

Transverse and longitudinal spin correlations in α -Fe₉₂Zr₇Sn

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⁵⁷Fe and ¹¹⁹Sn Mössbauer spectroscopy has been used to probe local spin correlations in α -Fe₉₂Zr₇Sn. Competing exchange interactions lead to two distinct magnetic transitions. The first, at T_c (180 K), marks the onset of long-range ($> 10 \mu\text{m}$) ferromagnetic order, and is followed by freezing of transverse spin components at T_{xy} (46 ± 6 K). Comparison of the average hyperfine fields at the ¹¹⁹Sn and ⁵⁷Fe sites indicates that the transverse spin components are strongly correlated on a nearest-neighbor length scale below T_{xy} while the ferromagnetic correlations show no detectable change on cooling through T_{xy} . By contrast, the magnetically similar $AuFe$ system shows a clear reduction in magnetic order on cooling that appears to be unrelated to the freezing of the transverse spin components.

I. INTRODUCTION

Competing ferromagnetic and antiferromagnetic interactions lead to a phenomenon known as exchange frustration and cause many magnetic materials to exhibit a noncollinear ground state. If the level of frustration is insufficient to make the system a spin glass, two distinct magnetic transitions are observed.¹ The first, at T_c , is to a collinear ferromagnetic state; however, substantial spin components transverse to the collinear order persist and precess rapidly so that their time average is zero. The second transition, at T_{xy} , marks the ordering of these transverse spin components. T_{xy} is identified, firstly, by the occurrence of deviation from the total local moment derived from Mössbauer spectra and the z component determined by magnetization measurements, and secondly, by the development of ordered spin components perpendicular to the magnetization. The freezing of the transverse spin components makes the spin system more resistant to rotation of the magnetization, and the rapid increase in anisotropy leads to a number of secondary effects being observed at T_{xy} , including reduction of the ac susceptibility and irreversibility in the dc susceptibility.

Many of the early observations of transverse spin freezing were made on $Au_{1-x}Fe_x$ ($0.15 \leq x \leq 0.2$) close to the percolation threshold²⁻⁵ and although there have been many suggestions that metallurgical instabilities may be influencing the behavior,⁶ the system remains one of the best examples. Iron-rich amorphous Fe_xZr_{1-x} ($0.88 \leq x \leq 0.93$) is an attractive alternative system in which to study the effects of partial exchange frustration.^{7,8} The rapid quenching used in the preparation of the alloys makes significant compositional modulation unlikely, while the high concentration of iron leads to relatively high ordering temperatures and eliminates possible percolation effects.

Although the magnetic phase diagrams for α -FeZr and $AuFe$ have been established, a detailed description of the magnetic order is still lacking. Both Lorentz microscopy⁹⁻¹¹ and neutron depolarization measurements¹¹⁻¹³

confirm that long-range ferromagnetic order is established at T_c and domains $\sim 10 \mu\text{m}$ across are observed. These domains persist unchanged through T_{xy} , clearly demonstrating that the freezing of the transverse spin components does not lead to a loss of collinear order. However, small-angle neutron scattering (SANS)¹⁴ shows that short-range (~ 10 nm) spin correlations also develop at T_c and that these appear to coexist with the long-range order that leads to the magnetic domains. The short-range correlations also persist essentially unchanged through T_{xy} . A complex modulation of the background within domains seen by Lorentz microscopy has also been attributed to fine structure in the magnetic order.^{9,15} Both the long- and short-range correlations are associated with the longitudinal spin components, and the presence of the short-range correlations within domains is probably caused by the exchange frustration.

