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Reexamination of the small-angle neutron scattering data on concentrated *Au*Fe spin-glasses

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The recent reinterpretation by Soukoulis, Grest, and Levin of the small-angle neutron scattering data on *Au*Fe alloys is criticized. It is argued that their analysis of the experimental data creates (and their theory accounts for) an additional anomaly not observed experimentally. Adequate theoretical explanation of the original q-dependent peaks in the small-angle scattering cross section of the alloys may still be lacking.

Small-angle neutron scattering measurements on *Au*Fe alloys containing between 10 and 15 at.% Fe (Ref. 1) show a series of q-dependent peaks (maxima) in the total scattering intensity at temperatures which appear to extrapolate smoothly as $q \rightarrow 0$ towards T_{sg}, the temperature of the maximum in the ac susceptibility. These results were interpreted by the present author as showing a series of q-dependent freezing temperatures.

Recently Soukoulis, Grest, and Levin[2] (hereafter referred to as SGL) have put forward a different interpretation of these results. They show that the subtraction from the measured total scattering cross section of a suitably chosen static component which decreases monotonically from its maximum value at $T = 0$ K and goes sharply to zero at a unique ordering temperature T_{sg} (for all q values) can leave the residue, i.e., the dynamic part (or the integrated frequency-dependent component)[3] with peaks at the same temperature T_{sg}, for all q values, and thus argue that the experimental results on the *Au*Fe alloys are not inconsistent with a unique ordering temperature T_{sg}. They have attempted to justify the above form of the static structure factor by means of a theory of the neutron scattering cross section in the mean-field random-phase approximation using the Edwards-Anderson-type Hamiltonian[4] for a system of well defined magnetic clusters with ferromagnetic intracluster interactions.

In the following it is pointed out that the effect of the subtraction of the static part of the cross section from the total cross sections as done by SGL, should not be misunderstood as causing the shift of the original q-dependent peaks in the total intensity to a unique temperature T_{sg} (for all q values) in the resultant dynamic part.

(a) In actual fact the resultant sharp peaks in the latter merely reflect, and are the direct result of, the sharp discontinuity at T_{sg} in the assumed form of the static structure factor. It is useful therefore to recognize that the resulting q-independent peaks in the dynamic component are "artificially" created additional anomalies in as much as they are assumed to be there to begin with. Furthermore, as demonstrated in Fig. 1 they bear no direct connection to the experimentally observed q-dependent peaks since they are obtained even if there were no peaks originally in the total cross section.

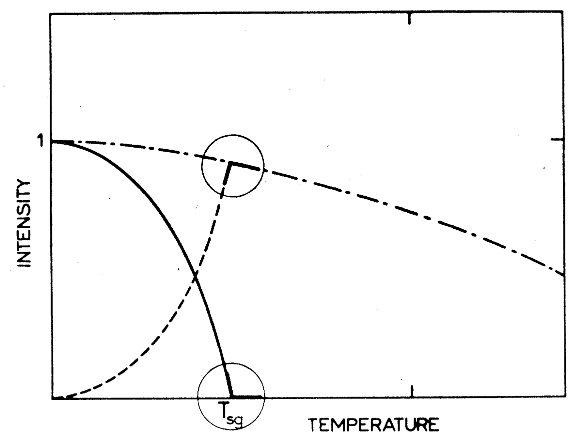

FIG. 1. Schematic variation of the intensity with temperature for one q value. The chain curve shows a possible monotonic form for the total measured intensity (note: no sharp peak originally). The solid curve represents the static (or Bragg) contribution with its sharp discontinuity at T_{sg} which is circled. The resultant dynamic component is given by the dashed curve together with the chain curve (above T_{sg}). It is clear that the sharp peak formed at the junction of the two curves (circled) is the direct results of the assumed sharp discontinuity at T_{sg} (also circled) in the static component. If T_{sg} is assumed to be the same for all q values, it is evident that the dynamic component [proportional to $\chi(q,T) \times T$] will have sharp maxima at the same temperature T_{sg} for all q (in addition to any other sharp features present in the total intensity curves).

