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Evolution of magnetic surfboards and spin glass behavior in $(Fe_{1-p}Ga_p)_2TiO_5$

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Abstract

The unusual anisotropy of the spin glass (SG) transition in the pseudobrookite system Fe_2TiO_5 has been interpreted as arising from an induced, van der Waals-like, interaction among magnetic clusters. Here we present susceptibility (χ) and specific heat data (C) for Fe_2TiO_5 diluted with non-magnetic Ga, $(Fe_{1-p}Ga_p)_2TiO_5$, for disorder parameter $p = 0$, 0.11, and 0.42, and elastic neutron scattering data for $p = 0.20$. A uniform suppression of T_g is observed upon increasing p, along with a value of $\chi\,(T_g)$ that increases as T_g decreases, i.e. $d\chi\,(T_g)/dT_g < 0$ We also observe $C\,(T) \propto T^2$ in the low temperature limit. The observed behavior places $(Fe_{1-p}Ga_p)_2TiO_5$ in the category of a strongly geometrically frustrated SG.

Keywords: anisotropic spin glasses, neutron scattering, geometric frustration

(Some figures may appear in colour only in the online journal)

1. Introduction

The pseudobrookite compound Fe_2TiO_5 is unusual for exhibiting spin glass (SG) freezing signatures—a cusp in susceptibility, $\chi\,(T)$, at the glass transition temperature, T_g, and hysteresis in $\chi\,(T)$ below T_g—only with the applied magnetic field along the crystallographic $\boldsymbol{c}$-axis [1]. When probed in the $\boldsymbol{a}$ and $\boldsymbol{b}$ directions, no anomaly is seen at T_g, which is highly unusual because the magnetic ion, Fe^{3+}, is an s-state ion and thus possesses no single-ion anisotropy. In addition, the Fe and Ti atoms randomly occupy the two cation sites. When a similar s-state ion, Mn^{2+}, resides on a periodic lattice, such as in MnF_2, anisotropy develops below the Néel transition due to the dipole–dipole energy, which establishes an ordering direction. It seems unlikely, therefore, given the randomness of the Fe^{3+} ions, that this mechanism can explain the anisotropy in Fe_2TiO_5. Thus, it is also likely impossible to describe the complete anisotropy in its SG response using atomic spins as the freezing degree of freedom. Recently we revisited this problem and showed, using neutron scattering, that the Fe spins develop nano-sized regions of antiferromagnetic (AF) order where the spins are either aligned or anti-aligned with the long direction, parallel to the $\boldsymbol{a}$-axis [2]. These regions take the shape of surfboards with correlations lengths

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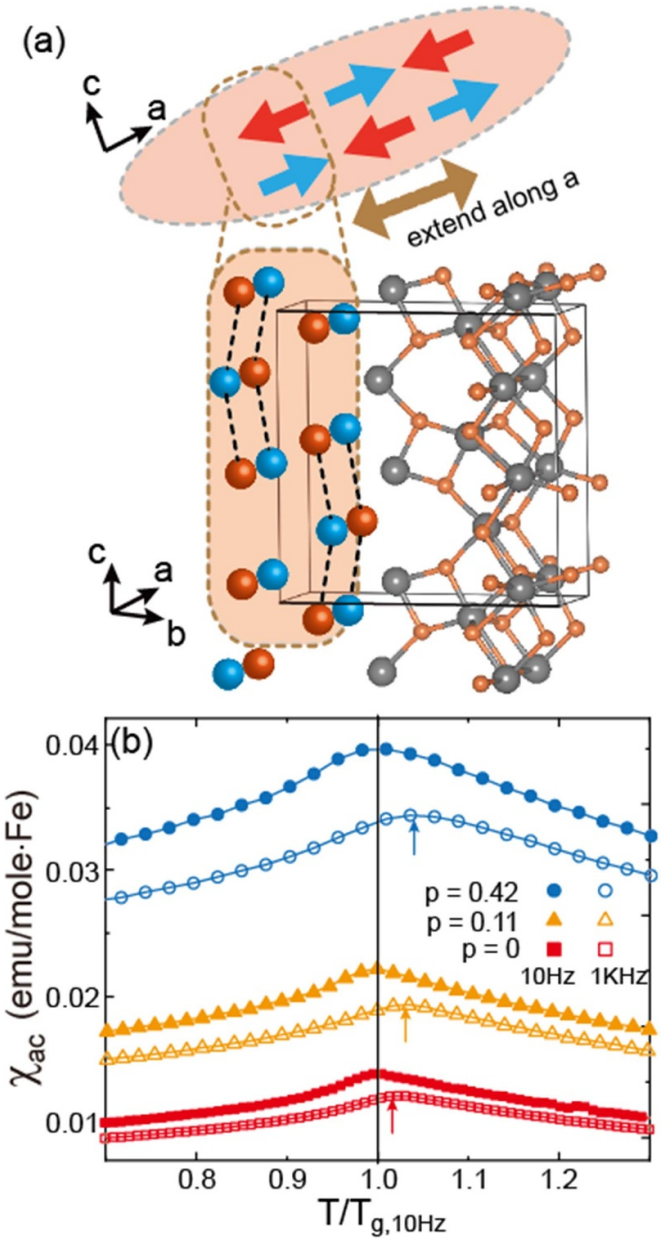

