Density wave orders in two-dimensional and layered materials are known to exhibit rich interplay with their underlying lattice motifs. For example, the honeycomb lattice exhibits the Kekulé bond-density-wave order. In graphene, such Kekulé order can give rise to massive Dirac fermions via chiral symmetry breaking, a mechanism analogous to dynamical mass generation of fundamental fermions in the Standard Model. Similarly, the kagome lattice supports a rich tapestry of charge density wave (CDW) orders, closely related to the Haldane model for Chern insulators and the Varma model for the cuprate superconductors. The exploration of new quantum materials exhibiting density wave orders under broken time-reversal symmetry, especially in the honeycomb and kagome lattices, promises the discovery of exotic many-body quantum phases, including exotic orbital current orders, unconventional topological superconductors and phases supporting fractional excitations.

The antiferromagnet FeGe assumes a crystal structure comprised of alternating honeycomb and kagome atomic layers. Recent neutron scattering, spectroscopy and transport measurements suggest a CDW in FeGe, providing the first example of a CDW in a kagome magnet. However, the nature of the structural distortion and the mechanism driving the CDW remain elusive. Here, using in-depth first-principles calculations with optimized atomic structure, we predict that FeGe hosts an unusual hybrid kagome-honeycomb CDW. We find a $2 \times 2 \times 1$ CDW, which takes the form of a generalized Kekulé distortion of the Ge honeycomb layers. While, the Fe kagome layers almost remain unchanged. Our analysis indicates that the honeycomb generalized Kekulé distortion is driven by the Fermi surface nesting of kagome electronic states. Our predicted honeycomb-kagome CDW is further substantiated by the matching with the atomic distribution of the electronic density of states data of scanning tunneling microscope (STM).

The pristine phase of FeGe is in the space group $P\bar{6}/mm\bar{m}$ (No.191) [Fig. 1(a)], where Fe atoms form kagome layers. There are two types of Ge atoms, one is located at the hexagonal center of the Fe kagome layer and the other forms honeycomb layers located in-between two kagome layers. By using the high-performance structure research software CALYPSO, we find four types of FeGe supercell structures (SM II). Among these, the $2 \times 2 \times 1$ and $2 \times 2 \times 2$ supercell structures are the lowest energy structures that compete with the pristine phase. No distortion is seen on the Fe kagome layer. The Ge atoms, which form the honeycomb lattice in different layers, move in opposite directions. As a result of the distortion, there are two inequivalent Fe atoms and six inequivalent Ge atoms in the $2 \times 2 \times 1$ cells.

To understand the phase transition between the phases, we plot the computed phonon band structure of the pristine phase and the $2 \times 2 \times 1$ CDW phase in Figs. 1(c) and (d), respectively. Both the pristine phase and the $2 \times 2 \times 1$ CDW phase of FeGe are seen to be dynamically stable. This is to be contrasted sharply from most of the previously reported materials with CDW.
FIG. 1. (a) A 2×2 supercell of the pristine phase, where the primitive cell is shown with black solid line. (b) The 2×2×1 CDW phase, where the blue and purple arrows show the in-plane movement of Ge atoms on the top and middle Ge layers, respectively. The out-of-plane moment of Ge atoms is given in SM II. (c), (d) Phonon spectra of pristine and 2×2×1 CDW phases. (e) Gibbs free energy of 2×2×1 CDW phases relative to the pristine phase. (f) Reaction path between the pristine and 2×2×1 CDW phase calculated by NEB[21–23]. Brown spheres are Fe atoms and the green arrows represent the direction of magnetic moment.

phase transition, where the phonon spectra of the pristine phase exhibits a softening mode at low temperature[24–27]. To further understand the phase transition of FeGe, we show the Gibbs free energy of 2×2×1 CDW phase relative to the pristine phase, which will increase with the increase in temperature [Fig. 1(e)]. At low temperature, the energy difference is negative, and the change of sign occurs at 80 K, where the phase transition will take place. This is close to the results of the latest experiments[16,17], where the CDW transition happens at the temperature of 100-110 K. We further plotted the energy barrier of the reaction path between pristine and 2×2×1 CDW phases in Fig. 1(f), where both phases are local minimum in energy and the CDW phase shows slightly lower energy. There is an energy barrier of ~147 meV/cell along the reaction path. This is different from the kagome material family AV₃Sb₅ (A = K, Rb, Cs)[9], where the pristine phase sits at the saddle point of the potential energy surface, so that movement of the atoms along the softening mode will trigger a spontaneous change to a low-energy phase.

Our calculations also show that both pristine and 2×2×1 CDW phases are collinear antiferromagnetic with magnetic moment perpendicular to kagome layers. In the latest experiments, the enhancement of magnetic moment was observed, which may be explained by chiral flux phase of circulating currents and electrons correlations independent from Fermi surface nesting[16]. Interestingly, our first-principles calculations also capture this enhancement of the magnetic moment. Compared to the pristine phase, the magnetic moment of each Fe atom in the 2×2×1 CDW phase is increased by 0.04 µB [Fig. 1(f), inset].