(b) It is important to note that after subtraction of a smoothly varying static component we can still be left with a shoulder in the resultant dynamic structure factor with a relatively sharp change of slope at the original position of the peak in the total cross section. It is evident that the sharp change of slope observed in the total intensity cannot simply disappear through subtraction of a function which varies smoothly over the temperature range of the anomaly. It can, however, be made less noticeable by subtraction of a suitably chosen static component with a structure complementary to that of the peak or with a relatively rapid temperature variation in the region of the peak.

(c) In this respect the weakly q-dependent maxima in the total intensity computed by SGL are much broader compared with the relatively sharp peaks observed experimentally, and these maxima in the calculated curves always lie below T_{sg} even for $q=0$. (If the model parameters are adjusted such that the maximum in the total intensity coincides with T_{sg} at $q=0$ then there is no q dependence at all in the temperatures of the maxima.)

(d) Finally, SGL's calculated total intensity shows a sharp change of slope at T_{sg}. This is a natural result of the mean-field approximation and might reasonably be expected also in a more sophisticated analysis involving a true phase transition at T_{sg}. It is emphasized that no evidence of such a change of slope at T_{sg} is present in the experimental results.

In Fig. 2 the data for a Au-13 at.% Fe alloy are shown over a small range of low q values where the scattering intensity is higher resulting in a better statistical accuracy of the data points. These are shown as triangles whose size is of the order of or larger than the statistical errors ($\sigma=\sqrt{N}$). Two different forms of the static structure factor have been assumed in treatment of the data. These are

$$I(q,T)=\begin{cases} I_0(q)(1-T/T_{sg})^{\beta} & \text{(1a)} \\ I_0(q)[1-(T/T_{sg})^{\beta'}] \ , & \text{(1b)} \end{cases}$$

where $I_0(q)$ is the scattering intensity for $q \rightarrow 0$ and $T_{sg}=40$ K is the temperature of the ac susceptibility peak for this concentration taken from the literature.[5] The values of β and β' were chosen arbitrarily as $\frac{1}{3}$ and 3 in order to simulate the forms for the static structure factor taken by SGL in their analysis. Subtraction of the static component from the total intensity yields data points for the dynamic component which are shown by the round points (below T_{sg}). A dashed curve is drawn through these as well as the data points above T_{sg} to identify the total dynamic component. It is apparent that the sharp maxima at a unique temperature T_{sg} in the dynamic structure factor indicated by the down-pointing arrows are the direct result of the assumed form of the static component with its sharp discontinuity at T_{sg}, whereas no such anomaly is present initially in the measured total intensity. In the diagram the up-pointing arrows mark the positions of the original anomalies (i.e., the peaks) and of the shoulders left over after subtraction of the static part. It is clear that although one may shift the position of the resultant shoulders in the dynamic part by a small amount relative to the position of the peak in the total intensity there is no question of shifting the anomaly all the way to coincide with T_{sg}. The two forms of the assumed static structure factor help to demonstrate that the anomaly represented by the peak can be made less noticeable by a suitable choice of the form of the static component. In particular, for the Au-10 at.% Fe and the Au-13 at.% Fe alloys the resultant shoulder in the dynamic structure factor which lies on a rapidly rising slope is "lost" within the statistical errors in the data points if the value of β' [Eq. (1b)] is increased to about 5. This, however, is not so for the Au-15 at.% Fe alloy where the scattering intensity and hence the statistical accuracy is significantly higher so that the anomaly remains noticeable especially at low q's after subtraction of any monotonic static component. The results for this alloy are shown in Fig. 3 where, for reasons of variety, the exponent β in Eq. (1a) is taken as 0.8, which is closer to the value $\beta=1$ predicted by mean-field theory,[6] but which still gives some curvature close to T_{sg}.