In order to probe correlations in the transverse spin components, it is necessary to separate the contribution of the longitudinal components from that of the transverse spin components. One method which permits such a separation, on a nearest-neighbor length scale, is measurement of the transferred hyperfine field at a nonmagnetic atom in the alloy. The magnetic hyperfine field B_{hf} , measured by Mössbauer spectroscopy, can be written as

$$B_{\text{hf}} = aS_0 + b \sum_{i=1}^n S_i, \quad (1)$$

where S_0 and S_i are the magnetic moments of the probe and neighbor atoms respectively and the sum runs over the n nearest neighbors. The coefficients a and b relate the transferred field at the probe nucleus to the atomic moments causing it. In the case of a Mössbauer measurement with a magnetic probe atom, such as ⁵⁷Fe, B_{hf} is dominated by the contribution from the moment of the probe atom itself [i.e., the first term of Eq. (1)], and the effect of the neighbors amounts to only a few percent of the total field. With a nonmagnetic atom however, the local contribution is zero and B_{hf} measured at the probe site is due solely to the magnetic nearest neighbors [i.e.,

the second term of Eq. (1)]. The transferred hyperfine field in this latter case will therefore reflect any local correlation of the neighboring magnetic moments. If the spins remain fully parallel, the temperature dependence of the hyperfine fields at magnetic and nonmagnetic atom sites will be the same, and the ratio of the two hyperfine fields is temperature independent. However, if the degree of correlation is in any way temperature dependent, this simple proportionality is lost. For example, the second term of Eq. (1) may be resolved into longitudinal (parallel to the z axis) and transverse (xy) components¹⁶:

$$\left| \sum_{i=1}^n \mathbf{S}_i \right| = \left[\left(\sum_{i=1}^n S_z^i \right)^2 + \left(\sum_{i=1}^n S_{xy}^i \right)^2 \right]^{1/2}. \quad (2)$$

If the transverse spin components are assumed to have a 2D spin-glass ordering below T_{xy} (i.e., $\langle S_{xy}^i S_{xy}^j \rangle = 0$ for $i \neq j$), then

$$\left(\sum_{i=1}^n S_z^i \right)^2 + \left(\sum_{i=1}^n S_{xy}^i \right)^2 = n^2 (S_z)^2 + n (S_{xy})^2 \quad (3)$$

and the transverse spin components partially cancel, leading to a reduced transferred hyperfine field at the nonmagnetic atom site. The hyperfine field at the magnetic site is almost entirely caused by the local moment and so it will increase at T_{xy} as the extra spin components order. As a result, the ratio $B_{\text{hf}}^{\text{nonmag}} / B_{\text{hf}}^{\text{mag}}$ falls at T_{xy} . In disordered alloys, particularly amorphous alloys, there will be some variation in the local environments and so we obtain a distribution of the hyperfine fields rather than a single value. However, the average hyperfine field $\langle B_{\text{hf}} \rangle$ will reflect the mean environment and the above argument still holds.

A search for transverse correlations in the $AuFe$ system has been made using ^{197}Au Mössbauer spectroscopy in $\text{Au}_{83.2}\text{Fe}_{16.8}$ (Ref. 17) and ^{119}Sn Mössbauer spectroscopy in $\text{Au}_{79}\text{Fe}_{19}\text{Sn}_2$.^{16,18} For both alloys, the average hyperfine field derived from ^{57}Fe Mössbauer spectra clearly shows a sudden increase at T_{xy} as the extra spin components freeze and contribute to the local hyperfine field. The same behavior of the average hyperfine field is observed in the corresponding ^{197}Au or ^{119}Sn Mössbauer spectra, indicating that the transverse components are indeed locally correlated. Furthermore, because $\langle B_{\text{hf}} \rangle(T)$ clearly changes slope at T_{xy} , it is possible to separate the contributions of the longitudinal and transverse components. Extrapolating the high-temperature region ($T_{xy} \leq T \leq T_c$) to $T=0$ allows the average longitudinal moment to be estimated, while the value actually measured at $T=0$ reflects the contributions from both the longitudinal and transverse components. Comparing the ratios of these two values as measured at the magnetic and nonmagnetic sites allows the degree of correlation to be estimated. Assuming that the transferred hyperfine field at the nonmagnetic atom site (^{197}Au or ^{119}Sn) is due only to the moments on the nearest-neighbor iron atom leads to the conclusion that the transverse components are strongly correlated over first neighbor distances; indeed, in one case, better than perfect correlation is required.¹⁶

One problem with the measurements on the $AuFe$ sys-

tem is the rather low iron concentration in the region of interest. This leads to over one-third of the nonmagnetic probe atoms having less than two iron nearest neighbors and therefore being insensitive to any correlations. We present here similar measurements on $a\text{-Fe}_{92}\text{Zr}_7\text{Sn}$ which has similar magnetic properties but the greatly increased iron concentration means that any probe atom will typically have 10–11 magnetic neighbors allowing the spin correlations to be more clearly examined.

