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Cascade of pressure induced competing CDWs in the kagome metal FeGe

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Electronic ordering is prevalent in correlated systems, which commonly exhibit competing interactions. Here, we use x-ray diffraction to demonstrate a cascade of pressure induced order-disorder transformations, with propagation vectors $\mathbf{q}_{\text{CDW}} = (\frac{1}{2} \ 0 \ \frac{1}{2})$, $\mathbf{q}^* = (\frac{1}{3} \ \frac{1}{3} \ \frac{1}{2})$ and $\mathbf{q}^\dagger = (\frac{1}{3} \ \frac{1}{3} \ \frac{1}{3})$, in the kagome metal FeGe. In the pressure interval between $4 < p < 10$ GPa, $\mathbf{q}_{\text{CDW}}$ and $\mathbf{q}^*$ coexist and the spatial extent of the $\sqrt{3} \times \sqrt{3}$ order is nearly long-range at ~ 15 GPa, ~ 30 unit cells. Above ~ 25 GPa, the periodic lattice distortion has a propagation wavevector of $\mathbf{q}^\dagger = (\frac{1}{3} \ \frac{1}{3} \ \frac{1}{3})$ at room temperature. The cascade of phase transitions are captured by the Ising model of frustrated triangular lattices and modeled by Monte Carlo simulations based on the dimerization of trigonal Ge₁. The pressure dependence of the integrated intensities and correlation lengths of $\mathbf{q}_{\text{CDW}}$, $\mathbf{q}^*$ and $\mathbf{q}^\dagger$ demonstrate a competition between the 2×2 and $\sqrt{3} \times \sqrt{3}$ phases prove the tunability of order-disorder phase transitions under pressure and the rich landscape of metastable/fragile phases of FeGe.

Correlated phases are usually characterized by a complex interplay of charge, spin, orbital and lattice degrees of freedom with comparable energy scales that allow for a fine tuning of their ground state by external stimuli [1–3]. Notable examples can be found in the gate-tuned superconductivity in SrTiO₃ [4], Mott insulators [5] or twisted bilayer graphene (TBG) [6]. Another example is the ground state of superconducting cuprates that exhibits an interplay of competing charge, spin and superconducting phases [7–9], highly tunable under pressure [10, 11], strain [12–14] and magnetic fields [15, 16].

Very recently, a cascade of correlated phases have been reported in the geometrically frustrated kagome lattices [17–21], believed to be rooted on their rich electronic band structure, featuring van Hove singularities (vHs), Dirac crossings (DC) and dispersionless flat bands [22–25]. Such electronic details could unveil novel unconventional electrodynamics, like chiral CDWs, superconductivity or possible chiral flux phases [26–28], which can be realized upon tuning the band fillings to the Fermi level. For instance, the non magnetic kagome metal AV₃Sb₅ (A=Cs, K, Rb) is characterized by a multiple- $\mathbf{q}$ CDW connecting the M and L points of the Brillouin zone (BZ) [29, 30], intertwined with a low temperature superconducting state [31, 32]. Such 3D CDW is strongly influenced by the dimensionality [33] and stacking sequence [34], magnetic field [35], dynamic disorder [36], uniaxial

strain [37, 38] and pressure [39, 40]. Intriguingly, the application of hydrostatic pressure results in the emergence of new electronic orders [41, 42] that demonstrate the tunability of the AV₃Sb₅ ground state to external parameters. Similarly, the long range 3D CDW in the double layer ScV₆Sn₆ is characterized by a $(\frac{1}{3} \ \frac{1}{3} \ \frac{1}{3})$ propagation vector [43] that competes with the high temperature short range charge fluctuations with wavevector $(\frac{1}{3} \ \frac{1}{3} \ \frac{1}{2})$ [44, 45], featuring a soft mode that becomes stable under moderate hydrostatic pressure [46].