Figure 1. Illustration of the crystal and magnetic structure in Fe_2TiO_5. The Fe/Ti positions are represented by large spheres and the oxygen atoms by small yellow spheres. The large spheres are colored red (blue), indicating spins that are parallel (anti-parallel) with the *a*-axis, respectively. The grey color indicates that the direction of the spins is undetermined due to geometric frustration. The oxygen atoms on the left part of the unit cell are omitted to better show the double corrugated spin chains as indicated by the dashed lines. The shaded area represents a cross-section of the mesoscopic surfboard (top) that extends along the *a*-axis, as determined in [2]. (b) Magnetic ac susceptibility along the *c*-axis were measured on $(Fe_{1-p}Ga_p)_2TiO_5$ compounds with $p = 0$, 0.11 and 0.42. A modest shift in T_g as expected for a spin glass is clearly seen to higher temperature with increasing frequency.

of Å × 10 Å along the ***a***- and ***c***-axis, respectively, but along the ***b***-axis, only nearest neighbor spins are correlated due to frustration (figure 1). The principal building block of these surfboards are corrugated AF chains, as depicted in figure 1(a). The frustrated coupling between such chains leads to the short-range order within surfboards. We also showed that the interaction between the surfboards comes from the fluctuations of their magnetization along the ***c***-direction, i.e. the direction transverse to the ordered spins. Such a *magnetic van der Waals* force can thus explain, at least qualitatively, the anisotropy of freezing in Fe_2TiO_5.

An important aspect of Fe_2TiO_5 is its strong geometrical frustration, as indicated by a large frustration parameter, $f \equiv \theta_W/T_g = 900/55 \approx 16$ [3], where θ_W is the Curie–Weiss temperature. As described in recent work by two of us [4], when varying the amount of quenched disorder in strongly geometrically frustrated (GF) systems, the SG transition temperature T_g and magnetic susceptibility $\chi(T_g)$ follow the universal trend $d\chi(T_g)/dT_g < 0$, which is opposite to the trend $d\chi(T_g)/dT_g > 0$ displayed by conventional SGs. Furthermore, T_g in strongly GF materials grows with decreasing the density of vacancies [4], the dominant source of quenched disorder. This trend calls into question the achievability of quantum spin liquids widely sought in these materials, because it suggests that the SG state, believed to be incompatible with quantum spin liquids, occurs even in very clean samples.

Although Fe_2TiO_5 belongs to the same class of strongly GF materials, it is distinct from other GF materials in that it develops the described surfboard-shaped clusters and these clusters apparently play an essential role in the SG freezing. The strong interaction between these clusters, in addition to the strong super-exchange interaction between Fe^{3+} spins, may be responsible, for example, for higher SG-transition temperatures in Fe_2TiO_5 than in the other GF systems [4]. The SG transition and the structure of the glass state in Fe_2TiO_5 thus require a separate careful investigation.