The cluster model of SGL comes close to describing the physical processes in spin-glasses. Its main limitations appear to be the use of the mean-field approximation and that the clusters of spins with their relatively strong intracluster interactions are well defined separate entities with a specified size distribution from the outset whereas the physical properties of spin-glasses indicate a continuous evolution of correlations in the spin system. Use of the mean-field approximation in equilibrium theories yields directly a finite static component below some temperature which naturally and unavoidably appears as a unique critical temperature. In this connection it is interesting to compare the discontinuity at T_{sg} in the slopes of SGL's computed total intensity curves, a feature in conflict with the experimental results, with a similar discrepancy in Soukoulis and Levin's[7] calculated curves for the specific heat of spin-glasses. In the latter case the authors obtain broad maxima in the temperature dependence of the specific heat which are qualitatively similar to the experimental results but, in addition, they obtain a sharp anomaly at T_{sg} not seen in the experimental data,[8] but which necessarily results from the use of the mean-field approximation in treating intercluster interactions in their model. In mean-field theory this anomaly in C_v has the same physical origin as the sharp kinks at T_{sg} in the total intensity curves and the q-independent maxima in the dynamic structure factor in SGL's cal-

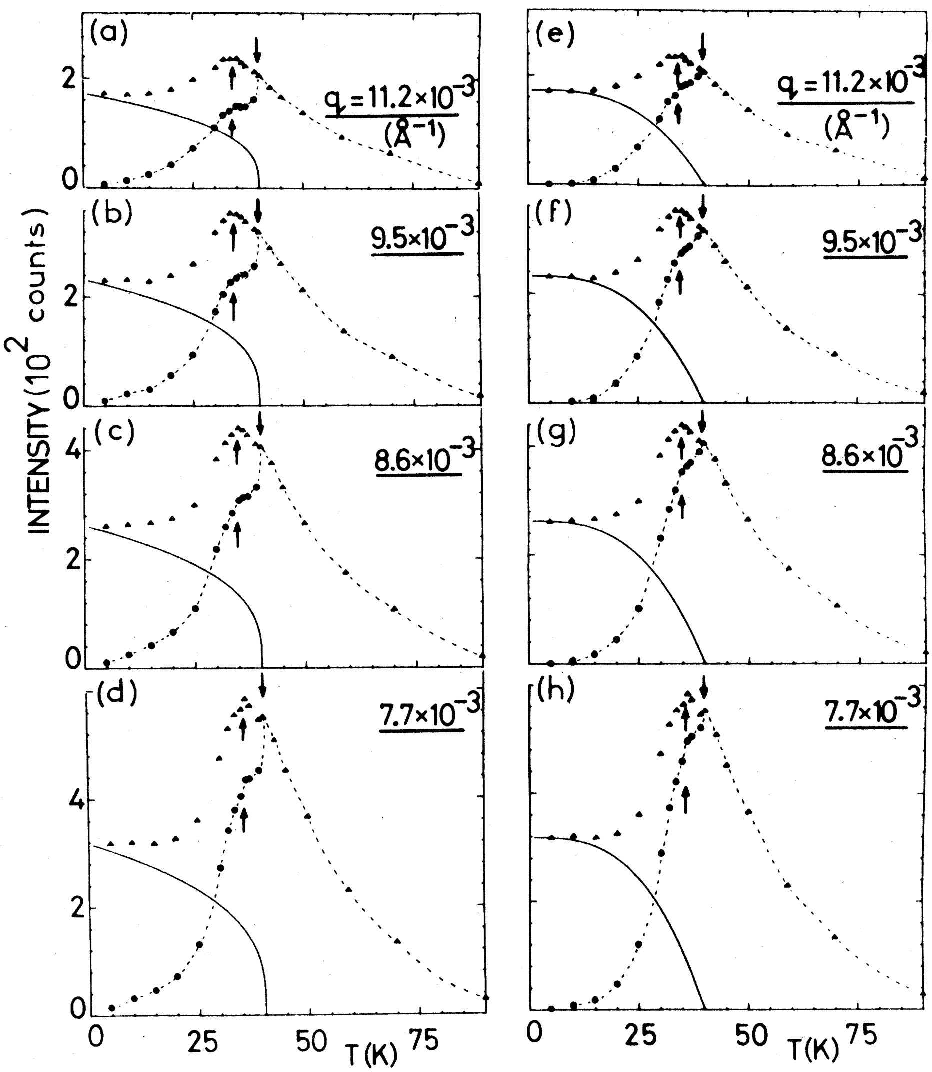

