Frustration-induced spin wave anomaly and dimension instability in FeGe$_2$

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Abstract

Inelastic neutron scattering was used to probe the spin dynamics of the long-range magnetic structure of single crystal FeGe$_2$. An unusual checkerboard-shaped anomaly located far from the magnetic satellite peaks of the incommensurate order was found in the magnetic dynamical structure factor. Using expanded ranges of wave-vector and energy transfer, we are able to develop a new model Hamiltonian that includes a heretofore unconsidered intraplane next-nearest neighbor interaction. The checkerboard-shaped anomaly is attributed to the near-perfect magnetic frustration in FeGe$_2$, which also facilitates the emergence of unexpected two-dimension magnetic order in the system.

Introduction

FeGe$_2$ has the same body-centered tetragonal crystal structure [1] (S.I. FIG. S1) as $\theta$-phase Al$_2$Cu with the space group $I4/mcm$. It exhibits two zero-field magnetic phase transitions as a function of cooling [2,3]: one is a second order Néel transition from paramagnetic phase to an incommensurate spin-density-wave state at 289 K, and the other is a first-order transition from an incommensurate state to commensurate antiferromagnetic state at 263 K. The ordering wavevector of the commensurate state $(2\pi/a)[1, 0, 0]$ changes to $(2\pi/a)[1+\delta, 0, 0]$ in the incommensurate state, where $\delta$ varies from 0 to 0.05. Along the $c$ axis, the nearest-neighbor distance between Fe atoms is 2.478 Å, which is quite close to that of elemental Fe (~2.482 Å), such that ferromagnetic exchange interaction $J_c$ is expected to be strong. In the $ab$ plane, only a weak nearest-neighbor antiferromagnetic exchange interaction $J_1$ has previously been considered [4,5]. While the spins of iron were known to lay on the basal plane, their exact orientation is still unclear. The magnetic moment is expected to be $\mu = g\sqrt{S(S+1)}\mu_B = 2.8\ \mu_B$ / Fe, assuming no electron transfer between Fe and Ge atoms and a low-spin configuration in a tetrahedral environment with spin $S = 1$ and g-factor of 2. However, the value was found to be $1.2 \pm 0.1\ \mu_B$ / Fe from a powder neutron study, explained as arising from hybridization of the 3d and 4s orbitals of the Fe atoms [6].

The spin wave along $[H, 0, 0]$ was previously measured using a thermal triple-axis neutron spectrometer at the Chalk River reactor [4], and it was found that the dispersion rises to a maximum energy of 26.5 meV at the zone boundary. This work was followed by a study of the high-energy spin wave along $[0, 0, L]$ using the direct geometry time-of-flight spectrometer HET at ISIS [5]. As a result, a nearest-neighbor (NN) Heisenberg model was proposed with $S_{j_c} = 136$ meV and $S_{j_1} = -8.8$ meV, where $S$ is the on-site spin magnitude.

Here we provide a combined scattering and computational study of FeGe$_2$ in examination of a portion of the magnetic spectrum that was not previously identified. We use inelastic neutron scattering (INS) to examine the lattice and magnetic dynamics of FeGe$_2$ over a range of temperatures. Phonons at finite temperatures were calculated from atomistic calculations and the results are in good agreement with experiments. The spin waves were simulated by classical linear spin wave theory (LSWT); however, an
Additional intraplane next-nearest neighbor interaction $J_2$ is found to be required to fully describe the spectrum. The excellent agreement between experiment and the new model corroborates the near-perfect magnetic frustration in FeGe$_2$.