Single crystals of FeGe [47, 48], crystallize in the same space group as AV₃Sb₅ and ScV₆Sn₆ (*P6/mmm*). However, FeGe orders antiferromagnetically below $T_N \sim 400$ K and develops a conical order at lower temperature [49]. At $T_{\text{CDW}} \sim 105$ K, diffraction experiments unveil a dimerization driven short-range charge density wave [50–57] that connect the M, L, and A high symmetry points of the BZ. The microscopic origin of the CDW is still under debate, as angle resolved photoemission (ARPES) and density functional theory (DFT) show saddle points at the M point close to the Fermi level [51, 58], while scattering techniques report a giant spin-phonon coupling above T_{CDW} [59] and a moderate phonon softening below T_{CDW} [50]. Moreover, DFT calculations indicate that both the real and imaginary parts of the electronic susceptibility peak at the $(\frac{1}{3} \ \frac{1}{3} \ 0)$ wavevector (K point in the 2D BZ) [60–62], in contrast to the propagation vectors observed experimentally. Finally, diffuse scattering (DS) locates the CDW within a canonical order-disorder transformation [47], thus hinting at the tunability of the electronic order [63] to external perturbations.

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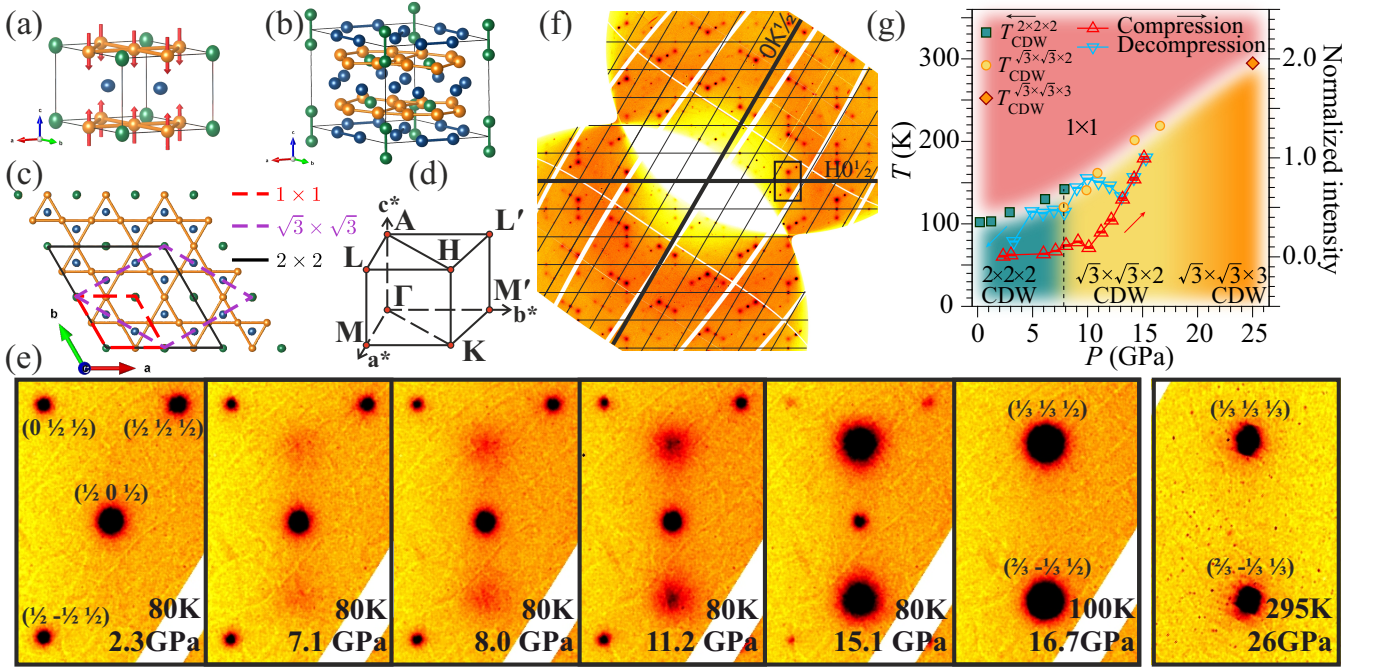


Figure 1. (a) Normal state structure (non-CDW) of FeGe. Arrows denote the spin polarization of the kagome planes, which will double the paramagnetic unit cell in the z -direction. Orange symbols are the Fe atoms, the green represents the Ge atoms in the kagome plane (trigonal Ge_1) and the blue symbols are the Ge atoms in the honeycomb layer (Ge_2). (b) Unit cell of the $\sqrt{3} \times \sqrt{3}$ CDW phase. (c) Top view of the FeGe kagome net, displaying the 1×1 , 2×2 and $\sqrt{3} \times \sqrt{3}$ superlattices. (d) High symmetry points in the non-magnetic Brillouin zone. (e) Detailed $(h\ k\ \frac{1}{2})$ maps (and $(h\ k\ 1/3)$ on the right) parsing the pressure evolution of the different CDW orders of FeGe around the reciprocal lattice vector $\mathbf{G}_{200}$. (f) Reciprocal space map (RSM) sweeping the $(h\ k\ \frac{1}{2})$ plane at 80 K and 8 GPa, highlighting the DS that characterizes the crossover from the 2×2 towards the $\sqrt{3} \times \sqrt{3}$ order. (g) Pressure-temperature (p - T) phase diagram of FeGe (closed symbols). Superimposed (open symbols), compression (red)-decompression (blue) pressure hysteresis (see text).

