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Recommended Citation

Y. Su et al. (2022). "Frustration-Induced Diffusive Scattering Anomaly And Dimension Change In FeGe₂". *Physical Review B*. Volume 106, Issue 2. DOI: 10.1103/PhysRevB.106.024406
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Frustration-induced diffusive scattering anomaly and dimension change in FeGe₂Yaokun Su¹, Hillary L. Smith², Matthew B. Stone³, Douglas L. Abernathy³, Mark D. Lumsden³, Carl P. Adams⁴, and Chen Li^{1,5,*}¹*Materials Science and Engineering, University of California, Riverside, Riverside, California 92521, USA*²*Physics and Astronomy, Swarthmore College, Swarthmore, Pennsylvania 19081, USA*³*Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA*⁴*Department of Physics, St. Francis Xavier University, Antigonish, Nova Scotia B2G 2W5, Canada*⁵*Mechanical Engineering, University of California, Riverside, Riverside, California 92521, USA* (Received 9 February 2022; revised 10 April 2022; accepted 10 June 2022; published 11 July 2022)

Magnetic frustration, arising from the competition of exchange interactions, has received great attention because of its relevance to exotic quantum phenomena in materials. In the current work, we report an unusual checkerboard-shaped scattering anomaly in FeGe₂, far from the known incommensurate magnetic satellite peaks, by inelastic neutron scattering. More surprisingly, such phenomenon appears as spin dynamics at low temperature, but it becomes prominent above Néel transition as elastic scattering. A model Hamiltonian that includes an intraplane next-nearest neighbor was proposed and such anomaly is attributed to the near-perfect magnetic frustration and the emergence of unexpected two-dimensional magnetic order in the quasi-one-dimensional FeGe₂.

DOI: [10.1103/PhysRevB.106.024406](https://doi.org/10.1103/PhysRevB.106.024406)

Magnetic frustration has attracted interest due to its relation to novel phases including quantum spin liquids, spin and electronic nematic phases, and unconventional superconductivity [1–5]. Generally, the magnetic frustration arises with a special geometry of lattice, but it can also be achieved as a consequence of the competition between different pair antiferromagnetic (AFM) interactions.

In recent years, AFM materials have been widely studied due to their robustness against disturbance and potential applications in high density data storage [6], resistive switching [7,8] and spintronics [9,10]. As a germanium-based AFM intermetallic, FeGe₂ has been explored by numerous experimental and theoretical studies for its complex magnetism [11–17]. Magnetic excitations have been measured in FeGe₂ [18,19], revealing a large anisotropy and an overdamped feature of the spin wave. A nearest-neighbor (NN) Heisenberg model was proposed with $SJ_c = 136$ meV and $SJ_1 = -8.8$ meV, where S is the on-site spin magnitude and J_s are the exchange constants. However, the measurements were highly restricted by the instruments used, and a detailed examination of its magnetic excitations throughout the full Brillouin zone is still missing.

FeGe₂ has the same body-centered-tetragonal crystal structure [20] (Fig. S1) as θ -phase Al₂Cu (space group $I4/mcm$). It exhibits two zero-field magnetic phase transitions on heating [17,21]: one first-order transition from a commensurate AFM state to an incommensurate spin-density-wave state at 263 K, and another second-order Néel transition from the incommensurate state to paramagnetic phase at 289 K. The ordering wave vector changes from $(2\pi/a)[1, 0, 0]$ for the

commensurate state to $(2\pi/a)[1 + \delta, 0, 0]$ for the incommensurate state, where δ varies from 0 to 0.05. Along the c axis, the NN distance between Fe atoms is 2.478 Å, which is close to that of elemental Fe (2.482 Å), such that ferromagnetic (FM) exchange interaction J_c is expected to be strong. In the a - b plane, only a weak NN AFM exchange interaction J_1 has been considered previously [18,19]. While the magnetic moments were known to lay in the basal plane with a value of $1.2 \mu_B$ per Fe atom, their exact orientation is still unclear [21].