Results

FIG. 1 shows a sample of the inelastic neutron scattering data acquired as a function of both temperature and energy transfer. FIG. 1(a)-(b) shows the elastic scattering in the (HK0) plane as a function of...
temperature. At 20 K, one sees the diffraction pattern with several aluminum powder line rings visible. As the temperature increases, an anomalous feature can be observed clearly in the HK0 slices of dynamical structural factor (FIG. 1 and S.I. FIG. S2). Besides the expected nuclear and magnetic Bragg peaks, there is extra intensity connecting nearest neighbor magnetic Bragg peaks. This intensity forms a checkerboard arrangement with rods along the X-M directions (see the Brillouin Zone map in S.I. FIG. S1), as depicted in the 3D rendering at L = 0 (FIG. 1(l)). This checkerboard-shaped anomaly is found in both elastic and inelastic scattering slices and does not strongly depend on energy transfer. At 20 K, such intensity can only be observed at finite energy transfers and appears to be detached from M points, forming a dot-dash-dot pattern (FIG. 1(c)). TAX data at 8 meV shows that each dash consists of two sections, which merge with the magnetic peaks at M points as the temperature increases (FIG. 1(e)-1(i)).

The extent of the phonon and magnetic excitation spectrum can be assessed by examining the scattering intensity as a function of energy and wave-vector transfer along the X-M direction for T = 20 K and T = 300 K as shown in FIG. 2(a) and 2(d) respectively. Magnetic excitations emerge from the M points and disperse up to approximately 30 meV. The spin-waves return to the elastic line albeit with much weaker intensity at wavevectors near X points. As wave-vector increases, the magnetic form factor causes the spin wave scattering intensity to decrease. At these larger wave-vectors, one can see the optic phonon excitations between approximately 15 and 35 meV.

Phonon simulations were performed using finite temperature effective force constants. The simulated phonons shown in FIG. 2(b) and 2(e) match well with experimental data. The magnetic excitations dispersing out of M points were reproduced by LSWT using the NN model. This is shown in FIG. 2(c) for SJ values from literature [5]. These values reproduce the gross features of the magnetic spectrum, although the spin waves measured appear steeper in the vicinity of the magnetic zone center.
There are important differences between the measurement and simulation. The main difference is that the continua-like intensity between neighboring M points is not reproduced in the NN model. This intensity is associated with the anomalous checkerboard intensity observed in the HK0 planes. Although over-damped, this intensity can be resolved at 20 K as collective dispersive excitations, which reach the minimums at X points. At 300 K, this dispersive excitation is further damped and merges to a broad response across a wide energy range along rod directions in the Brillouin zone. Spectral weight perpendicular to this direction (Γ-X) is weak and the dispersion is very steep around X points, (S.I. FIG. S3).

Discussion

To reveal the origin of the checkerboard anomaly, a Q-dependence analysis was performed. Upon examination of the ARCS data, it is found this anomalous intensity only appear where L is even, following the same behavior that is predicted for magnetic Bragg peaks (FIG. 1(k)). This observation indicates that the order behind the anomaly has the same periodicity along the c axis as the magnetic structure of FeGe2. To quantify the structural factor of the anomalous INS intensity, equivalent points of [1.3, 0.3, 0] in wave-vector space were selected on each rod at elastic (0 meV) and inelastic (8 meV) HK0 slices and their intensity was plotted as a function of |Q|. After excluding the points affected by background from sample environment, the data was compared to the function $A F^2(Q) + B$, where $F^2(Q)$ is the square of magnetic form factor of Fe atom, A and B are constants to be fit. Fe atom was used here instead of ionic forms since no electron transfer was found in the Bader analysis results. FIG. 3 shows the excellent agreement to the experimental data, further confirming that the anomalous intensity, in both the elastic and inelastic scattering, is indeed from magnetic excitations. At 20 K and 0 meV, as shown in FIG. 3(a), the intensity is dominated by the background, consistent with the fact that no rod intensity was observed in the low temperature elastic scattering.

Contributions to the checkerboard anomaly from other sources including electronic scattering or leftover incommensurate order were considered, but ultimately excluded. An scattering process, such as observed in CePd$_5$ [7], was considered. However, in order to produce the checkerboard-shaped pattern in elastic scattering, a specific geometry of the Fermi surface is needed to create the required electronic scattering channels. A non-magnetic Fermi surface calculation of FeGe2 was performed to show that the Fermi surface does not have this shape (S.I. FIG. S4). Additionally, the intensity anomaly cannot be explained by leftover incommensurate order either. The magnetic satellites intensity from incommensurate state, if exists, will be along different directions in the reciprocal space. Besides, the maximum value of $\delta$ is 0.05, which is too smaller to account for the anomaly far away from M points.
The NN model used previously to describe FeGe₂ includes the strong FM interaction J_c in the c direction and the nearest-neighbor AFM interaction J_1 in the plane but is insufficient to describe the anomalous intensity observed here. As shown in FIG. 2(c), the NN model predicts spin waves with a lower slope near zone centers, and it fails in reproducing the anomalous excitation observed between X and M. The former is consistent with the statements from earlier reports [4], but the latter is of more interest in this context.