Given the metastability of the 2×2 phase of FeGe and its short-range correlation length, here we apply hydrostatic pressure to tune a cascade of transformations from the 2×2 towards a $\sqrt{3} \times \sqrt{3} \times 2$, $\mathbf{q}^* = (\frac{1}{3}\ \frac{1}{3}\ \frac{1}{2})$, and $\sqrt{3} \times \sqrt{3} \times 3$, $\mathbf{q}^\dagger = (\frac{1}{3}\ \frac{1}{3}\ \frac{1}{3})$, periodic lattice distortion (PLD). We find that the onset temperature of the 2×2 order increases linearly with pressure up to nearly 150 K. Moreover, the intensity of $\mathbf{q}_{CDW}$ decreases up to $p^* = 15$ GPa, above which, the nearly long-range $\sqrt{3} \times \sqrt{3} \times 2$ order develops. At ~ 25 GPa, the $\sqrt{3} \times \sqrt{3} \times 3$ PLD becomes the ground state at room temperature and demonstrates a strong competition between the three CDW phases. DFT calculations confirm the stability of the $\sqrt{3} \times \sqrt{3}$ phase at high p as a result of the dimerization of the trigonal Ge_1 , that is well captured by the Ising model of frustrated triangular lattices and corroborates the extreme fragility of the electronic orders of FeGe.

Figure 1(a-b) sketches the unit cell of FeGe in its normal (NS), 2×2 and $\sqrt{3} \times \sqrt{3}$ state of FeGe, respectively. The NS unit cell consists on a 2D FeGe kagome layer and hexagonal Ge_2 layers, featuring a honeycomb lattice. At $p=0$, the trigonal Ge_1 in the kagome plane dimerizes along the c -direction, renormalizing the p_z -derived electron pocket at Γ below T_{CDW} [54], hence the

2×2 PLD is a result of the frustrated order driven by the dimerization of Ge_1 , as observed by x-ray diffraction [59, 64] and DFT calculations [55]. At 0 GPa, the CDW in FeGe is described by three independent $\mathbf{q}$ -vectors, namely: $(\frac{1}{2}\ 0\ \frac{1}{2})$ (L point), $(0\ 0\ \frac{1}{2})$ (A point) and $(\frac{1}{2}\ 0\ 0)$ (M point) [50, 59], see fig. 1(d). Focusing on the $\mathbf{G}_{200}$ reciprocal lattice vector, fig. 1(e), the intensity of the CDW peak with propagation vector $(\frac{1}{2}\ 0\ \frac{1}{2})$ shows a gradual weakening up to 15 GPa. Similar pressure dependence of the intensity of the 2×2 order is observed for $(\frac{1}{2}\ 0\ 0)$ (M point) and $(0\ 0\ \frac{1}{2})$ (A point). In the pressure interval between 5 and 10 GPa, broad diffuse satellites emerge at $(\frac{1}{3}\ \frac{1}{3}\ \frac{1}{2})$ and $(\frac{2}{3} - \frac{1}{3}\ \frac{1}{2})$ (H point) and gain spectral weight up to $p^* \sim 15$ GPa. Above p^* , the 2×2 order is reduced and falls below the detection limit. Overall, our experimental results up to 25 GPa are summarized in the p - T -phase diagram presented in the fig. 1(g) (see suppl. inf. Appendix A for the determination of the bulk modulus [65–67]). The onset of the CDW with propagation vector $\mathbf{q}^* = (\frac{1}{3}\ \frac{1}{3}\ \frac{1}{2})$ follows the same pressure dependence as the 2×2 order at a rate of $\Delta T/p \sim 7$ K/GPa, presumably due to the release of the dimerization driven lattice frustration [47], in marked contrast with the AV_3Sb_5 and ScV_6Sn_6 kagome

