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Phase separation in a spin-glass

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We have performed an extended x-ray absorption fine structure measurement of the Mn environment in a $Au_{0.95}Mn_{0.05}$ spin-glass. There is no evidence for changes in the Mn nearest neighbors with annealing, in spite of large changes in the magnitude of the cusp in the magnetic susceptibility. We propose a model based on phase separation which explains these apparently inconsistent observations in the context of the known structural and magnetic properties of the Au-Mn system. The effects of this annealing process on spin-glass behavior are discussed with reference to current theories.

I. INTRODUCTION

Spin-glasses are characterized by a prominent cusp in the magnetic susceptibility χ occurring at low temperatures ($\approx$ 15 K). The microscopic origin of this behavior is generally attributed to randomly coupled atomic spins.[1] The archetypal material is a dilute solution of transition-metal atoms in a noble-metal host. These atomic spins are conceived as being distributed randomly on the crystalline lattice. A regular but spatially oscillating pairwise interaction such as that of Ruderman-Kittel-Kasuya-Yosida (RKKY) will then lead to spin-spin interactions of varying sign and magnitude, random in a sense. It is this random spin-spin interaction which leads to the term "spin-glass," not a lack of atomic crystallinity. Typical systems include Au and Cu with 0.5 to 10% of the magnetic moment-bearing atom (Fe or Mn). Our EXAFS (extended x-ray absorption fine structure) experiment is motivated by an interesting observation for Au-Mn spin-glasses. The cusp in χ is enhanced strongly by annealing at high temperatures ($\approx$ 1200 K for a bulk sample and $\approx$ 600 K for a thin film).[2] A likely explanation for this annealing effect is that chemical clustering or even phase separation is occurring in the sample. Our insight into spin-glass behavior would be enhanced by understanding this phenomenon in detail in these spin-glasses. With that aim, we have performed an EXAFS measurement of the Mn environment in a $Au_{0.95}Mn_{0.05}$ spin-glass. We find no evidence for changes in the Mn nearest neighbors with annealing. We have formulated a model which explains the annealing dependence of the magnetic properties of Au-Mn spin-glasses in the absence of changes in the nearest-neighbor correlations. The model invokes localized phase separation in a manner consistent with the extensively studied structural and magnetic properties of compound phases in the Au-Mn system. The effects of this annealing process on spin-glass behavior are discussed with reference to current theories.

II. EXPERIMENT

We chose the Au-Mn system for this study because it has been the subject of extensive work in the composition range of compound formation (10–50 at.% Mn) as well as in the spin-glass regime ($<$10 at.% Mn). An earlier study[2] of thin-film $Au_{0.95}Mn_{0.05}$ alloys has shown that the cusp in the magnetic susceptibility χ increases substantially and sharpens upon annealing (see Fig. 1). Specifically, thin films were sputtered onto substrates held at 77 K, and were then annealed for up to 1 h at various temperatures T_a. For $T_a = 293$ K, χ is unchanged from the as-deposited films. For $T_a = 573$ or 1223 K, the magnitude of the cusp in χ increases by a factor of 2 or 3, respectively, from its value in the as-deposited films, although the temperature T_{sg} at which the cusp occurs is essentially unchanged. This is consistent with the well established but somewhat qualitative notion that the cusp in χ can be enhanced and sharpened in bulk materials by annealing at high temperatures.[3] For the EXAFS measurements, thin films of the crystalline $Au_{0.95}Mn_{0.05}$ were produced by sputtering onto thin aluminum substrates. Two different substrate temperatures were used during deposition: 77 K for our "unannealed" sample and 900 K for our "annealed" sample. While the "unannealed" sample

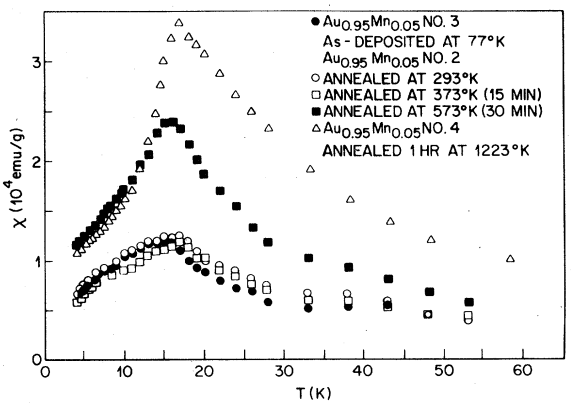

FIG. 1. Magnetic susceptibility as a function of temperature for various $Au_{0.95}Mn_{0.05}$ films annealed at different temperatures (from Ref. 2). Note the steady increase in χ with annealing.