II. EXPERIMENTAL METHODS

Amorphous ribbons of nominal composition $\text{Fe}_{92}\text{Zr}_7\text{Sn}$ were melt spun under helium from ingots prepared from the pure metals by arc melting under argon. The absence of crystallinity was confirmed by x-ray diffraction and room-temperature Mössbauer spectroscopy. Sample composition was checked by electron microprobe analysis and found to be within 0.3 at. % of the nominal value. 1 at. % of Sn was added in order to probe the spin correlations at low temperatures. No significant change of structure or magnetic properties was expected as a result of introducing such a small amount of Sn. This was confirmed by the fact that the difference in T_c , obtained from the temperature dependence of the Mössbauer absorption in zero field, between $\text{Fe}_{92}\text{Zr}_8$ and $\text{Fe}_{92}\text{Zr}_7\text{Sn}$ is about 5 K. As can be seen in Fig. 1, the average transferred hyperfine fields at the ^{119}Sn and ^{57}Fe sites show similar temperature dependences, and the onset of magnetic broadening occurs at both sites at the same temperature. These observations indicate that the sample is homogeneous and no segregation of Sn has occurred.

Mössbauer spectra presented here were obtained over a temperature range from 5 to 90 K on a conventional constant acceleration spectrometer with $^{57}\text{CoRh}$ and

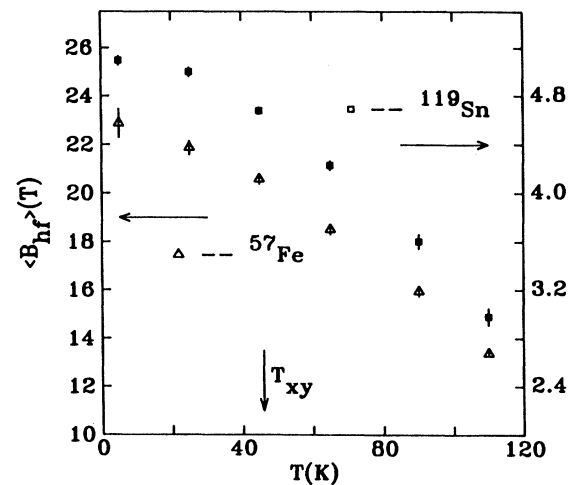


FIG. 1. Average hyperfine fields in $a\text{-Fe}_{92}\text{Zr}_7\text{Sn}$ derived from ^{57}Fe and ^{119}Sn Mössbauer spectra in zero field showing a smooth temperature dependence with no apparent break in slope at T_{xy} . The similar behavior of the two curves indicates that the sample is homogeneous.

$\text{Ca}^{119\text{m}}\text{SnO}_3$ sources at room temperature. High-field spectra were recorded with a 3.5 T field applied parallel to the γ beam using a superconducting solenoid. For these measurements, the source was located inside the cryostat at the null point of the magnet and the spectrometer was operated in sinusoidal mode. The field was applied above T_c and the spectra were obtained on field cooling. A field of 3.5 T was selected to place the material well onto the flat region of the magnetization curve (after demagnetizing fields are taken into account) where the system is technically saturated.