Here, we present $\chi(T)$ and specific heat $(C(T))$ results for the dilution series $(Fe_{1-p}Ga_p)_2TiO_5$ for $0 \leqslant p \leqslant 0.42$, in which non-magnetic Ga^{3+} substitutes for Fe^{3+}, as well as neutron scattering data for $p_{\text{nominal}} = 0.20$. Similar to other GF systems where $f \gtrsim 10$, we find that T_g decreases with increasing susceptibility, $d\chi(T_g)/dT_g < 0$, as disorder is increased with Ga content, p. The specific heat also displays a low-temperature quadratic-in-T term that grows with p, similar to that seen in other GF materials. Our neutron scattering measurements indicate the existence of surfboard-shaped correlated regions, like in Fe_2TiO_5, but substantially shrinking, as p is increased. Thus, whereas T_g reduction in other GF systems is due to an increase in vacancies around which 'quasispins' form and undergo freezing, in $(Fe_{1-p}Ga_p)_2TiO_5$ the quasispins are AF-ordered surfboard-shaped regions. This distinction, in addition to the large exchange coupling reflected by the θ_W, may explain why the energy scale for SG freezing is larger in the present material than in other GF systems. Nevertheless, in both types of systems the quasispin is a composite object, i.e. formed from many atomic spins and, thus noted, the argument of [4] pertains.

2. Methods

In the present study, we used crystals of $(Fe_{1-p}Ga_p)_2TiO_5$, grown by J P Remeika [5] for measurements of magnetic

susceptibility and specific heat. Because these crystals were grown from a flux, their nominal concentrations serve only as a coarse guide to the resultant concentrations. Also, since techniques for atom identification in the solid state, such as energy dispersive atomic spectroscopy, possess systematic uncertainty in ratios between constituent atoms, we rely here on the susceptibility itself to determine the concentration of Fe in each sample. This approach relies on our previously reported measurement of $\chi\,(T)$ in Fe_2TiO_5 for temperatures between 400 K and 900 K, which yielded an effective moment of $\mu_{\rm eff} = 6.12 \pm 0.05\,\mu_B$, in reasonable agreement with the value $\mu_{\rm eff} = 5.92\,\mu_B$ expected for a free $S = 5/2$ ion [2]. Since the most concentrated compound of the series yielded an effective moment close to the theoretical value, it is reasonable to expect that the effective moment in more dilute compounds will be identical since the chemical environment and oxidation states are the same. We note that, since Fe^{3+} and Ti^{4+} can each occupy either the A or B sites in the pseudobrookite A_2BO_5 structure, even for $p = 0$, the Fe occupation is random. In addition, due to the different oxidation states of Fe and Ti, the pure pseudobrookite compound, Fe_3O_5, cannot be made with only trivalent Fe ions, i.e. with zero magnetic disorder. Since Ga^{3+} is isovalent with Fe^{3+} but non-magnetic and similar in size, substituting it for Fe^{3+} will increase magnetic disorder while decreasing spin density. We studied flux-grown crystals for 3 nominal values: $p_{\rm nominal} = 0$, 0.3, and 0.5. Based on the effective moment from high temperature susceptibility, we estimated the corresponding actual concentrations to be $p_{\rm estimate} = 0$, 0.11, and 0.42; hereafter, we refer to the values of p by those determined via $p_{\rm estimate}$ for simplicity. A larger single crystal with $p_{\rm nominal} = 0.2$ for neutron scattering was grown at Argonne National Laboratory in a floating zone furnace. Based on comparison of T_g measured by magnetometry of a single crystal fragment grown by the floating zone to those measured for the flux-grown crystals, we estimated the concentration to be $p_{\rm estimate} = 0.17$. Structural refinements of full single-crystal x-ray diffraction datasets, collected using an Oxford Diffraction Xcalibur-2CCD diffractometer indicated almost complete random occupation of the A and B-sites in the pseudobrookite-type structure by Ti, Fe, and Ga, with a slightly higher electron count, of the order of 0.8 electrons or an average of 26 electrons on the B-site. This thus suggests that the increasing magnetic disorder on both A and B-sites with increasing p is notionally correct.