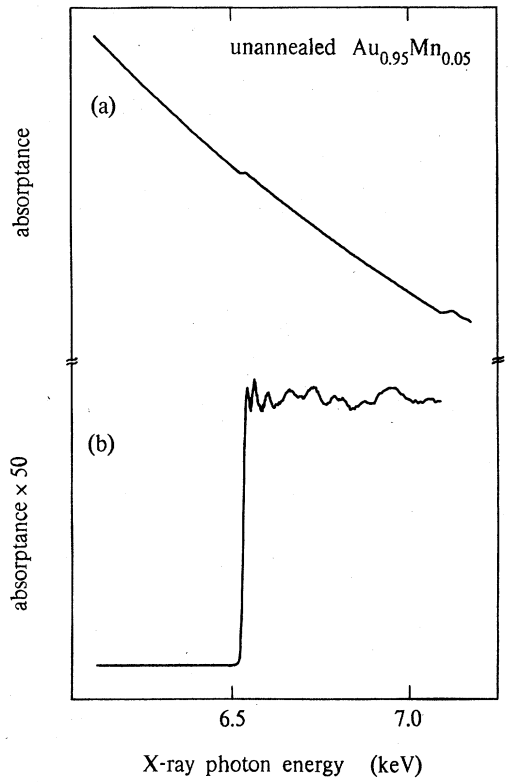

FIG. 2. (a) The absorptance of an unannealed sample of $Au_{0.95}Mn_{0.05}$, for a range of x-ray photon energies including the Mn K edge at 6.54 keV. The feature at 7.11 keV is due to iron impurities in the aluminum substrate. (b) The absorptance due to the Mn K edge alone, after subtracting a low-order polynomial from (a).

was, of course, unavoidably annealed at room temperature, the previous measurement[2] has shown that this produces no change in χ. The higher substrate temperature was chosen to produce a well-annealed sample, given the substantial kinetic energies available during sputtering. Crystalline thin films of Mn and of the compound Au_2Mn were prepared by sputtering onto substrates held at approximately 350 K. These materials serve as known structures in the subsequent analysis of the EXAFS data. The measurements were carried out at 77 K using the EXAFS facilities on the focused line at the Stanford Synchrotron Radiation Laboratory in a conventional transmission experiment.

For all the samples, the ratio of incident to transmitted intensity was recorded as a function of photon energy from 0.9 keV below to 1.2 keV above the Mn K edge at 6.54 keV. The $Au_{0.95}Mn_{0.05}$ alloy places stringent requirements upon a conventional transmission EXAFS experiment since the Mn K-shell absorptance is much smaller than the background Au absorptance. This is evident in the absorptance spectrum of unannealed $Au_{0.95}Mn_{0.05}$ shown in Fig. 2(a), where the Mn K edge is a very weak feature. The EXAFS is more clearly revealed after extraction of the prethreshold absorption with a low-order polynomial [see Fig. 2(b)]. The absorptance was measured repeatedly for the spin-glass samples and averaged. The EXAFS was extracted from the raw data using procedures described in detail elsewhere,[4] expressed as a function of final-state electron momentum k, and Fourier transformed to r space to give the ϕ's shown in Fig. 3 for unannealed and annealed $Au_{0.95}Mn_{0.05}$, Au_2Mn, and Mn. The real part and magnitude of the complex ϕ are displayed in the figure. For K-shell absorption, $\phi(r)$ can be expressed[5] as a sum of contributions from each shell of atoms surrounding the excited atom