A Checkerboard-shaped anomalous excitation without periodicity along [0, 0, L] has previously been observed in quasi-2D square lattice systems with magnetic frustration, such as CaCo₂₃As₂, and is described by the J₁-J₂ Heisenberg model [8,9]. Different from FeGe₂, the interlayer NN exchange interaction J_c is ignored in these systems since it is generally much weaker. Besides the NN exchange interaction J₁, the intraplane next-nearest-neighbor (NNN) exchange interaction J₂ is also important in describing the excitations in these systems. The ground state of the system could be determined by the frustration parameter η = J₁/J₂, where J₁ could be either ferromagnetic (FM) or antiferromagnetic (AFM) but J₂ is always AFM. Néel-type, stripe-type and FM ordering occurs for η > 1, |η| < 1 and η < -1, respectively. Perfect frustration happens between Néel-type and stripe-type when η = 1 and between stripe-type and FM when η = -1. Extreme spatial anisotropy due to the perfect frustration leads to effectively 1D behavior and corresponding plane-like features in INS [10].

Identical to the just mentioned quasi-2D square lattice systems, each magnetic atom in FeGe₂ has 4 intraplane NN atoms and 4 intraplane NNN atoms. To determine the extent of the role of J₂ interaction in FeGe₂, exchange constants were calculated using atomistic simulation based on density functional theory. Total energy calculations were made on five collinear magnetic configurations and the exchange parameters were fit to these energies under the assumption that the change of energy is only dependent on the selected exchange interactions (S.I. FIG. S5 and Table S1). Two models were used in the least-square fitting: the NN model containing only J₁ and J_c, and the NNN model that also includes J₂. The results are shown in Table 1.

|          | E₀  | J₁S² | J₂S² | J_cS² | η     |
|----------|-----|------|------|-------|-------|
| NN model | -556.6166 | -7.2 | - | 99.1  | -     |
| NNN model | -556.6495 | -7.2 | -2.6 | 99.1  | 1.4   |

Table 1. Parameters obtained from total energy calculations.

For the NN model, the calculations yield |J₁/J_c| ~ 13.8, this large ratio represents the anisotropic nature of in-plane and [0, 0, L] spin waves in FeGe₂. The ratio is consistent with previous reports of 15.5 [4]. For the NNN model, J₁S² and J_cS² remain almost the same as those values determined for the NN model and J₂S² has a value of -2.6 meV. J₁ and J₂ are of the same order of magnitude, indicating that both interactions play important roles in describing the spin dynamics and should not be neglected.

Exchange parameters calculated for the NNN model were used to perform spin wave simulations of the excitations in the low temperature AFM state, as shown in FIG. 2(f). An effective value of S = 0.8 was used here to scale the energy magnitude, accounting for the itinerant nature of FeGe₂. These simulations show the importance of J₂ in reducing the magnitude of the spin wave energy of dispersion near the X points in the Brillouin zone, and significantly improving agreement with the experimental results. It should be noted that along [0, 0, L] the spin waves are very steep, so even small integration range in making slices from ARCS S(Q, E) results in the feature that appear as a continuum of excitations. This has been accounted for by using the same integration range in the spin wave simulations as the experimental data. The simulated HK0 slices at constant energies (FIG. 1(j)) show strong “rod” intensity along X-M and only weak intensity along Γ-X near the X points for a large range of energy transfer, consistent with the experiments.
As temperature increases to 300 K, the dispersive magnetic excitations become more and more diffuse and soften to lower energy transfer. Constant Q cuts were obtained from the TAX data (FIG. 4(a)-(e)), revealing a dramatic softening of the spin wave from finite energy at low temperatures to near 0 meV at 300 K as seen in FIG. 4(f) and 4(g). This effect is also visible in FIG. 4(h) showing the constant energy scans at 8 meV from the same set of data. Between 5 and 275 K, two peaks between neighboring M points can be easily resolved. Above 300 K, the peaks vanish, and the intensity becomes flat between M points. Combining these findings, we believe that the well-defined spin waves collapse near 300 K as one enters the paramagnetic phase of FeGe₂.