3. Results

The crystal structure of the flux-grown crystals was confirmed by single crystal x-ray diffraction to be the pseudobrookite phase. The orthorhombic lattice constants $\{a,\ b,\ c\}$ in Ångstroms for $p = 0$, 0.11, and 0.42 are {3.732(1), 9.8125(2), 10.0744(2)}, {3.7023(1), 9.7872(2), 9.9670(2)} and {3.6857(1), 9.7903(2), 9.9681(2)} respectively. The dependence of $\{a,\ b,\ c\}$, as well as the volume of the unit cell $V(p)$, on p is shown in the upper left inset of figure 2, normalized to their $p = 0$ values. The volume monotonically decreases with increasing p, consistent with the smaller ionic radius of Ga^{3+} compared to Fe^{3+} and indicative of successful substitution. The monotonic $V(p)$ emanates primarily from the monotonic decrease in a with p, while there are less changes in b and c from p =0.11 to 0.42. Magnetization measurements were performed in two different Quantum Design Magnetic Property Measurement Systems (MPMS3s). For $\chi(T < 300$ K), a conventional sample holder was used. For $\chi(T > 300$ K), the sample was mounted with Zircar cement on a rod designed for high-temperature measurements. The specific heat, $C(T)$, was measured for $T > 2.5$ K using the relaxation technique in a Quantum Design Physical Property Measurement System (PPMS). For $T < 4$ K, $C(T)$ was measured using the semi-adiabatic technique in a top-loading dilution refrigerator. Energy dispersive x-ray absorption spectroscopy was performed on the samples and yielded a Fe:Ga ratio that confirmed the general trend expected for Ga substitution. The neutron scattering measurements were performed on the time-of-flight instrument Corelli (ORNL) using a pseudo-white beam [6]. This is the same instrument which was used to measure Fe_2TiO_5, as reported previously [2].

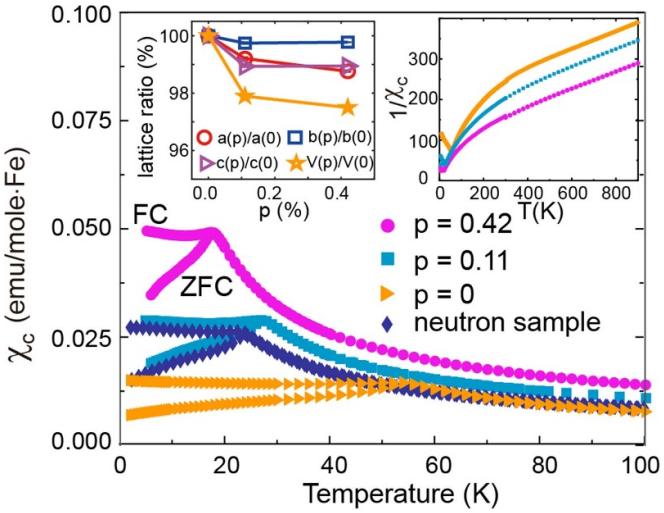

Figure 2. C-axis susceptibility of $(Fe_{(1-p)}Ga_p)_2TiO_5$ for various values of p measured with an applied field of $\mu_0 H = 0.01$ T. The lower curve at temperatures below each kink were taken after zero field cooling and the upper curves after field cooling. Inset: left, the dependence of lattice constants and the unit cell volume on p; right, the inverse susceptibility for the same samples. The p-values for the samples other than $p = 0$ were determined by adjusting the molar mass to yield the same high-temperature slope of $1/\chi\,(T)$ as the $p = 0$ sample, the Fe concentration of which is known exactly. The crystal labeled 'neutron sample' was a measurement of a small single crystal grown in the same floating zone furnace with the same nominal composition as that as the crystal measured by neutron diffraction.

In figure 2 are shown magnetic susceptibility with applied field along the c-axis, $\chi_c(T)$, characterizing the anisotropic SG transition in $(Fe_{(1-p)}Ga_p)_2TiO_5$ with $p = 0$, 0.11, and 0.42. It is important to recognize that, since Fe, Ga, and Ti can each occupy either of the two cation sites, the magnetic ion stoichiometry $p = 0$, 0.11, and 0.42 corresponds to Fe occupancies of $n_{\rm Fe} = 0.67$, 0.59, and 0.39. We see that T_g decreases with decreasing concentration of magnetic ions—$T_g = 55$ K,