$$\phi(r) = \sum_{\alpha = \text{Au,Mn}} \int_0^\infty dr' / r'^2 p_\alpha(r') \xi_\alpha(r - r') , \quad (1)$$

where $r > 0$ and p_α is the radial distribution function of atom species α about the excited Mn atom. p_α is defined so that $\int_0^\infty dr\, p_\alpha(r)$ equals the number of α atoms in the sample. Thus ϕ is a radial distribution function convolved with a peak function $\xi(r)$ (see Ref. 5 for a discussion). Previous work[6] has shown that ξ_α is sensitive to the backscattering atom species but not to changes in crystal structure, local bonding, thermal effects, etc. Our analysis of the data is based, accordingly, on the assumption that the ϕ differ only because of differences in p_{Au} and p_{Mn} from sample to sample. Thus the shift or broadening of a peak in the $p(r)$ appears as a linear shift of the peak function in ϕ or a convolution of it with a Gaussian, respectively. ϕ is then a linear combination of ξ's, appropriately shifted and broadened. The details of our analysis procedure are described elsewhere.[4,7]

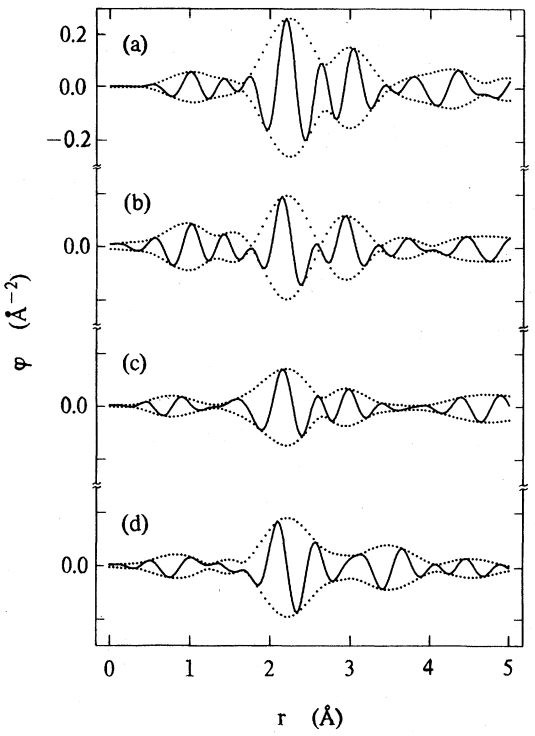

FIG. 3. The real part (solid line) and the magnitude of the Fourier transform of the EXAFS on the Mn K edge in four samples: (a) unannealed and (b) annealed $Au_{0.95}Mn_{0.05}$; (c) Au_2Mn (a compound); and (d) Mn. In each case, the k-space window is 3.15 to 8.80 $Å^{-1}$, broadened by 0.7 $Å^{-1}$. The vertical scales are identical.

The nearest neighbors of Mn in the Au_2Mn and Mn crystalline standards are Au and Mn atoms, respectively, at known distances. The shape of the peaks in Figs. 3(c) and 3(d), and, in particular, the detailed shape of the real part within the envelope (magnitude), should be taken as characteristic of Au and Mn nearest-neighbor backscattering atoms, respectively. Since Au is a heavy atom, even a single shell of Au neighbors will give rise in *every* instance to a double-peaked structure similar to that between 1.5 and 3.3 Å in Fig. 3(c).[7] The lighter Mn atoms give rise to a single-peaked structure similar to that between 1.7 and 2.8 Å in Fig. 3(d). The structures at lower r in Figs. 3(a)–3(d) arise from low-frequency noise in the data. A visual comparison of these spectra with the unannealed and annealed spin-glasses suggests that the nearest neighbors of Mn are Au in both cases. A detailed least-squares comparison among these spectra bears this out. In fact, there are no detectable Mn-Mn nearest neighbors in either spin-glass sample, strong evidence for the type of chemical ordering often seen in metallic glasses.[7] For example, in a least-squares comparison between the ϕ for the annealed sample and that for the unannealed, we find that the residual error R in the fit is least if it is assumed that there is no change in the number of Mn-Mn nearest neighbors with annealing. R increases by roughly a factor of 1.05 if 0.6 Mn atoms are added to or subtracted from the nearest-neighbor shell (0.6 Mn atoms would be 5% of the 12 neighbors in the first shell). Additionally, R increases by a factor of 1.3 if the unannealed sample is assumed to have as many as 0.6 Mn nearest neighbors. While the quality of the EXAFS data obtained from this difficult material does not allow the sort of detailed analysis possible in more ideal systems, it is safe to conclude that significant Mn clustering is unlikely. In addition, the Mn-Au nearest-neighbor distribution is a narrow peak. The only effects of annealing on the *nearest-neighbor* distribution are slight and probably insignificant changes in the width and position of this peak. These changes explain completely the apparently large differences between Figs. 3(a) and 3(b) in the region between 1.7 and 3.3 Å (even those for $r \approx 2.7$ Å).

