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## ABSTRACT

Magnetic skyrmions are topologically protected spin textures with promising prospects for applications in data storage. They can form a lattice state due to competing magnetic interactions and are commonly found in a small region of the temperature–magnetic field phase diagram. Recent work has demonstrated that these magnetic quasi-particles fluctuate at the  $\mu\text{eV}$  energy scale. Here, we use a coherent x-ray correlation method at an x-ray free-electron laser to investigate these fluctuations in a magnetic phase coexistence region near a first-order transition boundary where fluctuations are not expected to play a major role. Surprisingly, we find that the relaxation of the intermediate scattering function at this transition differs significantly compared to that deep in the skyrmion lattice phase. The observation of a compressed exponential behavior suggests solid-like dynamics, often associated with jamming. We assign this behavior to disorder and the phase coexistence observed in a narrow field-window near the transition, which can cause fluctuations that lead to glassy behavior.

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The modern theory of continuous phase transitions and critical phenomena is the linchpin of statistical mechanics and condensed matter physics. At the critical point, mean-field theory breaks down and the microscopic fluctuations become important at many length and time scales. This description originated from the study by Landau *et al.*<sup>1</sup> and was later reformulated by Wilson using renormalization group theory, a formal prescription for handling critical singularities.<sup>2</sup> Since the fluctuations are what drives the physics near these mathematical singularities,

the quantitative understanding of fluctuation phenomena has marked a great success in physics.

While the scaling of the fluctuations is well understood in the case of continuous transitions, first-order transitions do not typically exhibit critical fluctuations. In this case, the system does not go through a critical point but rather exhibits a singularity in a thermodynamic variable at the phase transition. Indicators of this are typically latent heat, hysteresis, and phase separation produced

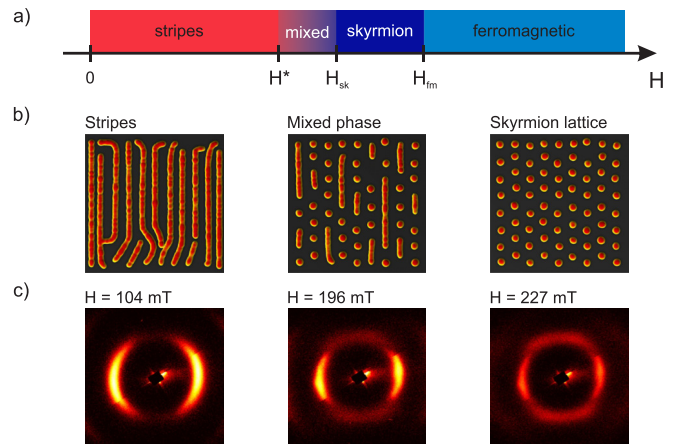
by the formation of stable nuclei. Interestingly, fluctuation-induced first-order phase transitions have been theoretically predicted however.<sup>3,4</sup> It was proposed that excessive critical fluctuations may lead to a first-order transition before reaching the critical point, thereby modifying an expected continuous transition to become first-order. Early experimental examples involve the nematic-to-smectic transition in liquid crystals,<sup>3,5</sup> and theoretical works have indicated that this mechanism could be of importance in superconducting transitions.<sup>6,7</sup>

Recently, fluctuation-induced first-order transitions have also been reported in skyrmionic systems.<sup>8–10</sup> The presence of such transitions seems to be a common feature to these chiral magnetic materials and calls for a detailed investigation of the fluctuations near discontinuous phase transitions in these systems.<sup>11</sup> One topic of interest, especially as it relates to the skyrmion lattice phase, is how topology affects the formation, fluctuation, and behavior of the skyrmion lattice where the thermodynamic free energy becomes non-analytic. This represents a vast and unexplored area of research in topological systems. While the inherent features of the fluctuation-induced discontinuities are important, their investigation nevertheless remains scarce, especially experimentally.

Here, we address this by reporting a study in the coexistence region at the phase boundary between the skyrmion lattice and ferromagnetic stripe phases in amorphous Fe–Gd thin films. We use short-pulsed coherent x-rays to detect spontaneous fluctuations in this region, giving direct insights into the temporal nature of how correlations decay at a first-order phase transition. The intermediate scattering function is shown to follow a compressed exponential relaxation, which could be due to either jamming or “glass-like” behavior found in the phase coexistence region.