FIG. 2. Small-angle neutron scattering intensity (in units of 10^2 counts) for the Au-13 at.% Fe alloy. The triangles show the measured data points. The solid curves in (a) to (d) represent the static structure factor as given by Eq. (1a) with $\beta = \frac{1}{3}$. In (e) to (h) it has the form given by Eq. (1b) with $\beta' = 3$. The dashed curves through the resultant data point below T_{sg} and those above T_{sg} identify the total dynamic scattering intensity.

culated results.

In the absence of adequate theoretical explanation of the observed q-dependent peaks it is perhaps useful to compare and contrast the freezing of spins in these concentrated spin-glass alloys close to the ferromagnetic critical concentration with a conventional second-order phase transition. In both cases correlations in the spin system evolve with decreasing temperature. For an isotropic, atomically ordered Heisenberg magnet the correlation length is a unique quantity determined by the temperature whereas in the spin-glass alloys the spin system evolves with a whole spectrum of correlation sizes. As the temperature reaches T_c for a ferromagnet the correlation length diverges, and the spin system becomes frozen with the scattering intensity from the dynamic correlations going into the static component which gives the Bragg peaks. In the spin-glass, however, the correlation length presumably never becomes infinite, but as the collective relaxation time of spins correlated within a region $2\pi/q$ becomes long compared with the effective measurement time constant ($\sim 10^{-6}$ s for neutrons in the total scattering intensity measurements)[9] the dynamic structure factor is replaced by the static structure factor for the corresponding q. As discussed previously[1] it seems plausible that the relaxation times of such correlated interacting regions of spins depend inversely on their size through an