Here we provide a combined scattering and computational study of FeGe₂ 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 FeGe₂, revealing an unusual checkerboard-shaped scattering anomaly. Such an anomaly appears in dynamic part at low temperature and turns into diffusive intensity above Néel transition. An additional intraplane next-nearest neighbor (NNN) interaction J_2 is found to be necessary to fully describe the spin wave spectrum. Exchange parameters calculated from atomistic simulations and the model including J_2 suggest the near-perfect magnetic frustration in FeGe₂, enabling us to reproduce the anomalous excitation in the low temperature AFM state. Instead of the one-dimensional (1D) correlation, unexpected two-dimensional (2D) correlations emerge as temperature increase, attributed to the appearance of stripe-type domains and the magnetic frustration. Such 2D correlations are believed to rationalize the anomalous checkerboard-shaped diffusive scattering.

A semicylindrical single crystal FeGe₂ with an approximate 15-mm radius and 40-mm length with a mass of 110 g was used for the INS measurements. This crystal, used in previous studies of FeGe₂ [19,22], was measured using

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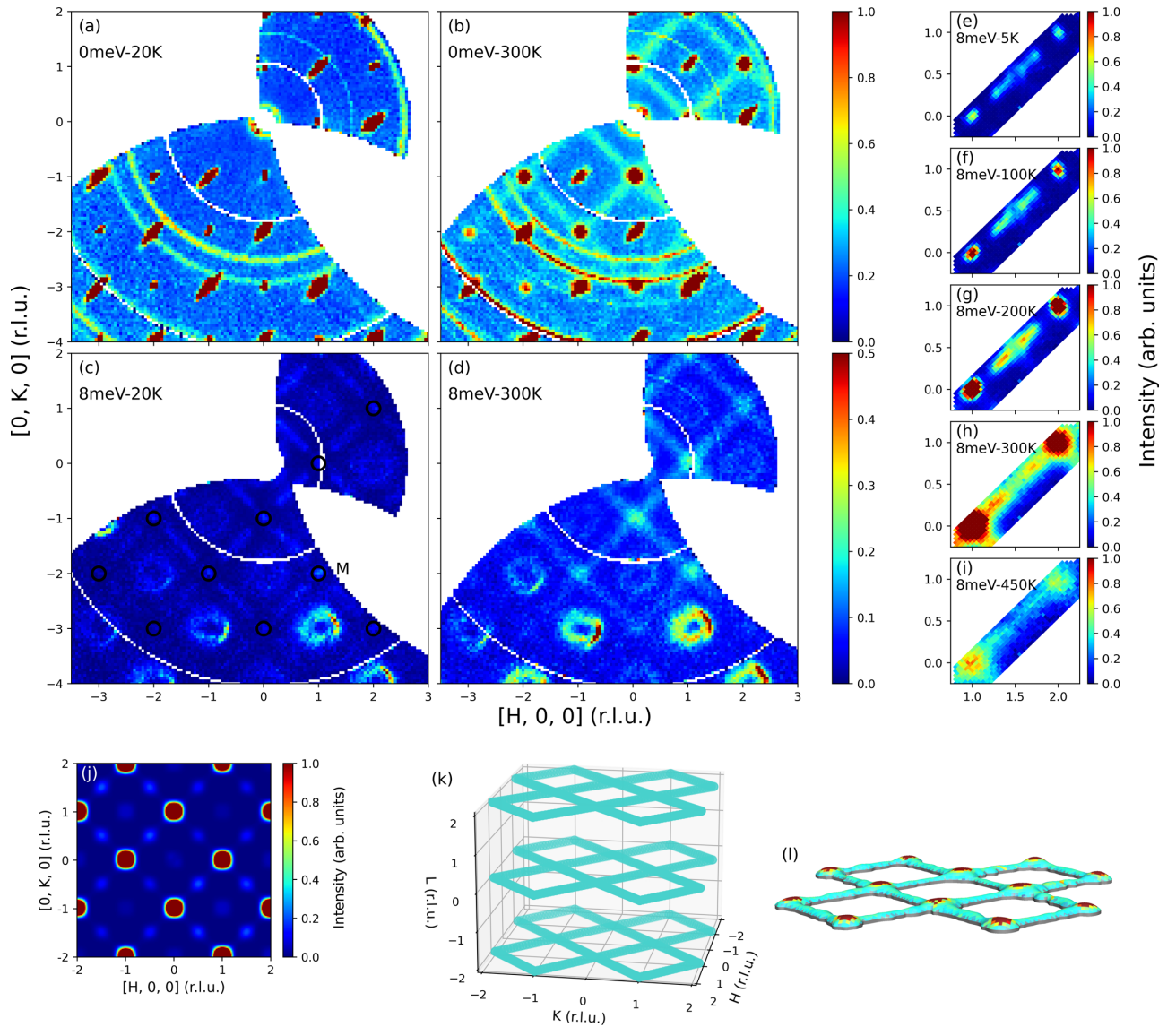