Skyrmions are stable topological objects that were theoretically proposed in 1962 by Skyrme.<sup>12</sup> Although they were initially proposed as a model for subatomic structures, they have become, after the observation of magnetic skyrmions in MnSi a decade ago, a main focus in condensed matter physics.<sup>13,14</sup> Since then, skyrmion systems have indeed attracted much interest, in light of their potential use in spintronics and data storage applications.<sup>15–19</sup> Recent work has demonstrated that fluctuations of the skyrmion lattice take place on the  $\mu\text{eV}$  energy scale,<sup>20</sup> but how this changes near a discontinuous transition is still not known.

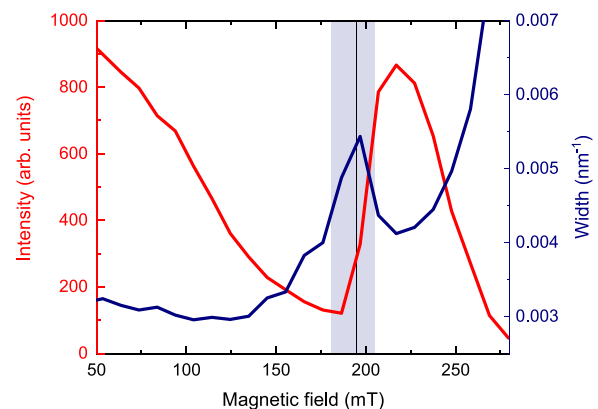
The system studied here is an Fe–Gd thin film heterostructure. The pronounced perpendicular magnetic anisotropy favors the emergence of exotic magnetic phases, such as stripes or skyrmions. The experiment was performed at the Linac Coherent Light Source (LCLS) x-ray free-electron laser<sup>21</sup> (FEL) using resonant magnetic soft x-ray scattering<sup>22</sup> to directly probe the different magnetic phases. A unique double electron bunch scheme is used to generate pairs of x-ray pulses with tunable delay in the ns range<sup>23</sup> (see the [supplementary material](#)). A one-dimensional schematic of the room temperature phases is shown in Fig. 1(a). The transition between the stripe and skyrmion phases occurs over a finite range of applied fields between approximately 180–205 mT, where the two phases coexist, possibly indicative of a disorder-broadened first-order transition.<sup>24</sup> The transition between the different phases can be determined from the intensity and the width of the weaker skyrmion x-ray scattering peaks, as shown in Fig. 2. In particular, the peak in intensity in this region signals the presence of the long-range ordered skyrmion lattice, and the phase



**FIG. 1.** Fe–Gd thin film heterostructure room temperature phases. (a) Sketch of the phase diagram at room temperature with a magnetic field.<sup>25,26</sup> The critical fields are  $H^* = 180$  mT,  $H_{sk} = 205$  mT, and  $H_{fm} \approx 265$  mT. The experiment reported here was performed at  $H = 195$  mT, well in the mixed phase region, where both the skyrmion lattice and the stripes coexist. (b) Sketch of the stripe, mixed, and skyrmion lattice phases in real space, based on simulations from previous work.<sup>26,27</sup> (c) Measured data of the soft x-ray FEL diffraction patterns at the Gd  $M_5$ -edge showing the stripe, mixed, and skyrmion lattice phases in reciprocal space. Because each image is the sum of about 500 shots, the speckle is not visible in these average images.

coexistence region can be identified by the broadening of the peak, indicative of a reduction of the correlation length. The intensity at lower field values is due to the tail of the strong stripe peaks (see Fig. 1), and the transition to the ferromagnetic state above 250 mT is shown by the decline of the peak intensity and the divergence of the peak width.

The stripe and skyrmion phases can be identified by their resonant diffraction pattern at the Fe  $L_3$  or Gd  $M_5$  edge [see Fig. 1(c)].<sup>25</sup> Traditionally, a change from a twofold symmetry, characteristic of the one-dimensional nature of the ferromagnetic stripe, to the sixfold symmetry of the hexagonal skyrmion lattice occurs in the transformation

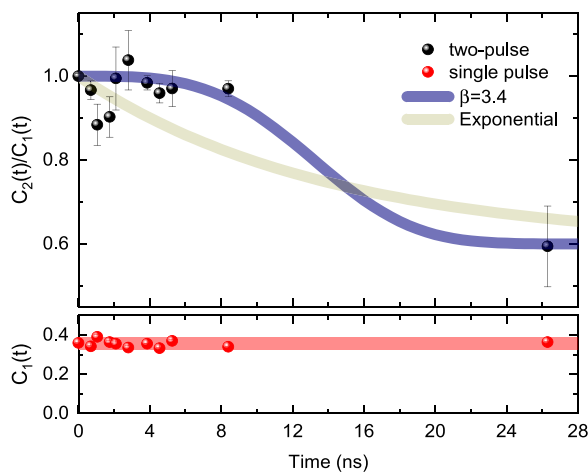


**FIG. 2.** Intensity and radial width of one of the skyrmion peaks as a function of the applied magnetic field. The shaded area indicates the stripe-skyrmion phase coexistence region, and the vertical black line indicates the field for which the fluctuations are measured.