At the same time as the spin waves begin to be overdamped, rods of scattering between neighboring M points connect with each other and form the checkerboard-shaped pattern in HK₀ elastic slices of S(Q, E). To gain a quantitative understanding of the spatial coherence behind these rods, correlation lengths were extracted by fitting elastic cuts with a Voigt function across the rod of scattering in the basal plane (Γ-X) and across-plane (Γ-Z), as described in Supplementary Information. Along the rod direction, the almost flat intensity suggests no correlation. At 300 K, the correlation length is about 12 Å in the across-rod direction and 23 Å in the across-plane direction. In this case, 3D order breaks down with 2D correlations of plates.
along directions bisecting the $a$ and $b$ directions remaining in the paramagnetic phase. These kinds of 2D correlations in real space are surprising in a quasi-1D system, where spins are weakly coupled in the $ab$ plane with fourfold symmetry. At 500 K, the correlation lengths become 4 Å and 11 Å, respectively, indicating a simultaneous decrease of short-range order upon further warming into the paramagnetic phase.

Our calculations show that a stripe-type configuration has the next-lowest ground state energy, about 3.7 meV per atom above the Néel-type AFM structure. When the temperature increases to 300 K, thermal fluctuations become comparable to the energy difference between these two configurations and exchange interactions can no longer stabilize the Néel-type structure. It is likely that stripe-type domains start to appear and occupy nearly half of the system. In the Néel-type structure, the effective coupling along both [1, 1, 0] and [1, -1, 0] are of $\sim J_1-2J_2$, which is close to zero when $\eta \sim 1$. Once the [1, 1, 0] stripe-type domains are formed, the effective coupling along [1, -1, 0] remains nearly zero but that along [1, 1, 0] increases to $\sim J_1+2J_2$. In this case the magnetic order could be viewed as plates perpendicular to the rods along [1, -1, 0], where there are strong in-plane correlations, but the neighboring plates are nearly decoupled because of the small effective correction between plates. Since [1, 1, 0] and [1, -1, 0] are equivalent directions in the system, two types of magnetic domains are necessary to account for the rods in the checkerboard arrangement.

Conclusions

From INS measurements of FeGe$_2$, we observe anomalous excitations at low temperature, as well as a checkerboard-shaped elastic scattering pattern developing at high temperature. We showed that both phenomena share a magnetic origin and are related to the intraplane NNN interaction $J_2$. This previously ignored interaction leads directly to generating the extra spin wave pattern for a large range of energy transfer. The near-perfect in-plane frustration introduced by the NNN interaction facilitates the emergence of 2D structure at high temperature instead of the expected 1D correlations.

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Methods

A semi-cylindrical single crystal FeGe$_2$ with an approximate 15 mm radius and 40 mm length with a mass of 110 g was used for the neutron scattering measurements. The crystal, used in previous studies of FeGe$_2$, was loaned from Oak Ridge National Laboratory (ORNL) [5,11]. Inelastic neutron scattering measurements were performed using two spectrometers at ORNL.

The majority of the data presented were obtained using the time-of-flight Wide Angular Range Chopper Spectrometer (ARCS) at the Spallation Neutron Source (SNS). The sample was mounted inside a low-background electrical resistance vacuum furnace with the (HK0) crystallographic plane horizontal. The measurements were performed with an incident neutron energy of 70 meV, at 20, 300, 500 and 635 K. The crystal was rotated about the vertical axis through 120° in increments of 0.5°. Temperature dependence of phonons in FeGe$_2$ were previously reported [11].