FIG. 1. Checkerboard-shaped anomaly is observed in the $(HK0)$ planes. (a)–(d): $S(\mathbf{Q}, E)$ slices from ARCS measurements, obtained by integrating $[-0.5, 0.5]$ meV in energy and $[-0.1, 0.1]$ reciprocal lattice units (r.l.u.) along $[0, 0, L]$. (a), (b): Elastic scattering results, (c), (d): inelastic scattering results with a neutron energy loss of 8 meV. In (a), (b), those peaks with $H+K$ even are nuclear Bragg peaks (Γ), and those with $H+K$ odd are magnetic Bragg peaks (M). In general, the intensities of nuclear Bragg peaks are stronger than that of magnetic ones and will increase with respect to the absolute value of momentum transfer $|\mathbf{Q}|$. The intensities of magnetic Bragg peaks are, on the other hand, weaker at larger $|\mathbf{Q}|$. (e)–(i): $S(\mathbf{Q}, E)$ slices from HB-3 measurements, which is more limited but has better resolution. (j): Spin wave simulation of $(HK0)$ plane at 8 meV from NNN model. (k): Three-dimensional (3D) schematic of the checkerboard-shaped anomaly. (l): 3D rendering of $L=0$ schematic.

the time-of-flight Wide Angular Range Chopper Spectrometer (ARCS) and Triple-Axis Spectrometer HB-3 at Oak Ridge National Laboratory (details in the Supplemental Material [23]).

Figure 1 shows a sample of the INS data acquired as a function of both temperature and energy transfer. Figs. 1(a) and 1(b) shows the elastic scattering in the $(HK0)$ planes. At 20 K, one sees the diffraction pattern with several aluminum powder line rings visible (from sample environment background). As the temperature increases, an anomalous feature can be observed clearly in the $(HK0)$ slices of dynamical structure factor $S(\mathbf{Q}, E)$ (Fig. 1 and Fig. S2). Besides the expected nuclear and magnetic Bragg peaks, there is extra

intensity connecting NN magnetic Bragg peaks. This intensity forms a checkerboard arrangement with rods along the X - M directions, 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)]. HB-3 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 [Figs. 1(e)–1(i)].

The extent of the phonon and magnetic excitation spectra can be assessed by examining the scattering intensity