III. PROPERTIES OF THE Au-Mn SYSTEM

We have formulated a model which explains the annealing dependence of the magnetic properties of Au-Mn spin-glasses in the absence of changes in the nearest-neighbor correlations. It is based on knowledge of the structural and magnetic properties of Au-rich compound phases in the Au-Mn system, Au_4Mn and Au_3Mn in particular. Consider firstly the structural information. At all temperatures, both of these compounds consist of a face-centered cubic (fcc) lattice of Au with Mn atoms occupying substitutional sites. At elevated temperatures, the Mn atoms are disordered on this fcc lattice. After long anneals at lower temperatures, the Mn atoms form ordered arrays characterized by large unit cells. In both cases, the fcc array of *lattice sites* (i.e., neglecting the atom species on the sites) becomes only slightly distorted, with the next-nearest-neighbor distance (i.e., cube edge) varying with direction by no more than ≈ 0.05 Å from its value for pure Au. In the fcc Au lattice, there are 12 neighbors at 2.88 Å, six at 4.07 Å, 24 at 4.99 Å, etc. Ordered Au_4Mn has the Ni_4Mo structure ($D1_a$) below 693 K, body-centered tetragonal with a unit cell 2.5 times as large as the underlying fcc cell.[8] The number of Mn neighbors of a Mn atom are 0, 2, and 8, respectively, for the first three near-neighbor positions. Ordered Au_3Mn has the Ni_3V structure ($D0_{22}$) below 918 K, tetragonal with a unit cell two times as large as the underlying fcc cell.[9,10] The number of Mn neighbors of a Mn atom are 0, 4, and 8, respectively. Au_2Mn has a layered body-centered tetragonal structure based on a body-centered-cubic (bcc) array of lattice sites.[11] The Mn

near neighbors of a Mn atom are four at 3.37 Å, four at 4.76 Å, and eight at 4.98 Å. From these three compounds, one could conclude that the Mn atoms repel one another at separations of 2.88 and 4.07 Å in an Au environment, approaching one another only as *required* to satisfy the stoichiometry. This conclusion is reinforced by both theoretical and experimental studies. General treatments of phase stability and spinodal decomposition[12,13] have led to the conclusion that both the first and the second-nearest-neighbor interactions must be repulsive between like atoms in systems which form the above structures. Further, the ratio of the second-neighbor repulsion to the first must be between 0.0 and 0.5. In a study of short-range order in disordered Au_3Mn (Refs. 10 and 14) using diffuse electron scattering, it was concluded similarly that the first and second neighbor interactions were both such as to repel like atoms, and that the second-neighbor repulsion was ≈ 0.08 of the first. The weakness of this second-neighbor interaction is consistent with the long anneals needed to achieve the ordered structures. In summary, the known structural properties of Au-rich compounds in the Au-Mn system lead us to expect that annealing will result in Mn atoms being driven out from first and second-near neighbor positions relative to other Mn atoms, to the extent permitted by the fixed stoichiometry.