Data reduction and analysis of the ARCS data were performed with MANTID [12]. The data were normalized by the proton current on the spallation target. Bad detector pixels were identified and masked, and the data were corrected for detector efficiency using a measurement of a vanadium standard. After data
reduction, neutron events at different detectors were combined to generate the four-dimensional dynamical structure factor $S(Q, E)$, where $Q$ is the neutron wavevector transfer and $E$ is the energy transfer. Two-dimensional slices and one-dimensional cuts were then be obtained by integrating portions of $S(Q, E)$ over small ranges.

Additional data were collected with the Triple-Axis Spectrometer (TAX) HB-3 at the High Flux Isotope Reactor (HFIR). The measurements were made between 5 and 450 K with a fixed final energy of 14.7 meV. A pyrolytic graphite (002) monochromator and an analyzer were used and the horizontal spectrometer collimation was set to 48°-40°-40°-120°. The sample was aligned roughly in the [HHL] scattering plane.

The finite-temperature force constants were calculated with the TDEP method [13–15]. In this procedure, the Born-Oppenheimer surface of FeGe$_2$ at 20 and 300 K was sampled using molecular dynamics (MD) implemented in the VASP package [16–18]. Then, the model Hamiltonian

$$
\hat{H} = U_0 + \sum_i \frac{p_i^2}{2m_i} + \frac{1}{2} \sum_{ij} \sum_{\alpha\beta} \Phi_{ij}^{\alpha\beta} u_i^\alpha u_j^\beta + \frac{1}{3!} \sum_{ijk} \sum_{\alpha\beta\gamma} \Phi_{ijk}^{\alpha\beta\gamma} u_i^\alpha u_j^\beta u_k^\gamma
$$

was fit with this surface, which is done by comparing the forces of the model and the MD at each time step and minimizing the difference.

Here, $U_0$ is the potential energy and $\Phi_{ij}$ and $\Phi_{ijk}$ are the second- and third-order force constants. The displacement of atom $i$ from ideal positions is denoted as $u_i$, its momentum $p_i$, and $\alpha, \beta, \gamma$ are Cartesian indices.

For MD calculations, the projector augmented wave (PAW) pseudopotentials [19] were used with the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional [20]. The plane-wave cutoff was 630 eV. A supercell of a $2 \times 2 \times 2$ conventional unit cell with 96 atoms was used with a $4 \times 4 \times 4$ k-point grid. Lattice constants were relaxed before MD to $a = 5.914$ Å and $c = 4.948$ Å, which are slightly larger than the results from neutron powder diffraction at both temperatures [11]. Bader analysis was performed after the relaxation. Phonopy [21] was used to simulate phonon $S(Q, E)$ from the finite temperature effective force constants. The Fermi surface was calculated with a $10 \times 10 \times 10$ k-point grid and visualized using Xcrysden [22]. Exchange constants were calculated as described in Supplementary Information. The SpinW program [23] was used to simulate the spin waves and corresponding $S(Q, E)$ using both the NN model and NNN model. The energy resolution function of ARCS was convoluted with the models for both phonon and spin wave simulations. In addition, a momentum resolution of 0.1 r.l.u. was convoluted in the phonon simulation, and an extra 6 meV energy resolution was convoluted in the spin wave simulation to compensate for momentum resolution and the spin wave damping effects.
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Supplementary Information:

**Frustration-induced spin wave anomaly and dimension instability in FeGe$_2$**

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Crystal structure and Brillouin zone map of FeGe$_2$

FeGe$_2$ has the same body-centered tetragonal crystal structure (FIG. S1) as $\theta$-phase Al$_2$Cu with the space group $I4/mcm$. It exhibits two zero-field transition temperatures, one is a second order Néel transition from paramagnetic phase to an incommensurate spin-density-wave state at 289 K, the other is a first-order transition from an incommensurate state to commensurate antiferromagnetic state at 263 K. (HK0) slices were defined such that at the M points $Q = [100]$ and at the X points $Q = \left[ \frac{1}{2} \frac{1}{2} 0 \right]$

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FIG. S1. (a) Crystal structure of FeGe$_2$ with Neel-type AFM magnetic configuration. (b) The Brillouin Zone map of FeGe$_2$. 
Anomalous magnetic scattering in the HK0 planes

The anomalous intensity is observed at 20 K in finite energy transfer up to 25 meV. At 300 K, a checkerboard-shaped pattern was formed from 0 to 20 meV. The anomaly weakens at 500 K and finally vanishes at 635 K as magnetic order collapses.