To further prove that the distorted 2×2×1 CDW phase correctly captures the experimentally observed CDW ground state, we show our STM measurements of the density of states and compare these with our theoretical predictions. Figure 2(a) simulates the charge distribution on kagome layer of pristine phase, where the black parallelogram represents the primitive cell. (b) Tunneling current maps measured by STM, where the blue lines represent the kagome lattice. (c) The simulated surface charge distribution on kagome layer of 2×2×1 CDW phase, where the black parallelogram represents the primitive cell. The upper, middle and lower panels in (a)-(c) are the experimental STM images or simulated surface charge distribution at 40, 20 and -40 meV relative to the Fermi level, respectively.
the charge is mainly distributed on the Fe-triangles. The surface charge distribution based on the $2 \times 2 \times 1$ CDW phase [Fig. 2(c)] presented in this work captures the same behavior as that observed in STM. Bearing all these results in mind, we believe that our predicted $2 \times 2 \times 1$ CDW phase correctly captures the distorted $2 \times 2$ charge ordering structure observed experimentally in FeGe.

Now we turn to discuss the origin of the CDW interaction that induces atomic distortions. We first checked the Fermi surface of the pristine phase [Figs. 3(a)-(c)]. Interestingly we observed a very similar hexagonal Fermi surface both at $k_z=0$ and $k_z=\pi$ planes [colored blue in Fig. 3(a)]. This indicates colored states have very small dispersion along the $z$-direction and a hexagonal prism shape presents in the Fermi surface (Fig. S5 in SM III). The Fermi surface in the $yz$ plane also demonstrates quasi-2D behavior [Figs. 3(b)-(c)]. The momentum separation of the parallel sides of the hexagonal prism-shaped Fermi surface is about half of the unit vector of the pristine phase, as indicated by the yellow arrows in Figs. 3(a)-(c). These results indicate that the Fermi-surface nesting is likely to be the origin of the CDW interaction. To further demonstrate it, we calculate the zero frequency susceptibility $\chi^0(q, \omega = 0)$, i.e., Lindhard response function $^{29–32}$ to determine the nesting vector. The two dimensional renormalized Lindhard function is plotted in Fig. 3(d). Indeed the maxima are located at the center of the boundary of the first Brillouin zone (BZ) with the nesting vectors $Q_1 = (0.5, 0)$ and $Q_2 = (0, 0.5)$. This is in accord with the experimental observation $^{16,17}$.

To gain further insight into the Fermi surface nesting scenario, we calculated the band structure of the pristine phase. Figure 4(a) depicts the electronic band structure of the pristine phase with SOC, and the Fe-$d$ orbital-projected band structures to focus on the singularity of the kagome lattice are shown in Figs. 4(b)-(c). The “flat” bands lie slightly above the Fermi level with large band-widths, which is consistent with the previous theoretical results $^{28}$. Our calculations identify five Dirac cones (DCs) derived from the kagome lattice, which display van Hove singularities at M/L points and Dirac points at K/H points. The schematic diagram of these Dirac cones is plotted in Fig. 4(d). Notably, DC2 and DC3 have similar dispersion, but van Hove’s singularities are at different energy levels. This is because $d_{xy}/d_{2-y^2}$ orbitals, which dominate DC2 and DC3, lie in the kagome plane, and the hybridization with their upper and lower Ge atoms is weak. In contrast, the other three Dirac cones are mainly contributed by the out-of-plane $d_{xz}/d_{yz}$ orbitals, and due to their strong hybridization with Ge orbitals, they show significantly different dispersion in $k_z=0$ and $k_z=\pi$ planes. The DC1 and DC3 Dirac cones shown by solid blue lines in Fig. 4(d) play major roles in the hexagonal Fermi surface at $k_z=0$ plane, while the main contribution of the hexagonal Fermi surface gradually becomes DC3 as the 2D Fermi surface lifts from $k_z=0$ to $\pi$.

Now we discuss the effects of CDW interaction on the band structure. The E-K dispersion of $2 \times 2 \times 1$ CDW phase with the inclusion of SOC is given in Fig. 4(e). Most kagome features: the Dirac points and the flat bands can still be observed in the $2 \times 2 \times 1$ CDW phase. The main effects of the CDW interactions are on the states that are nested by the CDW Q-vectors. Taking the constant energy contour at $k_z=\pi$ at 10 meV below Fermi level as an example, the blue hexagon Fermi surface in pristine phase changes into a circle-like Fermi surface in $2 \times 2 \times 1$ CDW phase [Fig. 4(f) and Fig. S6 in SM III]. The deformation of the Fermi surface weakens the Fermi surface nesting that induces the CDW instability. Energy dispersion along with L’-A’-L’ in cut 2 plane of the pristine phase is shown in Fig. 4(g), and the unfolded band structure of the $2 \times 2 \times 1$ CDW phase along the same path in Fig. 4(h). A clear gap-opening due to CDW interactions can be seen along this path.