to the skyrmion phase. Importantly, the intensity distribution among the different peaks in the mixed phase region of the phase diagram retains the twofold symmetry of the stripe phase. Deep in the skyrmion phase of this system, however, this asymmetry is not due to the phase coexistence with the stripes but rather the presence of skyrmion bound pairs, breaking the perfect sixfold symmetry in the skyrmion lattice phase.<sup>25</sup> With a proper magnetic field application procedure, the lattice of skyrmion bound pairs, or the so-called “bi-skyrmion” phase, can be formed in this material. This phase has been studied in great detail in other systems, and it has been shown that it can be electrically driven with orders of magnitude lower current density than that for the conventional ferromagnetic domain walls.<sup>28</sup>

In the bound-pair, or bi-skyrmion, lattice phase, referred from this point simply as the skyrmion lattice phase, the contribution of each phase is somewhat difficult to disentangle. In the phase coexistence region, the stripe peaks are overlapped with the stronger skyrmion reflections. In the transition region, which is the focus of this study, we are thus sensitive to both order parameters simultaneously when measuring the stronger peaks. While this can be a problem for some experiments, here this enables the study of the fluctuations arbitrarily close to the transition, as the signal from each phase can be comparable when approaching the first order phase transition.

The contrast function  $C(q, t) = C(t)$  for the stronger peaks in the mixed phase is shown in Fig. 3 for integrals over the Bragg peak with a reasonable intensity. These are measured by extracting the number of degrees of freedom from the speckle pattern formulated under photon counting conditions.<sup>29</sup> This contrast function has been shown to provide direct information on the stochastic fluctuations in the system and is related to the intermediate scattering function, as measured with traditional or serial x-ray photon correlation spectroscopy.<sup>30</sup>



**FIG. 3.** Fluctuations in the stripe-skyrmion mixed phase. Top: normalized two-pulse contrast  $C_2(t)/C_1(t)$ . Slower, compressed exponential dynamics are observed and differ significantly compared to the pure exponential decay reported in the middle of the skyrmion phase.<sup>20</sup> Bottom: single pulse contrast  $C_1(t)$ . The time axis for this curve indicates the setting of the accelerator configuration, which generates the pulse separation of each pair and serves to highlight the stability of the beam parameters and the resilience of the sample under prolonged exposure.

A reference single-pulse measurement  $C_1(t)$  is shown in the bottom panel of Fig. 3. This is the contrast function measured using single-pulse measurements only, for different separation time configurations of the accelerator. In addition to demonstrating the stability of the beam properties,  $C_1(t)$  also serves to establish that the sample response is not affected under prolonged exposure.<sup>31</sup>

The top panel in Fig. 3 shows the normalized double pulse contrast, which encodes the fluctuations at the discontinuous phase transition. Interestingly, this normalized contrast function, or normalized intermediate scattering function, does not start decaying until 5–10 ns. Hence, the data cannot be modeled by a simple exponential decay, characteristic of diffusive behavior. This is in stark contrast to the pure skyrmion phase where the relaxation follows an exponential decay.<sup>20</sup> Furthermore, the sample reaches full decorrelation over the short span of tens of nanoseconds. The contrast can only decay to a minimum of  $C(t) = 3/5$  when using a two-pulse method in a mixed phase, where two different types of components contribute to the scattering (see the [supplementary material](#)). So, although the skyrmion lattice phase has been shown to exhibit a static or frozen-in component at ns levels for some regions of the lattice phase,<sup>20</sup> this is not the case here in the mixed phase. For the state near the first order phase transition, the fluctuations are more pronounced and occur over the whole probed volume.

A delayed response such as that observed here can be reproduced by a Kohlrausch–Williams–Watt (KWW) function,

$$S(q, t) = A_0 + A_1 \exp(-[t/\tau_0]^\beta), \quad (1)$$

with a compressed exponent,  $\beta > 1$ . In order to reflect the theoretical minimum contrast value, the background constant  $A_0$  is constrained between 0.6 and 1 (see the [supplementary material](#)). The optimum fit gives a quite high value for the compressed exponent  $\beta = 3.4$ , which captures best the delayed decay of the intermediate scattering function. This is shown together with an exponential fit to the data in Fig. 3.

