can be chosen to be temperature-dependent constants. The full curves of figure 1 represent the fits to equation (1) with $\beta = 0.6$, and other T-dependent parameters are listed in table 1; these indicate that τ increases by over three orders of magnitude in the T-region 39 to 1.5 K. The plot of ln τ versus $1/T$ for temperatures above T_f (figure 2) is almost linear, showing an Arrhenius-type thermally activated dynamics, $\tau = \tau_0 \exp(-E_0/kT)$, with an activation energy of $E_0 \simeq 33$ meV and a microscopic relaxation time of $\tau_0 = 10^{-13}$ s.

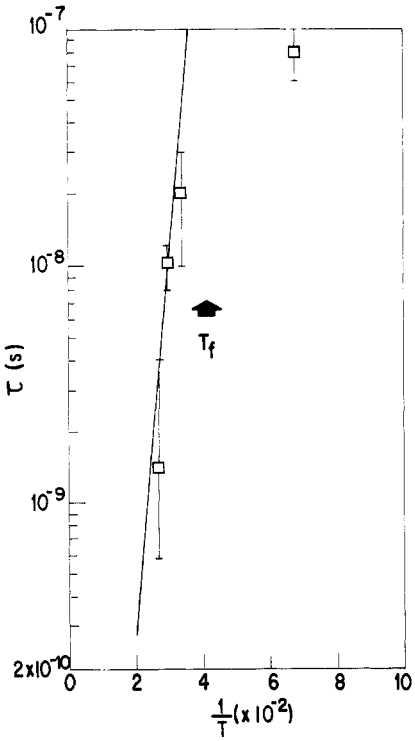

Figure 2. A semilogarithmic plot of ln τ versus $1/T$; the straight line shows the Arrhenius-type thermally activated dynamics above T_f.

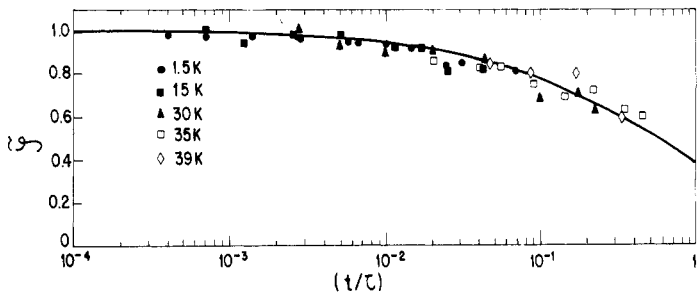

Figure 3. Plots of $\tilde{\varphi}$ versus the reduced time t/τ for various temperatures. The full curve represents $\tilde{\varphi} = \exp(-(t/\tau)^{\beta})$, with $\beta = 0.6$. Certain points are not plotted, to avoid overlapping.

To illustrate the validity of our analysis, we have replotted the results of figure 1 in terms of the quantity $\tilde{\varphi}$ $(=\varphi_S + \varphi_F)$ versus the reduced time (t/τ) on a semilogarithmic plot in figure 3; the data can be described by a single curve $\tilde{\varphi} = \exp(-(t/\tau)^{\beta})$; the fit shows that the Kohlrausch anomalous relaxation law describes the dynamics of the clusters for temperatures above and below T_f. Our analysis indicates the presence of a single macroscopic relaxation time τ, which determines the overall slow-time-decay behaviour of the dynamical processes and grows rapidly as T is reduced to zero. The slow-time-decay behaviour is a general phenomenon of dynamics associated with spin

freezing; such behaviour has also been observed in other spin glasses, such as **Cu**–Mn (Mezei and Murani 1979).

It is known that the dielectric relaxations in a non-ergodic glassy material can be well described by a Kohlrausch stretched exponential relation (Jonscher 1977); such a relation is also found to describe the time dependence of the remnant magnetization in spin glasses (Chamberlin *et al* 1984), and to provide a fit to the glassy relaxations observed in NSE studies (Mezei *et al* 1987). There has been considerable theoretical interest in stretched exponential time behaviour, similar to that described by (1). Certain features of theoretical work should be mentioned that show some correspondence with the cluster dynamics in **Au**–15%Fe presented above: an interesting approach to describing the stretched exponential time behaviour is given by the cooperative relaxation theory of Ngai (1980). In this model the deviation from the simple exponential time behaviour is the consequence of the strong coupling between the microstates and the environment. The Ngai model seems to characterize the time dependence of $S(Q, t)/S(Q, 0)$; however, a significant inconsistency still exists since Ngai's theory is valid only for long times, which are never achieved in our experiments. An alternative approach to the stretched exponential time behaviour is the hierarchically constrained dynamic scheme of Palmer *et al* (1984). In this scheme, Kohlrausch relaxation is the result of successive dynamic constraints over many energy barriers associated with cluster size distribution. In this picture, the smaller fast clusters will constrain the large slow clusters over many time-scales (the relaxation time for a cluster increases with the cluster size). A natural consequence of this model is that clusters of different sizes freeze at different temperatures; the model thus describes the emergence of a continuous distribution of the relaxation times with respect to the temperatures. Palmer *et al* (1984) find that the characteristic time obtained from a Kohlrausch law diverges in the Vogel–Fulcher manner. Although this interpretation seems to have close connections with our experimental results, the temperature evolution of the characteristic time τ deduced from our data corresponds more closely to an Arrhenius-type thermally activated dynamics.

To summarize, our results show that the cluster relaxations in **Au**–15%Fe alloy have the Kohlrausch stretched exponential form $\exp(-(t/\tau)^{\beta})$, like the dynamics of the glassy relaxation. The macroscopic relaxation time τ deduced from the Kohlrausch law grows as T is reduced to T_f in a manner similar to an Arrhenius-type thermally activated dynamics.

The author wishes to thank A P Murani for making available the specimen used in this work, and M A Continentino for valuable discussions.

References

Chamberlin R V, Mozurkewich G and Orbach R 1984 *Phys. Rev. Lett.* **52** 867
Jonscher A K 1977 *Nature* **267** 673
Mezei F, Knaak W and Farago B 1987 *Phys. Rev. Lett.* **58** 571
Mezei F and Murani A P 1979 *J. Magn. Magn. Mater.* **14** 211
Ngai K L 1980 *Comment. Solid State Phys.* **9** 141
Palmer R G, Stein D L, Abrahams E and Anderson P W 1984 *Phys. Rev. Lett.* **53** 958
Sarkissian B V B 1990 *J. Phys.: Condens. Matter* submitted