The magnetic properties of these compounds also form an interesting pattern. Au_4Mn is ferromagnetic with a Curie point at 361 K.[15] Au_3Mn and Au_2Mn are both antiferromagnetic with Néel points at 145 K (Ref. 15) and 363 K (Ref. 16), respectively. The correlation between the growing number of Mn-Mn near neighbors and the progression from ferromagnetism to antiferromagnetism can be understood qualitatively on the basis of the RKKY interaction. For atomic spins which are this far apart (i.e., beyond 3 Å), the strongest spin-spin interaction is likely to be that derived from spin polarization of the conduction-electron gas as proposed by Ruderman, Kittel, Kasuya, and Yosida.[17] If the Fermi level of pure Au is preserved in the alloys, then the calculation of Geldart[18] suggests that the RKKY coupling should be ferromagnetic for radii below roughly 3.6 Å, antiferromagnetic from there until 5.1 Å, and oscillating thereafter as a sinusoid with argument $2k_Fr$ (plus a phase). The first antiferromagnetic maximum in the interaction occurs at approximately 4.4 Å. It is safe to assume that this calculation of the RKKY coupling is suggestive of the actual spin-spin interaction, but may be inaccurate in detail. From the known magnetic properties, one may conclude that the net interaction, considering *all* neighbors, is ferromagnetic if the number of Mn-Mn neighbors in the second-neighbor position (4.07 Å) are two or fewer (as in Au_4Mn). If this number is increased, their fundamentally antiferromagnetic coupling swings the balance in that direction (as in Au_3Mn where there are four). In Au_2Mn, the Mn-Mn near neighbors are even closer and more numereous, leading to more solidly antiferromagnetic behavior. As regards the annealing of the spin-glass samples, we might expect that the driving of Mn-Mn near neighbors to larger separations will result in a tendency to progress from antiferromagnetic to ferromagnetic behavior, at least on a local scale.

Support for these ideas comes from other annealing studies of dilute alloys of Mn in Au. It has been observed[3] that the formation of localized regions of Au_4Mn would be consistent with the magnetic properties. There is some indirect supporting evidence for such a phase separation. It was concluded from diffuse neutron scattering from 15 at. % Mn in Au that the short-range order after annealing resembled that in either Au_4Mn or Au_3Mn.[19] A Mössbauer study of 5, 10, and 20 at. % Mn in Au led to the conclusion that the short-range order which resulted from annealing was similar to that in Au_4Mn, in particular.[20] While neither these experiments nor the EXAFS data has yielded a definitive identification of the annealing process, there is a substantial accumulation of circumstantial evidence.

IV. MODEL FOR THE ANNEALING PROCESS

We are led to the following model for the effect of annealing on the structural and magnetic properties of our samples. Consider first the unannealed sample. After "annealing" at room temperature, the sputtered films are random *except* that there are no Mn-Mn nearest neighbors (as required by the EXAFS data). This is the chemical ordering which has been observed in metallic glasses,[7] driven by the large like-neighbor repulsion. The result is shown schematically for one dimension in Fig. 4(b). A "random" arrangement is represented in Fig. 4(a) for comparison purposes. For this illustration, the arrangement in Fig. 4(b) has been generated from that in Fig. 4(a) by moving any nearest-neighbor Mn atoms apart. Note that this process of excluding Mn-Mn nearest neighbors (beginning with a random arrangement) will result in a larger number of second neighbors than expected for a random system. This can be made quantitative by counting the number of near neighbor pairs. For the random arrangement shown in Fig. 4(a), there are 2, 1, 2, and 2 pairs of spins in the first, second, third, and fourth near-neighbor positions, respectively. After first-neighbor exclusion has taken place [Fig. 4(b)], these numbers become 0, 3, 2, and 5. To the extent that there are a large number of Mn-Mn second-nearest neighbors in the unannealed sample, there will exist many local environments within which the coupling is primarily antiferromagnetic.