^{57}Fe spectra were fitted using both Window's method,¹⁹ where no assumptions about the shape of hyperfine distribution, $P(B_{\text{hf}})$, are made, and an asymmetric Gaussian distribution with independent widths above and below the most probable field. The average hyperfine field (the main parameter of interest here) differed by less than 2% between the two methods, although Window's method gave slightly better fits. Results derived from the first method are used here. For the ^{119}Sn spectra, $P(B_{\text{hf}})$ was proportionately much broader and Window's method proved unstable. For this reason, the ^{119}Sn spectra were fitted using the simpler asymmetric Gaussian distribution. This model fits the data quite well and eliminates the oscillatory tail frequency observed in the low-field part of $P(B_{\text{hf}})$ obtained by Window's method.

III. RESULTS AND DISCUSSION

^{57}Fe Mössbauer spectra of $\text{Fe}_{92}\text{Zr}_7\text{Sn}_1$ measured in a 3.5 T field applied parallel to the γ beam are shown in Fig. 2.

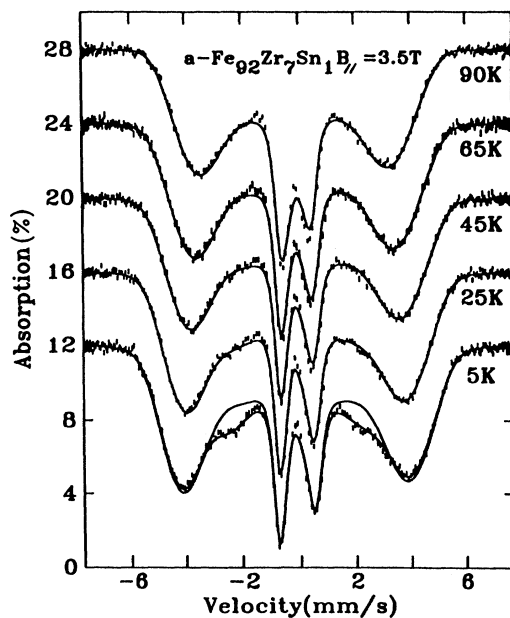


FIG. 2. ^{57}Fe Mössbauer spectra, measured with a magnetic field of 3.5 T applied parallel to the γ beam, showing the growth of lines 2 and 5 as the transverse spin components freeze below $T_{xy} = 46 \pm 6$ K.

The six lines observed in a magnetically split spectrum have intensities 3:R:1:1:R:3, where $R = 4 \sin^2\theta / (1 + \cos^2\theta)$, and θ is the angle between the magnetic moment and the direction of the γ beam. It is clear in Fig. 2 that lines 2 and 5, which are absent at high temperatures, appear at low temperatures. In Fig. 3, the fitted value of R is plotted as a function of temperature and shows the development of noncollinearity as the transverse spin components freeze below T_{xy} . R is zero in the collinear state above T_{xy} as the sample is fully magnetized parallel to the γ beam. On cooling through T_{xy} , the transverse components order perpendicular to the field (and γ beam) direction and R increases. The plot of R vs T in Fig. 3 shows that T_{xy} is 46 ± 6 K. While this value was obtained for an internal field of ~ 2 T (after correcting for demagnetizing effects), the field dependence of this transition temperature is either zero^{20,21} or at worst < 0.8 K/T (Ref. 22) and thus any possible field induced shift in T_{xy} is negligible in comparison with the uncertainty in the measurement. To get a quantitative measure of the deviation of the spin directions from the field direction, we consider a simple asperomagnetic model where the iron moments are distributed randomly within a cone of half-angle ψ .²³ At 5 K ψ is estimated to be $\sim 34^\circ$. Since the applied field (3.5 T) rotates the spins toward the field direction, we would expect a slightly larger deviation angle without the external field.