Notably, the Fermi surface nesting comes from the
kagome-derived Dirac cones, but structural distortions originate from motions of Ge atoms, forming generalized Kekulé distortions\textsuperscript{33–35}, where the shortest bonds form an “O”/“Y” shaped pattern as shown in Fig. 4(i). The distortion of Ge atoms modifies the charge hopping on the kagome layer, which affects on the Fe-kagome bands and weakens the Fermi surface nesting. Moreover, the Ge distortion results in greater spin splitting of each Fe atom. When the density of states is integrated into the Fermi level, the net charge of spin-up and spin-down states at each Fe atom induces magnetism. It can be clearly seen in Fig. S7 (SM IV) that the density of states of pristine and 2×2×1 CDW phase are similar, but in 2×2×1 CDW phase, the spin-up electronic states increase and spin-down electronic states decrease for the top Fe-kagome layer, and vice versa for the bottom Fe-kagome layer. In this way, in 2×2×1 CDW phase, the magnetic moment on each Fe atom is increased by 0.04 \( \mu_B \) to 1.48 \( \mu_B \), consistent with the experimental observation that magnetic moment enhances in CDW phase\textsuperscript{16,17}.

In summary, we have investigated the structural and electronic properties of antiferromagnetic kagome metal FeGe. We first discuss in-depth the ground state 2×2×1 CDW phase and our analysis indicates that this phase will transform into the pristine phase at a temperature of \( \sim 80 \) K, which is comparable to the corresponding experimentally measured transition temperature of \( \sim 100-110 \) K\textsuperscript{16,17}. Our predicted 2×2×1 CDW phase accurately captures the atomic topography and spectroscopies we observed in our STM experiments and the enhancement of magnetic moments\textsuperscript{16,17}. We demonstrate that our 2×2×1 CDW phase is driven by Fermi surface nesting, which mainly involves Fe-kagome-derived Dirac cones. In sharp contrast with earlier studies, our 2×2×1 CDW phase exhibits a generalized Kekulé distortion in Ge honeycomb layers, forming an O-shaped bond texture in the top Ge layer and a Y-shaped bond texture in the middle Ge layer. While, the Fe kagome layers almost remain unchanged. Kekulé distortions in graphene open new conduction channels between the valleys and influence optical absorption and conductivity\textsuperscript{35}, suggesting that generalized Kekulé distortions in FeGe might also yield interesting physical effects. Our study provides insight into the CDW phase and indicates that further exploration of the topological nature of the ground state of magnetic kagome-honeycomb lattices and their transport and optical responses will be interesting. We also identify the structure of 2×2×1 CDW phase and two metastable \( 1\times\sqrt{3}\times1 \) and 2×2\( \sqrt{3}\times1 \) structures (SM II), which could provide theoretical support for future experiments.

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1 C.-Y. Hou, C. Chamon, and C. Mudry, Electron fractionalization in two-dimensional graphene like structures. Phys. Rev. Lett. 98, 186809 (2007).
2 C. Gutiérrez et al., Imaging chiral symmetry breaking from Kekulé bond order in graphene. Nat. Phys. 12, 950 (2016).
3 C. Bao et al., Experimental Evidence of Chiral Symmetry Breaking in Kekulé-Ordered Graphene. Phys. Rev. Lett. 126, 206804 (2021).
4 H. Chen et al., Roton pair density wave in a strong-coupling kagome superconductor. Nature 599, 222 (2021).
5 H. Zhao et al., Cascade of correlated electron states in the kagome superconductor CsV_Sb. Nature 599, 216 (2021).
6 L. Nie et al., Charge-density-wave-driven electronic nematicity in a kagome superconductor. Nature 604, 59 (2022).
7 Y.-X. Jiang et al., Unconventional chiral charge order in kagome superconductor KV_Sb. Nat. Mater. 20, 1353 (2021).
8 H. Li et al., Observation of unconventional charge density wave without acoustic phonon anomaly in kagome superconductors AV_Sb(A=Rb, Cs). Phys. Rev. X 11, 031050 (2021).
9 H. Tan, Y. Liu, Z. Wang, and B. Yan, Charge density waves and electronic properties of superconducting kagome metals. Phys. Rev. Lett. 127, 046401 (2021).
10 T. Neupert et al., Charge order and superconductivity in kagome materials. Nat. Phys. 18, 137 (2022).
11 F. H. Yu et al., Unusual competition of superconductivity and charge-density-wave state in a compressed topological kagome metal. Nat. Commun. 12, 3645 (2021).
12 C. M. III et al., Time-reversal symmetry-breaking charge order in a kagome superconductor. Nature 602, 245 (2022).
13 H. J. Yang et al., Intertwining orbital current order and superconductivity in Kagome metal. (2022). arXiv:2203.07365.
14 C. M. III et al., Nodeless kagome superconductivity in LaRu_Si. Phys. Rev. Materials 5, 034803 (2021).
15 Y.-X. Jiang et al., Unconventional chiral charge order in kagome superconductor KV_Sb. Nat. Mater. 20, 1353 (2021).
16 X. Teng et al., Discovery of charge density wave in a correlated kagome lattice antiferromagnet (2022). arXiv:2203.11467.
17 J.-X. Yin et al., Discovery of charge order and corresponding edge state in kagome magnet FeGe (2022). arXiv:2203.01888.