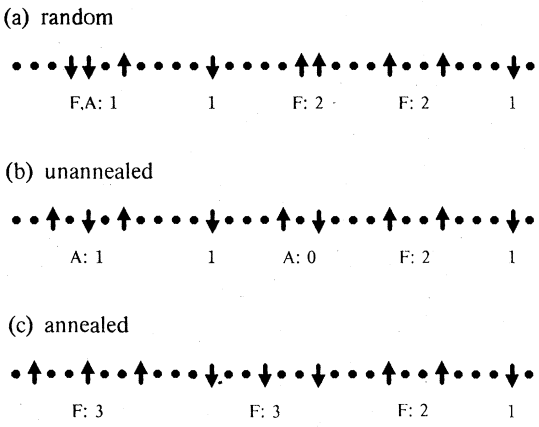

FIG. 4. A schematic one-dimensional representation of the position of Mn atoms in an Au lattice, and the orientation of the Mn spins, in $Au_{0.95}Mn_{0.05}$: (a) "random"; (b) the state characterizing the "unannealed" sample; and (c) the state characterizing the "annealed" sample. The Au atoms are represented by solid circles and the Mn atoms by arrows. Beneath each cluster of Mn atomic spins are noted its internal magnetic coupling (ferromagnetic or antiferromagnetic) and its net number of spins, as discussed in the text.

The effect of this on the magnetic properties can be understood qualitatively in the following way. The spin-spin interactions in our system presumably vary continuously from strongly ferromagnetic to insignificant to strongly antiferromagnetic depending on the distance between the spins, in a manner similar to that given by the RKKY expression. At a given intermediate temperature, some of these interactions will be strong enough to couple the spins effectively in spite of the disordering effect of temperature, while others will be too weak. At sufficiently high temperatures, none of the interactions are strong enough and the spins are effectively independent. In that limit, the paramagnetic susceptibility is given by the usual expression for noninteracting spins

$$\chi T \propto \eta = \sum_i s_i(s_i+1) , \qquad (2)$$

where the sum includes all spins. In order to assess the variation of η within our model for the annealing process, it is necessary to choose a value for s. The effective moment of Mn determined from the high temperature χ of Au_4Mn would yield $s=2$ for $g=2$.[16] This value of s would be appropriate for Mn with six $3d$ electrons. We choose $s=2$ accordingly. With this value $\eta=54$ for the nine spins shown in Figs. 4(a)–4(c) in the high-temperature (noninteracting) limit. As the temperature is lowered the spins which interact via the stronger forces will slowly become coupled. The susceptibility may be understood *qualitatively* through an arbitrary division of the interactions into two categories: treat all of the interactions with energy $> k_B T$ as leading to rigid couplings, and neglect all the weaker ones. This division is necessarily temperature dependent. For the present discussion of Fig. 4, let us consider a temperature such that we may treat only three interactions as nonvanishing: first, second, and third-nearest-neighbor interactions which are ferromagnetic, antiferromagnetic, and ferromagnetic, respectively. A "cluster" is a group of spins interconnected by these nonvanishing interactions. If the nonvanishing interactions are sufficiently short range, the concentration of spins sufficiently dilute, and the spin positions sufficiently random, then the system will consist of many clusters of varying size and separation. The spins in Figs. 4(a) and 4(b) have been oriented accordingly, yielding five clusters in each case. In this idealized case, the magnetic susceptibility is given by Eq. (2), except that s must be replaced with S, the vector sum of the spins within each cluster ($S_i = \sum_j s_{ij}$). While the number of atomic spins (Mn atoms) is conserved, the predominantly antiferromagnetic coupling within the clusters has the effect of preventing many of them from contributing to χ (i.e., S_i is reduced). For the situation in Fig. 4(b), $\eta=38$. At these lower temperatures, Fig. 4(a) corresponds to $\eta=58$. This difference is one consequence of the deviations from "randomness" in our model for the unannealed material. In the real system, of course, the arbitrary division of interactions into "strong" and "negligible" which underlies the concept of a cluster would be too unrealistic for a quantitative treatment. The "clusters" are not rigidly coupled internally, and always interact with one another to some degree via the weaker spin-spin forces. We expect the same qualitative behavior nonetheless.