The ^{57}Fe and ^{119}Sn Mössbauer spectra at different temperatures are shown with fits in Figs. 4(a) and 4(b). As can be seen, the six lines in the ^{119}Sn spectra are poorly resolved and the spectra appear to consist of only two peaks. It is not possible to obtain a unique value for R by fitting a spectrum under these conditions, and we cannot simply assume that $R = 2$, since there is always some magnetic texture present as a result of demagnetizing fields and quenched-in stresses introduced during preparation. In ribbon samples this frequently leads to R smaller than 2 for ^{57}Fe spectra.²⁴ The fitted shape of $P(B_{\text{hf}})$ (and hence its average value) depends somewhat on the value of R used in the fit, therefore an uncertainty in R translates into an uncertainty in $\langle B_{\text{hf}} \rangle$. The size of this uncertainty has been estimated by comparing the

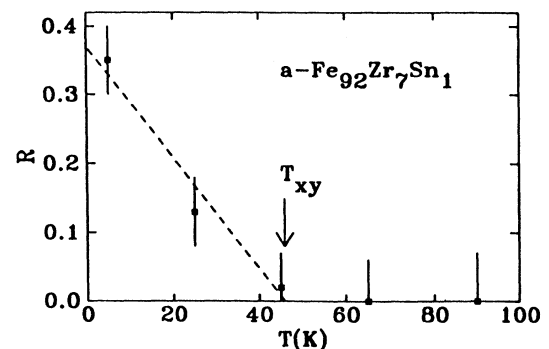


FIG. 3. Relative intensity of lines 2 and 5 with $B_{\parallel} = 3.5$ T showing the increase in R below T_{xy} as the transverse spin components order.

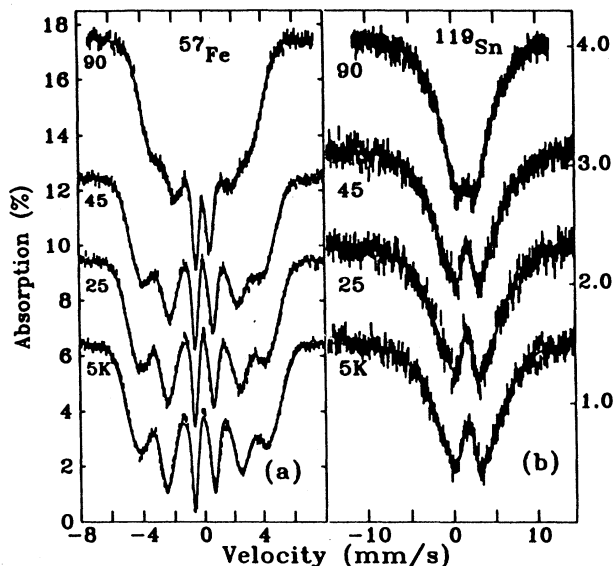


FIG. 4. Mössbauer spectra of $a\text{-Fe}_{92}\text{Zr}_7\text{Sn}$ with fitted curves measured at different temperatures with (a) ^{57}Fe and (b) ^{119}Sn sources.

average hyperfine field obtained using $R = 2$ (appropriate for a random spin distribution) with the value obtained using R derived from fitting the ^{57}Fe spectrum at the same temperature. In the temperature range from 5 to 90 K, the difference was found to be less than 2%.

In contrast with AuFe where $\langle B_{\text{hf}} \rangle(T)$ shows a clear break in slope at T_{xy} (Refs. 16–18), $\langle B_{\text{hf}} \rangle(T)$ in $a\text{-Fe}_x\text{Zr}_{100-x}$ increases smoothly with falling temperature.^{8,25,26} Figure 1 shows this behavior is observed at both the ^{57}Fe and the ^{119}Sn sites in $a\text{-Fe}_{92}\text{Zr}_7\text{Sn}$. The absence of an observed break in slope is probably caused by the much greater spread in local environments encountered in glasses, which broadens the lines making observation of the break difficult, and may also be expected to smear out the transition at T_{xy} thus masking the change. Furthermore, the strong correlation between R and the shape of $P(B_{\text{hf}})$ coupled with the fact that R changes at T_{xy} means that $\langle B_{\text{hf}} \rangle$ cannot be determined to better than a few percent unless a separate, independent determination of the shape of $P(B_{\text{hf}})$ is also made.²⁴ As we do not observe a break in the slope of $\langle B_{\text{hf}} \rangle(T)$ at T_{xy} , we cannot attempt to separate the longitudinal and transverse spin components as has been done previously.^{16–18} However, an analysis of the temperature dependence of the average hyperfine fields on the magnetic and nonmagnetic atom sites is still possible and information about spin correlations can be obtained. Using the value of ψ derived from the asperomagnetic cone model described above, the average transverse component is estimated to be about $0.40 \mathcal{S}$, where \mathcal{S} is the total iron moment. If we now assume a random arrangement of these transverse spin components below T_{xy} , then the ratio of the average ^{119}Sn hyperfine field to the ^{57}Fe hyperfine field should exhibit an 8% drop below T_{xy} . The temperature dependence corresponding to this situation is plotted as a solid