FIG. S2. S(Q, E) slices in the (HK0) plane from ARCS measurements. Slices were obtained by integrating from -0.5 to 0.5 meV in energy and from -0.1 to 0.1 reciprocal lattice units (r.l.u.) in [0, 0, L].
Across-rod spectra

Spectra intensity perpendicular to the rods (Γ-X) is weak and the dispersion is very steep around X points, revealing great anisotropy of this excitation.

FIG. S3. Across-rod spectra in several Brillouin zones in slices along [-H, -H, 0]. The anomalous intensity is very steep and only exist near X points (half-integer H values). Slices were obtained by integrating from -0.1 to 0.1 r.l.u. in [0, 0, L] and from -0.05 to 0.05 r.l.u. in [H, H, 0].
**Fermi surface**

The Fermi surface is calculated with a non-magnetic primitive cell. There are three bands across Fermi surface, shown in different colors (FIG. S4). In order to produce the checkerboard-shaped pattern in elastic scattering, certain scattering channels for electrons should exist, which require specific shape of the Fermi surface. However, the condition is not meet for all three bands after exploring the data.

![FIG. S4. Fermi surface of non-magnetic FeGe2. (a)(b)(c) Top views. (d)(e)(f) Side views.](image)
Exchange parameters calculation

Exchange constants were calculated using atomistic simulation based on density functional theory. Total energy calculations were made on five collinear magnetic configurations and the exchange parameters were fit to these energies under the assumption that the change of energy is only dependent on the exchange interactions. The plane-wave cutoff was 850 eV. A supercell of a $2 \times 2 \times 2$ conventional unit cell with 96 atoms was used with a $12 \times 12 \times 12$ k-point grid. Results are shown in main text.

It should be noted that a stripe-type configuration has the next-lowest ground state energy. At 300 K, the energy difference between stripe-type and Néel-type configurations is about 3.7 meV per atom and the $k_B T$ becomes 25.8 meV, so the ratio between these two configurations $N_3/N_1 = e^{-\Delta E/k_B T} \approx 0.86$.

\[ E = E_0 - N_1 J_1 s^2 - N_2 J_2 s^2 - N_4 J_4 s^2 \]

\[ E = E_0 - N_1 J_1 s^2 - N_2 J_2 s^2 \]

| Configuration | Energy / eV |
|---------------|------------|
| 1             | -560.13035462 |
| 2             | -559.21035056 |
| 3             | -560.01067121 |
| 4             | -559.79226090 |
| 5             | -559.79225802 |

Table S1. Total energy of five collinear magnetic configurations.

Correlation length fittings

To gain a quantitative understanding of the spatial coherence behind these rods, correlation lengths were extracted by fittings elastic cuts with a Voigt function across the rod of scattering in the basal plane ($\Gamma$-X) and across-plane ($\Gamma$-Z)(Table S2). Along the rod direction, the almost flat intensity suggests no correlation.

The Voigt function is of the form:
Where $A$ is amplitude, $\sigma$ is the standard deviation determined by fitting a nearby nuclear Bragg peak to a Gaussian function, $\gamma = 1/\xi$ is the inverse of the real-space correlation length and $q$ is the wave vector away from the peak center.

\[
I(q) \propto A \int_{-\infty}^{\infty} e^{-\frac{r^2}{2\sigma^2}} \frac{\gamma}{(q - \tau)^2 + \gamma^2} d\tau
\]

Table S2. Correlation lengths extracted along across-rod and across-plane at 300 K and 500 K.

| Temperature / K | Across-rod $\xi$ / Å | Across-plane $\xi$ / Å |
|-----------------|-----------------------|------------------------|
| 300 K           | 12                    | 23                     |
| 500 K           | 4                     | 11                     |