We view the annealing as a migration of Mn atoms from second to third, and possibly more distant, neighbor positions with respect to other Mn. In other words, the near-neighbor environment becomes more like that of Au_4Mn than Au_3Mn. The nearest neighbors are unaffected, as required by the EXAFS data. Furthermore, this movement occurs only upon substantial annealing because the driving force is so weak.[10] We have represented the result of this process schematically in Fig. 4(c). Since our alloys contain only 5 at. % Mn, the second-neighbor position about each Mn atom could be swept completely clean of other Mn, as shown. The numbers of near-neighbor pairs in Fig. 4(c) are 0, 0, 5, and 3, respectively. Note the unstatistically large number of ferromagnetically coupled third neighbors. This is directly analogous to the behavior of the compound phases, where such a migration will lead to the growth of ferromagnetic "clusters" of composition similar to Au_4Mn at the expense of antiferromagnetic "clusters" of Au_3Mn. This will increase each of the

cluster S's, and is expected to lead to a substantial enhancement in χ for temperatures low enough that the third-neighbor ferromagnetic interaction is important. This is illustrated clearly in Fig. 4(c), where $\eta = 110$, nearly three times larger than in Fig. 4(b). This model explains the observed dramatic effect of annealing on the magnitude of the low-temperature magnetic susceptibility in the absence of any detectible changes in the nearest-neighbor environment.

V. EFFECT ON SPIN-GLASS BEHAVIOR

We now consider a second feature of the annealing dependence of the magnetic behavior of our samples. Although the magnetic susceptibility increases, the temperature T_{sg} at which the spin-glass cusp occurs is unchanged by annealing. The effect which a microscopic annealing process such as we have described will have on T_{sg} depends upon the physical origin of the spin-glass behavior, which is by no means universally agreed upon at present.[21,22] In general terms, the characteristic cusp in the χ for a spin-glass results as the high-temperature Curie-Weiss behavior of the spins is moderated by the spin-spin magnetic interactions, which become more effective as the temperature is lowered.[1] There are at least two distinct theoretical approaches to quantifying this behavior. The first regards T_{sg} as the temperature of a thermodynamic "spin-freezing" transition, characterized by an order parameter which is nonzero only below T_{sg}.[23–26] The other considers T_{sg} to be the temperature at which a percolation condition is met and an infinite cluster is abruptly formed from clusters of correlated spins which grow in size with decreasing temperature.[27,28]

In the order-parameter theories,[23–26] the atomic spins s_i are distributed in a regular array. The disorder comes from assuming that the spin-spin exchange interactions J_{ij} have a Gaussian distribution with a mean value J_0 and a half-width ΔJ. For $J_0 = 0$, T_{sg} is typically given by an expression of the form

$$k_B T_{sg} \propto c\,\Delta J s^2 , \tag{3}$$

where c is the concentration of spins. Sherrington and Kirkpatrick[24] considered a distribution with nonzero mean, and found that the spin-glass ground state is unstable relative to a ferromagnetic state if $J_0 > \Delta J$. This condition corresponds to the spin-spin coupling being too predominantly ferromagnetic. In other respects, T_{sg} is independent of J_0. For an adequate amount of antiferromagnetic coupling (i.e., $J_0 < \Delta J$), the susceptibility χ has the form

$$\chi(T) = \chi_0(T)[1 - J_0\chi_0(T)]^{-1} , \tag{4}$$

where $\chi_0(T)$ is the spin-glass susceptibility for $J_0 = 0$. The migration of Mn atoms which we have proposed to explain the annealing process has the effect of altering the distribution of exchange interactions. As we have described it, the predominant effect in an order-parameter model would be a shift of J_0 to larger values (increasingly ferromagnetic). This will increase χ according to Eq. (4), as we argued from general considerations. T_{sg} would be unchanged, however, except as the Mn migration might narrow the spread in the exchange interactions ΔJ. It is difficult to quantify this effect, but we would expect any change in T_{sg} to be rather smaller than the change in the magnitude of χ.