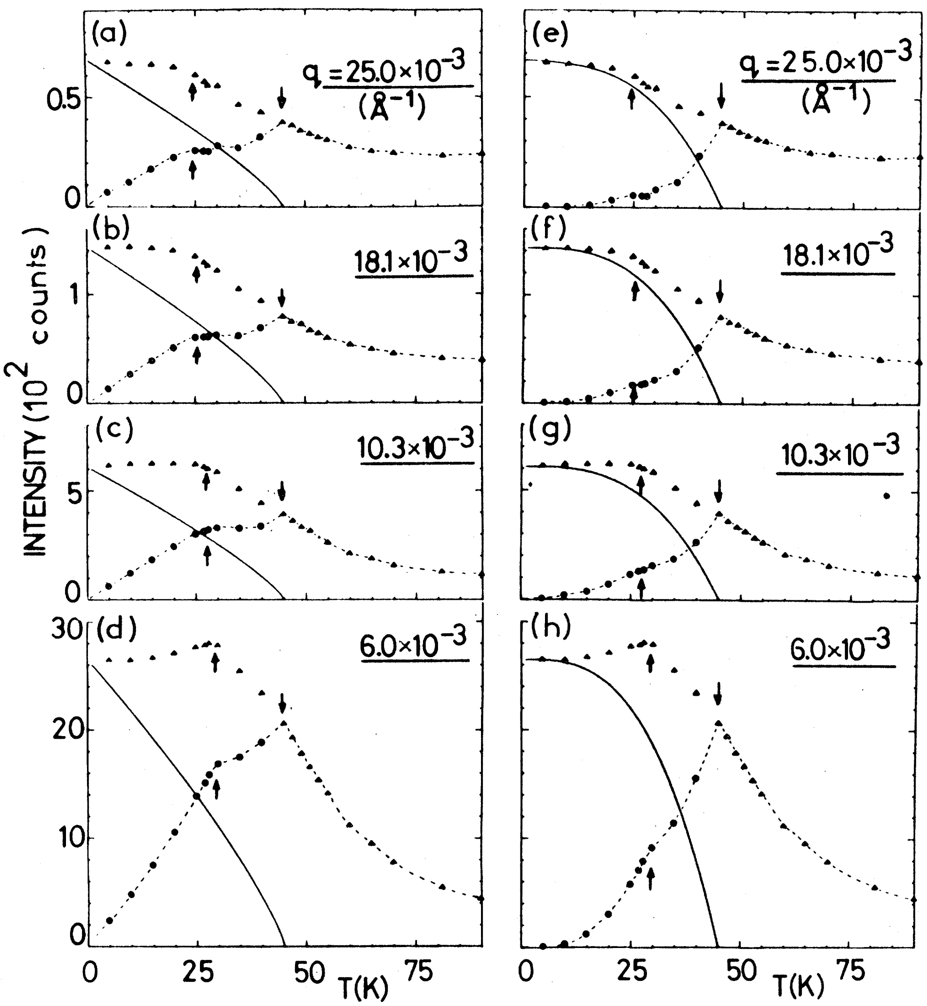

FIG. 3. Small-angle scattering intensity (in units of 10^2 counts) for the Au-15 at.% alloy. The exponent β [Eq. (1a)] for the static structure factor in (a) to (d) is 0.8 and $\beta'=3$ [Eq. (1b)] for (e) to (h).

Arrhenius relationship. We could see therefore the freezing process spread over a finite temperature range, as the spin system becomes frozen over progressively smaller regions with decreasing temperature.

The experimental results on *Au*Fe alloys appear also to bear qualitatively on some of the ideas contained in the localization theory interpretation of spin-glass behavior given by Anderson.[10] In this paper the author examined the problem of a crystalline lattice with random occupancy of sites coupled by near-neighbor interactions J_{ij} but with the concentration of occupied sites assumed to be below the percolation threshold for long-range order. For such a system, the eigenvalues of the random matrix J_{ij} are localized and the susceptibility χ^α of a localized eigenstate, i.e., a cluster, is given in the mean-field theory by

$$\chi^\alpha = C/(T-T_{c\alpha}) , \qquad (2)$$

where $T_{c\alpha}$ are nonsharp ordering temperatures forming a random continuum and there is no macroscopic ordering. Anderson[10] further argued that as with random lattices in general, localization could occur in the magnetic problem even with infinite range forces so long as the interaction matrix is sufficiently random, localization implying the exponential falling off of the wave functions at the edges. The $T_{c\alpha}$'s are now interrelated so that when $T_{c\alpha}$ of one of the localized eigenstates is reached it leads to renormalization of $T_{c\alpha}$'s of the others; i.e., the $T_{c\alpha}$'s are functions of temperature themselves. Hence with decreasing temperature the system passes through a series of nonsharp $T_{c\alpha}$'s.

In conclusion, it is hoped that the above reexamination of the experimental data demonstrates that the q-dependent peaks in the small-angle scattering cross section of *Au*Fe spin-glasses are real significant anomalies. It is emphasized that SGL's analysis of the experimental data creates an additional anomaly at T_{sg} which is well accounted for by their mean-field theory, but the observed q-dependent peaks in the total intensity may still be unexplained. The observed spread of temperatures of the peaks over the range of q values is large, $\Delta T/T_{sg} \sim 20\%$. which should be contrasted with the much smaller spread $\Delta T/T_c < 1\%$ for the temperatures of the critical scattering peaks in ferromagnets.[11,12]

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