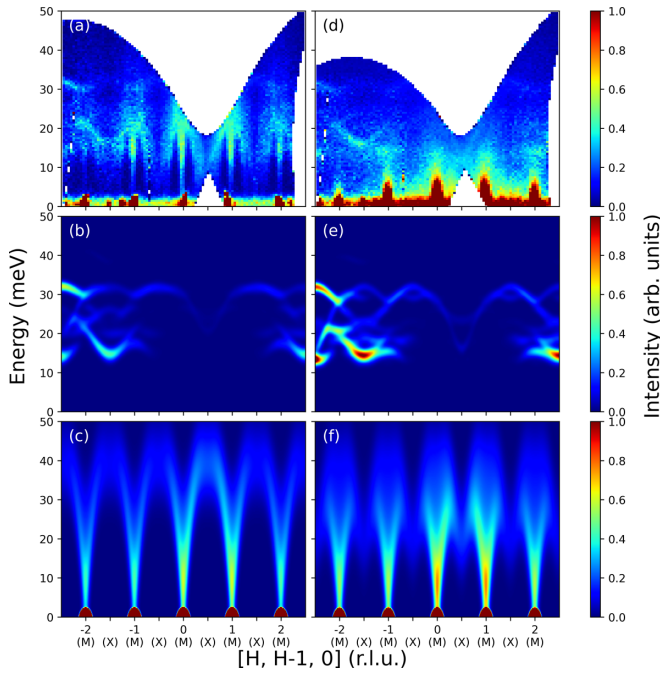


FIG. 2. Dispersive scattering intensity in several Brillouin zones in slices along $[H, H-1, 0]$. (a), (b) 20 K. (d), (e) 300 K. (a) and (d) are $S(\mathbf{Q}, E)$ slices from ARCS measurements, obtained by integrating $[-0.1, 0.1]$ r.l.u. along $[0, 0, L]$ and $[-0.05, 0.05]$ r.l.u. along $[H, -H, 0]$. (b)(e) are simulated phonons. (c)(f) are simulated spin waves from the NN model and NNN model.

as a function of energy and momentum transfer along the X - M direction for 20 and 300 K as shown in Figs. 2(a) and 2(d), respectively. Magnetic excitations emerge from the M points and disperse up to approximately 30 meV. As momentum transfer increases, the magnetic form factor causes the spin wave scattering intensity to decrease. At larger momentum transfers, one can see the optical phonon excitations between approximately 15 and 35 meV.

Phonon simulations were performed using finite temperature effective force constants (details in the Supplemental Material [23]). The simulated phonons match well with experimental data, as shown in Figs. 2(b), 2(e) for the X - M direction. The magnetic excitations dispersing out of M points were reproduced as described in the Supplemental Material [23] using the NN model, with SJ values from literature [19]. The NN model reproduces some other features of the magnetic spectrum. Along the Γ - M direction, the simulated spin wave agrees perfectly with experimental data. Along the X - M direction, the agreement is still good in the vicinity of the magnetic zone center, although the measured spin wave appears steeper [Fig. 2(c)].

There are important differences between the measurement and simulation. The main difference is that the continualike intensity between neighboring M points is not reproduced in either phonon or spin wave simulation. This intensity is associated with the anomalous checkerboard intensity observed in the $(HK0)$ planes. Although overdamped, 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

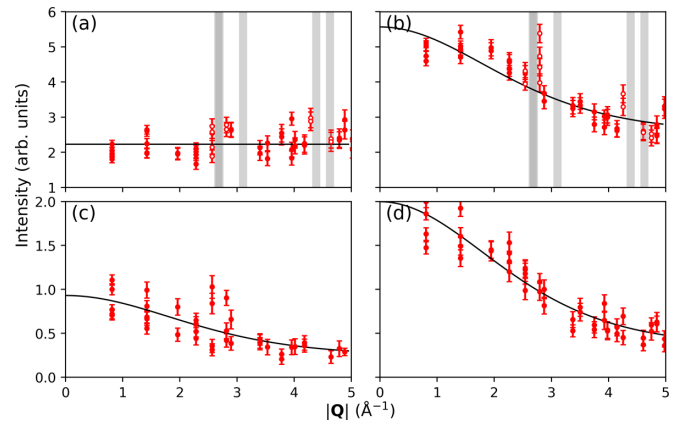


FIG. 3. The $|\mathbf{Q}|$ dependence of the checkerboard-shaped anomaly matches well with magnetic form factor of Fe. The fittings were done on 20 K (a), (c) and 300 K (b), (d) ARCS data for energy transfer of 0 meV (a), (b) and 8 meV (c), (d). The red solid circles are experimental data used for fitting, and the black solid lines are the fitted results. The gray vertical stripes in (a), (b) are places where data points were excluded (red open circles) due to the sample environment background.

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 (Fig. S3).

To reveal the origin of the checkerboard anomaly, a \mathbf{Q} -dependence analysis was performed. It is found this anomalous intensity only appear where L is even [Fig. 1(k)], following the same behavior of magnetic Bragg peaks. This observation indicates that the order behind the anomaly has the same periodicity along the c axis as the magnetic structure of FeGe_2 . To quantify the structural factor of the anomalous INS intensity, equivalent points of $[1.3, 0.3, 0]$ in momentum space were selected on each rod at 0 meV and 8 meV ($HK0$) slices and their intensity was compared to $Af^2(\mathbf{Q}) + B$, where $F(\mathbf{Q})$ is the magnetic form factor of charge-neutral Fe atom, A and B are constants to be fit. Fe atom was used here instead of ions since no charge transfer was found in our charge distribution calculation [24]. Figure 3 shows the excellent agreement between the fitted curves and the experimental data, further confirming that the anomalous intensity, in both the elastic and inelastic scattering, is indeed from the magnetic origin. 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 excluded, as described in the Supplemental Material [23].