We note that Soukoulis[26] has formulated an order-parameter theory for a system in which sharply identifiable clusters have temperature-dependent moments M determined by the internal dynamics of the clusters. Interactions between the cluster moments lead to results similar to Eqs. (3) and (4), except that s is replaced by $M(T_{sg})$ in Eq. (3) and J refers to the intercluster interactions. Annealing in our model affects primarily the first few shells of near-neighbor spins, and hence only the largest exchange interactions. Presumably, these would be *intra*cluster interactions in such a model. In the Soukoulis model, then, we would expect that annealing will increase the cluster moment M as the near-neighbor interactions become predominantly ferromagnetic. Not only will this increase χ, as we have discussed, but it will also increase T_{sg} unless there is an offsetting decrease in the width of the intercluster interactions ΔJ. Thus the constancy of T_{sg} is not so easily understood in a model with sharply defined clusters. A similar observation has been made in a treatment of glassy MnSi, a concentrated spin-glass.[29] On the other hand, we would not expect such a model to be appropriate for our materials, partly due to the high concentration of atomic spins.

In cluster-percolation models,[27,28] the cusp in χ occurs when the infinite cluster is generated, at the percolation limit. This takes place at a fixed concentration of spins c because the range of interaction L increases with decreasing temperature. Specifically, L is the separation at which the RKKY interaction energy between two spins first falls below $k_B T$ (L increases with decreasing T). The percolation limit occurs when "enough" spins fall within the interaction range, and accordingly depends upon both c and L. In terms of percolation theory, one would say that the critical concentration required for percolation falls with decreasing T. T_{sg} is the temperature at which this critical concentration falls to the actual spin concentration c. Smith[27] finds for this model that

$$k_B T_{sg} \propto c s^2 , \tag{5}$$

where c is the concentration of spins on the lattice and s is the atomic spin magnitude which enters into determining L.[30] Futhermore, the expression[28] for χ is similar to our Eq. (2), so that its magnitude will increase if the near-neighbor ferromagnetic coupling is

enhanced on annealing, as we propose. Following Smith's analysis for $c = 0.05$ in an fcc lattice, the value of L at the percolation limit is 1.5 times the cube edge, intermediate between the fourth and fifth-near-neighbor shell distances. Since annealing as we have described it affects primarily the nearer neighbors, there should be little change in those interactions responsible for the growth of the infinite cluster. Accordingly, we would expect T_{sg} to be insensitive to annealing in cluster-percolation models as well.

As one would expect from the general considerations which we introduced earlier, the magnitude of χ is expected to increase with annealing within each of these models of spin-glass behavior. The effect of annealing on the temperature T_{sg} at which the spin-glass cusp occurs is more difficult to predict. We expect T_{sg} to be relatively insensitive, however, to the sort of annealing process which we are proposing. In both spin-glass theories, the cusp results from the net effect of many weak interactions of varying sign, coming from more distant shells than those principally affected by the annealing. This reflects the importance of "frustrations" which enhance the impact of the weaker interactions on cooperative magnetic phenomena. On the other hand, the annealing certainly causes deviations from "randomness" which should have *some* effect on any spin-glass model.

VI. SUMMARY

This EXAFS study of the Mn environment as a function of annealing in a $Au_{0.95}Mn_{0.05}$ spin-glass has led to the conclusion that the nearest neighbors of Mn are unchanged in spite of the large change in the magnetic properties on annealing. There is no evidence for Mn clustering. A model has been proposed for the annealing effects, based on the known structural and magnetic properties of the Au-rich compounds in the Au-Mn system. The model calls for the increasing separation of Mn near neighbors with annealing. This is not only consistent with other structural studies, but would also account for the observed large increase in the cusp in the magnetic susceptibility. These atomic movements are a very local manifestation of phase separation. The effects of this annealing process on spin-glass behavior have been discussed in the context of current theories, with emphasis on understanding the observed constancy of T_{sg}. Since the observed effects of annealing on dilute Cu-Mn and Au-Fe alloys are similar, one could conclude that understanding the systematic aspects of these phenomena would yield new insight into spin-glass behavior.

ACKNOWLEDGMENTS

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