27 K, and 18 K, for $p = 0$, 0.11, and 0.42 respectively. While a decrease in T_g with decreasing concentration of magnetic ions is also seen in conventional SG systems such as *Cu*Mn [7], *Au*Fe [8, 9], and $Eu_xSr_{1-x}S$ [10], the increase of $\chi\,(T_g)$ with decreasing T_g, $d\chi\,(T_g)/dT_g < 0$, is accounted as an effect of vacancy defects on GF magnets [11]. We note here that although the nature of short-range order in the present system, and GF materials in general, is not fully understood, Syzranov has provided a mechanism for the observed behavior of $\chi\,(T_g)$ with dilution in the [11]. As discussed in [4], such behavior is, however, a distinct signature of the SG transition in all other GF for which $f \geqslant 10$. The inset of figure 2 shows the inverse susceptibility for the same compounds where, as previously indicated, the $p = 0.11$ and 0.42 data have been scaled to exhibit the same slope and thus the same μ_{eff} as the $p = 0$ data. To further confirm the SG behavior in doped compounds, we compare the temperature dependent AC susceptibility of p =0, 0.11, and 0.42 around the normalized spin-glass freezing temperatures (i.e. T/T_g) in figure 1(b). All three samples have a shift in T_g to higher temperature with increasing frequency, consistent with behavior in conventional SGs. Indeed, we observed an increasing frequency dispersion (i.e. a larger shift in the normalized T_g) as the value of p was increased. Previous work on the AC susceptibility of Fe_2TiO_5 indicated that the frequency dependence of T_g follows a Vogel–Fulcher law [12, 13].

In figure 3 are shown specific heat data for $p = 0$, 0.11, and 0.42 over different temperature ranges. In figure 3(a), $C\,(T)\,/T$ versus T up to 300 K is plotted per 'mole-formula unit', appropriate for comparing the main contributions, namely lattice and magnetic excitations. These data show a broad hump centered at approximately 140 K, the high-temperature decrease of which indicates an approach on warming into the Dulong–Petit region. The data in this frame are mainly due to lattice excitations, as can be judged from the area of the shaded region which is equal to the total magnetic entropy for $p = 0$ and approximately 10% of the total entropy from $T = 0$–300 K. As the Fe concentration decreases, one can see the height of the broad peak decrease slightly while the entropy below 50 K increases. This entropy shift may be indicating that, as Fe site-occupancy decreases, a greater fraction of spins find themselves in the low-energy region of the rough energy landscape associated with random freezing. In figure 3(b) are shown data at intermediate temperatures. Here, the differences among the data sets reflect the entropy transfer mentioned above. Finally, in figure 3(c) are shown $C\,(T)\,/T$ for T down to 200 mK, below which an additional, possibly nuclear, contribution appears as a rise in $C\,(T)\,/T$ on decreasing T for the last few points, especially noticeable for $p = 0.42$. Taking this contribution into account, the data are consistent with a linear term that is too small to be unambiguously identified. More apparent is a temperature dependence below $T = 3K$ that is consistent with $C\,(T) \propto T^2$, behavior which has been seen in several other strongly GF systems [14–17]. While the precise origin of the $C\,(T) \propto T^2$ behavior is not well understood [17], it is reasonable to ascribe the associated excitations to inter-surfboard spins, and not to intra-surfboard spin waves, since these are