While the stretched exponential ( $\beta < 1$ ) behavior is typically found in liquid-like systems, compressed exponentials are typically found in collective, solid-like dynamical systems.<sup>32–38</sup> This anomalous diffusion can be accounted for by continuous time Levy flights—random walks with heavy-tailed probability distributions—which can lead to both stretched and compressed relaxation.<sup>37</sup> The common observation of such relaxation in widely different systems suggests a generic underlying mechanism, yet the microscopic origin of such relaxation is still poorly understood.<sup>37,38</sup> For instance, recent work on the microscopic role underlying compressed exponential, or super-diffusive, behavior found that  $\beta$  was strongly dependent on the deformation properties in colloids,<sup>39</sup> which could have analogous behavior in topological systems.

Other systems that show compressed exponential behavior are found in the context of jamming.<sup>32,35,36</sup> Jamming is known to describe the universal behavior of complex systems under stress, such as those found in glasses, colloid systems, and frustrated magnets.<sup>40–43</sup> It is usually due to close-packing in systems where limited or non-Gaussian dynamics is possible and is often found with the values of the exponent  $\beta$  close to 1.5. For instance, the elastic relaxation of internal stress in a variety of soft materials, such as colloidal gels, seems to play an important role in aging properties and has been shown to explain the compressed relaxation of the dynamical structure factor.<sup>33</sup> A fit with the compressed exponent  $\beta$  fixed to 1.5 (not shown) also can describe



the data reasonably well with a decorrelation time of the same order, indicating that jammed dynamics may explain the decay observed here.

Although this is a study of the inherent, spontaneous fluctuations, the question arises as to whether other types of fluctuations could be responsible for the dynamics observed here with compressed exponential behavior. For instance, a flow of scatterers due to temperature or field gradients was shown to yield, in the simplest case, a Gaussian decay (KWW with  $\beta = 2$ ).<sup>38</sup> Drifts of the skyrmion lattice with velocities of the order of  $10^4 - 10^5 \text{ nm s}^{-1}$  have been observed under the application of low electric currents.<sup>44,45</sup> However, over the tens of ns considered here, this corresponds to distances orders of magnitude smaller than the skyrmion diameter, and such small motion is unlikely to affect the speckle pattern in a significant way. Thus, the stochastic fluctuations at the discontinuous phase boundary are much more significant than that detected in driven skyrmion systems.

Alternatively, the behavior discovered here could also emerge when a system exhibits a reasonable amount of disorder in the mixed phase region. Disorder is known to exist in this system and would exhibit slow relaxation as seen near a glass-like transition. It is possible that the KWW behavior is due to this glass-like nature that occurs in the mixed phase region where the competition between the two different states is at play. Although glasses are typically discussed in terms of stretched exponential behavior,<sup>46</sup> structural and metallic glasses do indeed also exhibit compressed exponential behavior.<sup>48</sup> Further work is needed, however, to conclusively determine which model can describe the physics of the fluctuation-induced 1st order phase transition in this skyrmion system. In particular, future studies that focus on the  $q$ -dependence are needed, which will allow us to unravel the mechanisms at work here. A linear  $q$ -dependence of the relaxation rate  $\tau$  is expected in the jammed system,<sup>35</sup> while de Gennes narrowing<sup>47</sup> would be featured if the transition is glass-like.<sup>48</sup>

Next-generation x-ray free electron lasers with the increased repetition rate will present the opportunity to unravel these differing models in topological magnets in the future. In order to remain in a non-perturbative regime, the x-ray pulses are heavily attenuated, preventing the opportunity to take full advantage of the high, single-pulse intensity of the source. With such a physical limitation, the signal-to-noise is mainly limited by the total number of pulse pairs that can be measured in an experimentally reasonable time. These types of experiments will thus profit tremendously from the increased repetition rate of these novel light sources beginning to come online.

We report a study of fast, magnetic fluctuations in the phase coexistence region at a first-order transition in a topological skyrmion system. The observation of a compressed relaxation was interpreted as indicative of a jammed or glassy state in this region of the phase diagram. It appears plausible that the limited volume available for each phase in the phase coexistence region limits their dynamics, which could lead to the jammed state. Further verification is, however, necessary to resolve if this indeed could represent a *skyrmion glass*. These observations naturally raise the question of the universality of this phenomenon and whether the observed behavior is a general feature in fluctuation-induced first-order phase transition phenomena.

See the [supplementary material](#) for experimental details, data analysis, and discussion of speckle statistics.

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The data that support the findings of this study are available from the corresponding author upon reasonable request.

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