To describe our observation, a new model is needed. A checkerboard-shaped anomalous excitation without periodicity along $[0, 0, L]$ has previously been observed in quasi-2D square lattice systems with magnetic frustration, and is described by the J_1 - J_2 Heisenberg model [25,26]. Different from FeGe_2 , the interlayer interaction J_c is ignored in these systems since it is generally much weaker. Besides the NN interaction J_1 , the intraplane NNN interaction J_2 is also

TABLE I. Parameters obtained from total energy calculations.

Model	$J_1 S^2$ (meV)	$J_2 S^2$ (meV)	$J_c S^2$ (meV)	η
NN	-7.2 ± 2.2	—	83 ± 4	—
NNN	-7.2 ± 0.5	-2.6 ± 0.4	81.1 ± 0.9	1.4

important in describing the excitations in these systems. The ground state of these systems could be determined by the frustration parameter $\eta = J_1/2J_2$, where J_1 could be either FM or AFM but J_2 is always AFM. Néel-type, stripe-type and FM ordering occurs for $\eta > 1$, $|\eta| < 1$ and $\eta < -1$, respectively. Perfect frustration happens when $|\eta| = 1$. Extreme spatial anisotropy due to the perfect frustration leads to effectively 1D behavior and corresponding planelike features in INS [27].

Identical to the magnetic atom in these systems, each Fe in FeGe₂ has four intraplane NN atoms and four intraplane NNN atoms. However, FeGe₂ has an additional strong interplane interaction J_c . To determine the extent of the role of J_2 interaction in FeGe₂, 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 (Fig. S5 and Table SI). Two models were used in the least-squares fitting: the NN model containing only J_1 and J_c , and the NNN model that also includes J_2 . The results are shown in Table I.

For the NN model, the calculations yield $|J_c/J_1| \sim 11.6$, this large ratio represents the anisotropic nature of in-plane and Γ -Z spin waves in FeGe₂, consistent with a previous report [18]. For the NNN model, $J_1 S^2$ and $J_c S^2$ remain almost unchanged from those values determined for the NN model and $J_2 S^2$ has a value of -2.6 meV. J_1 and J_2 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 then used to perform spin wave simulations. Assuming no charge transfer between Fe and Ge atoms and a low-spin configuration in a tetrahedral environment with spin $S = 1$ and a g factor of 2, the magnetic moment is expected to be $\mu = g\sqrt{S(S+1)}\mu_B = 2.8 \mu_B$ per Fe atom. However, this is about twice the experimental value, explained as arising from hybridization of the $3d$ and $4s$ orbitals of the Fe atoms [14]. As a result, a smaller effective value of S is needed to account for the itinerant nature of FeGe₂. A value of 0.5 was chosen here so that spin wave along Γ -M matches with result from the literature [18]. The simulation in Fig. 2(f) shows the importance of J_2 in reducing the spin wave energy near the X points in the Brillouin zone, and significantly improving agreement with the experimental results. A large energy broadening of the spin wave is needed in reproducing the measured spectra, and an additional 10 meV energy broadening is included in the simulation. Such damping of the spin wave may come from the frustration in FeGe₂. The simulated ($HK0$) slice at 8 meV [Fig. 1(j)] shows strong “rod” intensity along X -M and only