line in Fig. 5. Within the quoted uncertainty, the ratio is independent of temperature over the entire range covered here, as indicated by the dotted line. The observed temperature dependence is therefore inconsistent with a high degree of disorder in the transverse spin components. Furthermore, Fig. 5 also rules out the possibility that there is any loss of order in the collinear components at any temperature below T_c , including T_{xy} . If we take the drop in the ratio at 5 K to be real, we can set a limit on the degree of disorder in the transverse components. As a simple model, we modify the 3D asperomagnetic cone model to describe the transverse components as lying within a sector in the xy plane with a half-angle ϕ about a locally preferred direction. From the decrease of the ratio at 5 K, a ϕ of $\leq 50^\circ$ is estimated. We emphasize that the measurement is consistent with $\phi = 0^\circ$. The method is not particularly sensitive even to relatively large deviations from collinear order in the xy plane because the transverse moment fraction is quite small in this alloy.

It is instructive to repeat our analysis on the ^{197}Au and ^{119}Sn Mössbauer data from the AuFe alloys. Figure 6 shows a plot of the ratios of average hyperfine fields as a function of temperature for the two data sets. Comparison with the Fe-Zr data in Fig. 5 shows that the behavior of the transferred field ratio is quite different. The T_c and T_{xy} values of the two AuFe alloys are similar both to each other and to the present $a\text{-Fe-Zr}$ alloy and may be expected to exhibit comparable magnetic properties. However, in the AuFe data, the hyperfine field ratio falls steadily on cooling. For the ^{197}Au Mössbauer measurement on $\text{Au}_{83.2}\text{Fe}_{16.8}$, the ratio drops about 26% between 80 and 5 K. Even if random ordering of the transverse spin components is assumed, the expected reduction of the ratio is much smaller than that observed; both longitudinal and transverse components of the iron moments

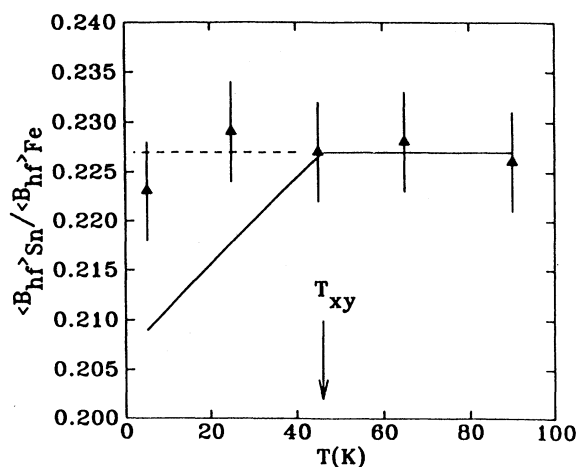


FIG. 5. Ratio of the average transferred hyperfine field at ^{119}Sn sites to that at the ^{57}Fe sites as a function of temperature in $a\text{-Fe}_{92}\text{Zr}_7\text{Sn}$. The absence of any temperature dependence indicates that both the longitudinal and transverse spin components are strongly correlated over nearest-neighbor distances. The solid line shows the expected behavior if the transverse spin components were to freeze in random orientations.