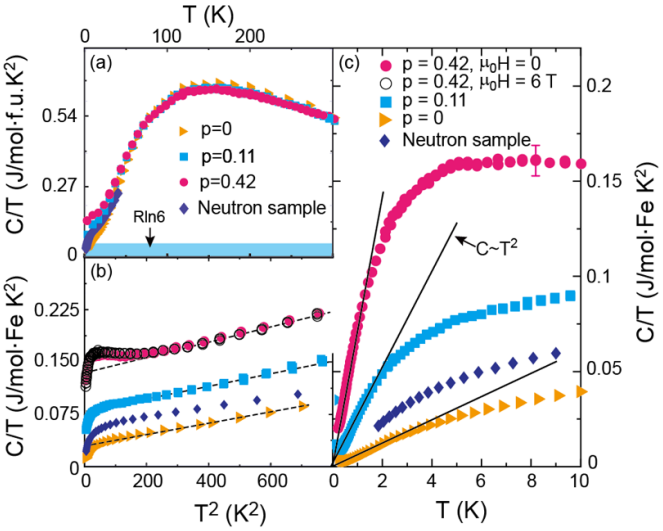

Figure 3. Specific heat divided by temperature of $(Fe_{1-p}Ga_p)_2TiO_5$ for various values of p. (a) $(T)/T$ vs. temperature for $p = 0$, 0.11, and 0.42, each showing a broad maximum at $T \approx 140$ K, which results from a dominantly phonon contribution to $C(T)$. The low temperature shoulder is a consequence of a magnetic contribution. (b) $(T)/T$ vs T^2 in the intermediate temperature regime shows a crossover from magnetically dominated region (low T) to the phonon dominated region where $C(T) \propto T^3$ expected for phonons. (c) $(T)/T$ vs T for $T < 10$ K, showing that for each composition $C(T) \propto T^2$ for $T < 3$ K. The crystal labeled 'neutron sample' was a measurement of a small single crystal grown in the same floating zone furnace with the same nominal composition as that as the crystal measured by neutron diffraction.

expected to follow $C\,(T) \propto T^3$, albeit with a strong finite-size effect, which should produce a gap in the spin wave spectrum. In general, the specific heat of a disordered spin system may come from itinerant spin modes, such as Halperin-Saslow [18, 19] modes or spin–lattice 'photons' [20–24], or from localized excitations [19]. Such itinerant modes can result in the $C\,(T) \propto T^2$ in an effectively 2D, e.g. layered, system. In 3D, however, they will lead to the $C\,(T) \propto T^3$ temperature dependence, inconsistent with our observations here.

It is reasonable, therefore, to ascribe the observed quadratic specific heat to localized excitations among inter-surfboard spins. The quadratic dependence of the specific heat requires that the density of states (DoS) of these excitations (the density of levels per unit energy in an ensemble of excitations), $\nu\,(E)$, be linear in energy E. As shown in [25], in a strongly disordered medium with short-range interactions, a linear-in-E DoS is a universal feature of excitations corresponding to shifting any conserved quantity between nearby sites (quasi-localized states). The role of such a quantity may be played by spin density or 'charge' in the Coulomb phase [20, 21, 26–28] of a GF magnet. The corresponding mechanism requires that the shifts of the conserved quantity do not become arbitrarily large (which is possible in metastable states such as SGs). Because the DoS of excitations is field-independent in strongly disordered systems, the respective contribution to the specific heat will be field-independent, as is observed between $H = 0$