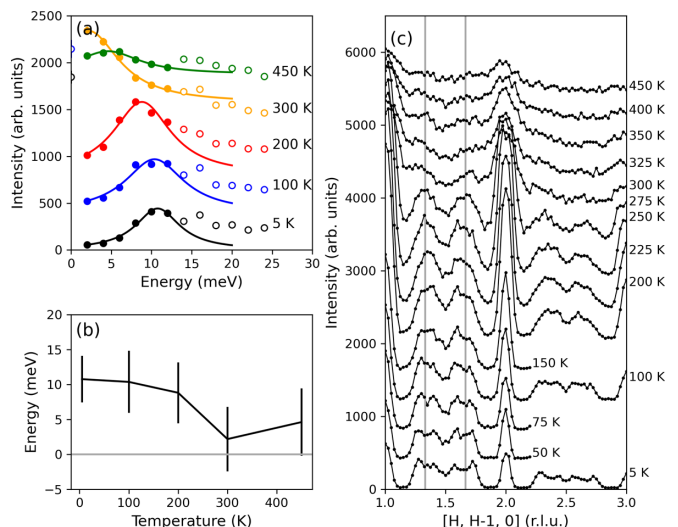


FIG. 4. Temperature dependence of the anomalous intensity along $[H, H-1, 0]$. (a) Constant \mathbf{Q} cuts at $[1.6, 0.6, 0]$ from HB-3 data. (b) Lorentzian fit to the first peak (solid circles) in (a), error bars represent the full width at half maximum. (c) Constant E scans along $[H, H-1, 0]$ at 8 meV with the black solid lines as guides to the eye. The vertical gray lines indicate peaks at about 1 and 2 r.l.u. Error bars are smaller than the symbols.

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 \mathbf{Q} cuts were obtained from the HB-3 data (Fig. S6), revealing a dramatic softening of the spin wave from finite energy at low temperatures to near 0 meV at 300 K as seen in Figs. 4(a) and 4(b). This effect is also visible in Fig. 4(c) showing the constant energy scans at 8 meV with denser temperature points. 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 wave collapses near 300 K as one enters the paramagnetic phase of FeGe₂.

At the same time as the spin wave collapses, rods of scattering between neighboring M points connect with each other and form the checkerboard-shaped diffuse scattering pattern in ($HK0$) elastic slices of $S(\mathbf{Q}, E)$. 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 the Supplemental Material [23]. 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, comparable with each other. In this case, 3D order breaks down with 2D correlations of plates along directions bisecting the a and b axes remaining in the paramagnetic phase. The 2D correlations unveiled in real space are surprising in FeGe₂. In such a quasi-1D system, spins are weakly coupled in the a - b plane with fourfold symmetry, and it is expected that correlations in

this plane disappear simultaneously with only 1D correlation along c axis left. At 500 K, the correlation lengths become 4 and 11 Å, respectively, indicating a simultaneous decrease of short-range order upon further warming into the paramagnetic phase.

This striking dimension change can also be explained by J_2 and magnetic frustration. 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 magnitude of 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-plate correlations, but the neighboring plates are nearly decoupled because of the small effective correlations between plates. Since $[1,1,0]$ and $[1, -1, 0]$ are equivalent directions in the system, stripe-type domains along the other direction are equally likely to be formed, together they can account for the rods in the checkerboard arrangement.

In conclusion, we observe anomalous excitations at low temperature, as well as a checkerboard-shaped diffusive scat-

tering pattern developing at high temperature. We showed that these two phenomena, though different in underlying mechanism, both have a magnetic origin and are related to the intraplane NNN interaction J_2 . This previously ignored interaction generates the extra spin wave feature for a large range of the reciprocal space. Our *ab initio* calculations show J_2 leads directly to the near-perfect in-plane magnetic frustration, which facilitates the emergence of unexpected 2D short-range magnetic order at high temperature. Low dimensional FeGe₂ has been synthesized for potential spintronic applications [28,29]. Our revelation of the magnetic frustration and its roles may provide some insights on these studies. Our work also sheds light on the potential of controlling the magnetic dimensionality and corresponding properties of materials by frustration.

This material is based upon work supported by the National Science Foundation under Grant No. 1750786. The neutron scattering portion of this research used resources at the Spallation Neutron Source and the High Flux Isotope Reactor, both of which are Department of Energy (DOE) Office of Science User Facilities operated by the Oak Ridge National Laboratory.

D.L.A. and C.L. performed the INS measurements on ARCS. M.B.S., M.D.L., and C.P.A. performed the INS measurements on HB-3. Y.S. performed the theoretical calculation. Y.S. and C.L. analyzed the data and wrote the manuscript. All authors contributed to discussing and editing the manuscript.

The authors declare no competing interests.

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