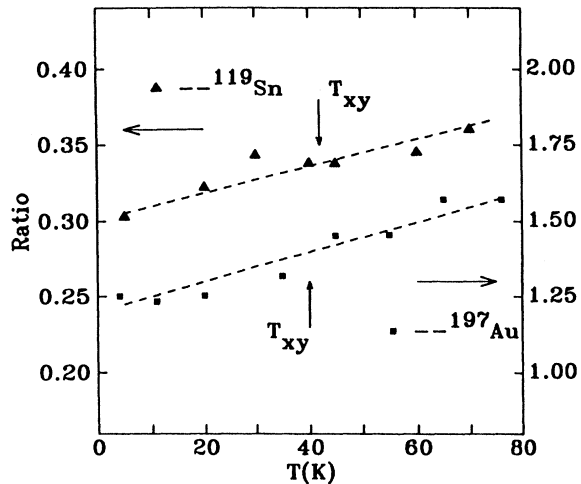


FIG. 6. Ratios of the average transferred hyperfine field at ^{197}Au and ^{119}Sn sites in two AuFe alloys to that measured at the ^{57}Fe sites in the corresponding alloy (calculated from the data of Refs. 17 and 16). The strong temperature dependence and absence of any feature at T_{xy} reflects a significant reduction in the correlations between the longitudinal spin components in these alloys.

can be estimated from ^{57}Fe Mössbauer spectra and the average transferred hyperfine fields at the ^{197}Au sites can be calculated. The decrease in the ratio estimated on this basis is only 14%, and moreover, the decline should only start below T_{xy} . The observed behavior is quite distinct from that of the Fe-Zr system and suggests a decrease in the correlation length of longitudinal spin components on cooling. Recent neutron depolarization measurements on Fe-Zr and AuFe alloys^{11,13} are also consistent with this conclusion. Although neutron depolarization is not sensitive on length scales smaller than the order of a micron, it does allow measurement of the size and average internal magnetization of domains. For $\text{Fe}_x\text{Zr}_{1-x}$, the polarization decreases strongly at T_c for alloys with $x < 0.92$, indicating the formation of domains, and these domains persist through T_{xy} (Ref. 11). For all samples, polarization decreases to 15 K and gives no evidence of a reduc-

tion of correlation length as the temperature is lowered. By contrast, in the AuFe alloys, the polarization first decreases below T_c as expected, then starts to increase again.¹³ The increase of polarization indicates a reduction of either the mean domain size or the internal domain magnetization, and requires a decrease in the correlation length of the longitudinal spin components.

Despite the strong similarities between the magnetic phase diagrams of the AuFe and $a\text{-Fe-Zr}$ systems and the values of the transition temperatures of the particular alloys studied here, quite different spin correlation behavior is observed. The longitudinal spin correlations in $a\text{-Fe-Zr}$ are quite robust, being unaffected by the ordering of the transverse spin components which also appear to be strongly correlated. By contrast, even the longitudinal correlations seem to decay in AuFe . This distinction was unexpected; however, several possible sources for it may be identified. $a\text{-Fe-Zr}$ has been shown to exhibit displaced hysteresis loops²⁷ whereas AuFe does not²⁸ and it is possible that the stiffness of the spin system below T_{xy} plays a role in stabilizing the spin correlations in the presence of exchange frustration. Another possibility is that the proximity of the AuFe alloys to the percolation threshold leads to a fragmentation of the order at low temperatures either as a result of random-field effects from the transverse spin components or because the effects of small compositional fluctuations are more significant in such marginal systems. While the true origin of the difference remains unclear, the effect is well defined and further work on a variety of similar systems is under way to identify the controlling factor.

IV. CONCLUSIONS

Transverse spin freezing has been observed in $a\text{-Fe}_{92}\text{Zr}_7\text{Sn}$ below $T_{xy} = 46 \pm 6$ K. The temperature dependence of the ratio of the average hyperfine fields at the ^{119}Sn and ^{57}Fe sites indicates that the transverse spin components are strongly correlated on a local scale below T_{xy} , while the longitudinal components are unaffected by the additional ordering. Comparison of our $a\text{-Fe-Zr}$ data with data on AuFe alloys shows that although both systems exhibit transverse spin freezing there is a significant reduction in the spin correlations in the latter system at low temperatures.

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