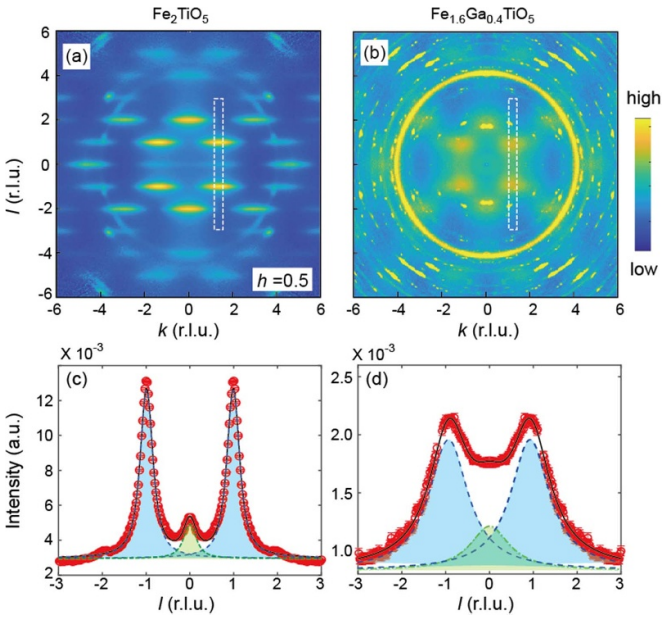

Figure 4. Total neutron scattering intensity in the [0.5, *k*, *l*] planes for Fe_2TiO_5 at $T = 5$ K (a) and $Fe_{1.6}Ga_{0.4}TiO_5$ ($p = 0.20$) at $T = 6$ K (b). (c) and (d) Total neutron scattering intensity along the *l*-direction as indicated by the dashed rectangle in (a) and (b), respectively. The intensity was fitted with three Lorentz functions convoluted with a Gaussian resolution function. The three Lorentz peaks are set to have the same HWHM whose reversal is the correlation length along the *l*-direction and is estimated to be 3.0 ± 0.1 Å at 6 K and 2.7 ± 0.1 Å at 50 K for $Fe_{1.6}Ga_{0.4}TiO_5$ and 10.9 ± 0.15 Å at 5 K and 9.6 ± 0.15 Å at 60 K for Fe_2TiO_5.

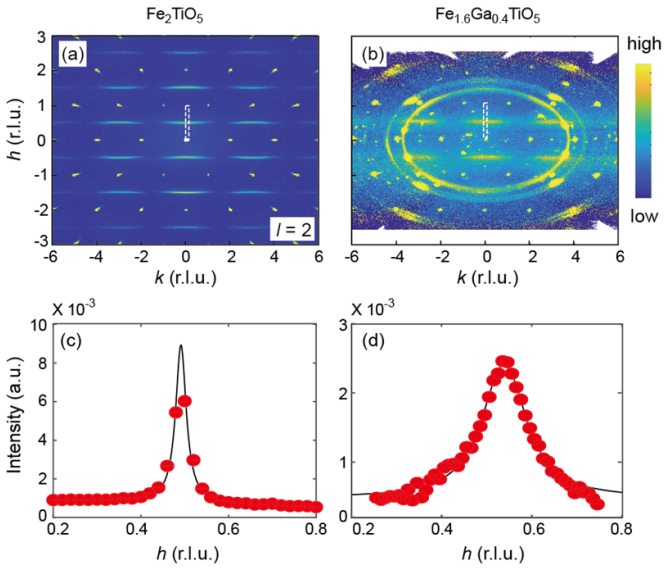

Figure 5. Total neutron scattering intensity in the (*h*, *k*, 2) planes for Fe_2TiO_5 at $T = 5$ K (a) and $Fe_{1.6}Ga_{0.4}TiO_5$ at $T = 6$ K (b). (c) and (d) Total neutron scattering intensity along the *h*-direction as indicated by the dashed rectangle in (a) and (b), respectively. The intensity was fitted with single Lorentzian functions convoluted with a Gaussian resolution function. The correlation length along the *h*-direction is estimated to be 10.8 ± 0.7 Å at 5 K and 7.5 ± 0.6 Å at 50 K for $Fe_{1.6}Ga_{0.4}TiO_5$ and 40.5 ± 1.0 Å at 5 K and 32.1 ± 0.9 Å at 60 K for Fe_2TiO_5.

and 6 T for the $p = 0.42$ compound, shown in figure 3(c). We leave, however, an investigation of the exact nature of the corresponding excitations for future studies.

Despite similarities with other GF systems, an important distinction can be drawn since, in $(Fe_{(1-p)}Ga_p)_2TiO_5$, the degrees of freedom that undergo SG freezing are postulated to be the surfboard-shaped ordered regions seen in neutron scattering [2]. The size of these regions in the undoped limit ($p = 0$) is approximately 40 Å × 3 Å × 10 Å, and thus an order of magnitude larger in the *a*-direction than the size of typical vacancy-induced quasi-spins which are thought to undergo freezing in other GF systems [29, 30]. Indeed, the large surfboard size, coupled with the observation that the ordered intra-surfboard spins point along the ***a***-axis, whereas spin freezing is seen only along the ***c***-axis, suggested a surfboard-surfboard interaction induced by fluctuations in the magnetization that are transverse to the spin ordering direction in the surfboards [2]. Below we describe the evolution of surfboard size and shape with Fe^{3+} dilution.

Comparison of the magnetic diffuse neutron diffraction patterns of the $p = 0$ and $p = 0.20$ is shown in figures 4 and 5. Figures 4(a) and (b) contrast the data measured in the ($1/2$ *k l*) plane for these two compositions. The peaks in scattering intensity are highly elliptical for $p = 0$, being broad along *k* and sharper along *l*, which reflects the differing correlation lengths along these two directions. Correlations along ***b*** are limited to nearest neighbors; however, along ***c*** the correlation length is about 11 Å as determined from the inverse of the half-width-half maximum of Lorentzian fits of a cut through the peaks along *l* as shown in figure 4(c). These results are consistent with our previous results for $p = 0$ [2]. For $p = 0.20$, the peaks occur at the same positions in reciprocal space as for $p = 0$, indicating the continued presence of surfboard-shaped correlation regions for smaller Fe site occupancy. The peak widths along *k* indicate that the correlations are similarly nearest neighbor-limited along ***b***. However, the peaks are significantly broader along *l* for $p = 0.20$ then for $p = 0$, and the Lorentzian fits shown in figure 4(d) indicate that the correlation length is significantly reduced at $T = 5$ K to only ~ 3 Å, approaching the correlation length along ***b***. Further, it is quite apparent that the peaks in the (*h k* 2) planes (figures 5(a) and (b)) are broader for $p = 0.20$ than for $p = 0$. Fits along *h* indicate that, despite considerable reduction induced by Ga dopants, the correlation length along ***a*** for $p = 0.20$ at 5 K remains significantly longer than the lattice constant, with a value of ∼11 Å, resulting in regions with a size ∼11 Å × 3 Å × 3 Å. Thus, while the aspect ratio of the correlated regions remains surfboard-like, the shape has become more prolate than for $p = 0$.

4. Discussion

The decrease in surfboard size (magnetic correlation length) with increasing *p* is consistent with the associated decrease

in T_g: smaller surfboard sizes correspond to smaller fluctuations of the magnetization on the surfboard, weaker interactions between the surfboards and, concomitantly, lower glass transition temperature T_g (in accordance with the quantitative description developed in [2] for the $p = 0$ case). Increasing the density of Ga^{3+} ions, or non-magnetic vacancies, leads to the existence of fewer regions of large-enough Fe^{3+} density to support the surfboard-like correlations, in addition to fewer Fe^{3+} in the regions between surfboards. Both of these effects combine to reduce the inter-surfboard interactions leading to a reduced T_g. An accurate quantitative description of the behavior of T_g will require the determination of the inter-surfboard distance and other microscopic details whose investigation we leave for future studies.

We noted already that the behavior of the magnetic susceptibility and the glass transition temperature follow the trend $d\chi(T_g)/T_g < 0$ that has been shown [4] to exist in all strongly GF magnets where T_g is modified by vacancy density. This trend can be re-expressed as the growth of the glass-transition temperature with *decreasing* vacancy density towards a finite value in the limit of zero vacancies. This finite temperature, T^*, has been called the 'hidden energy scale' [4] because it has not been addressed in microscopic theories of GF systems. This energy scale cannot be approached closely in $(Fe_{(1-p)}Ga_p)_2TiO_5$ due to the different valence states of Fe and Ti, but extrapolates to $T^* \sim 100\,K$, a value far higher than seen in other strongly GF systems where $5\,K < T^* < 20\,K$. This difference may be due to (1) the very large θ_W found for Fe_2TiO_5 and (2) the difference between the freezing degrees of freedom. In Fe_2TiO_5 these are fairly large objects, compared to an atomic spin, and their moment is induced. In the other GF systems, these are quasispins, or, the region around a vacancy and their moment is the distortion in the local spin arrangement caused by this vacancy. Despite these differences, it is reasonable to discuss $(Fe_{(1-p)}Ga_p)_2TiO_5$ in the same framework as the other strongly GF systems since it exhibits the characteristic behavior $d\chi\,(T_g)\,/dT_g < 0$.

5. Summary

In summary, we have measured $\chi\,(T)$, $C\,(T)$, and total neutron scattering in single crystals of $(Fe_{(1-p)}Ga_p)_2TiO_5$, for $p > 0$. We find that $d\chi\,(T_g)\,/T_g < 0$ as the SG T_g is made to decrease with increasing p, similar to behavior seen in other GF magnets. The model proposed to explain the unusual anisotropy of the freezing anomaly at T_g for $p = 0$ is validated for $p = 0.20$, in which correlated regions, albeit smaller than seen for $p = 0$, are observed with a concomitant reduction in T_g.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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