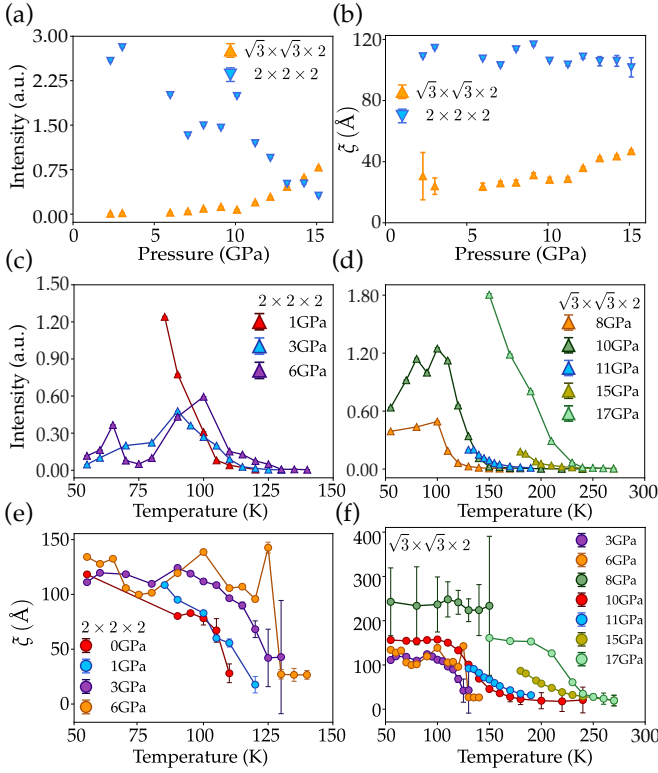


Figure 2. (a) Pressure dependence of the integrated intensities and correlation lengths (b) of the 2×2 and $\sqrt{3} \times \sqrt{3} \times 2$ orders at 80 K. (c) Temperature dependence of the intensity of the 2×2 and (d) $\sqrt{3} \times \sqrt{3} \times 2$ phases at different values of pressure. (e) Temperature dependence of the correlation length of the 2×2 and (f) $\sqrt{3} \times \sqrt{3} \times 2$ orders at selected pressures.

metals [40, 42]. Further increase of pressure leads to the disappearance of the $\sqrt{3} \times \sqrt{3} \times 2$ phase and a tripling of the unit cell along the c -direction ($\sqrt{3} \times \sqrt{3} \times 3$ PLD) at ~ 25 GPa and room temperature, $\mathbf{q}^\dagger = (\frac{1}{3} \frac{1}{3} \frac{1}{3})$.

To further delve into the evolution of the 2×2 and $\sqrt{3} \times \sqrt{3}$ orders as a function of p , we parse the x-ray reciprocal space maps (RSM), see suppl. inf. fig. 3. From the pressure dependence of the 2×2 and $\sqrt{3} \times \sqrt{3} \times 2$ integrated intensities at $T=80$ K (where the integrated intensities reach their maximum value), we infer a mutual competition between both orders, fig. 2(a). On the other hand, the correlation length of the 2×2 order remains practically unaffected by pressure (~ 100 Å), while the $\sqrt{3} \times \sqrt{3} \times 2$ phase, yet less correlated, increases with p , fig. 2(b). Furthermore, the spatial distribution of the $\sqrt{3} \times \sqrt{3} \times 2$ phase extends to ~ 30 unit cells in the ab -plane at low temperature ($T < 50$ K) and 17 GPa, fig. 2(f), hinting at a quasi-long-range order at high p .

Notably, once the pressure is cranked up, the temperature dependence of the integrated intensity at constant p of the 2×2 and $\sqrt{3} \times \sqrt{3} \times 2$ superlattice reflections start to peak right below the CDW onset and further weakens down to 10 K, fig. 2(c-d). Similar behavior has been observed in FeGe_{0.9} crystals [47], presumably associated to

quenched disorder or the internal strain induced by the Ge deficiency, and in correlated systems with competing/coexisting orders [9, 68]. On the other hand, the temperature dependence of the correlation lengths for both orders, fig. 2(e-f) follows a gradual decrease up to T_{CDW} due to either the formation of dislocations or to a shrinking of the ordered domains into smaller regions. Intriguingly, upon pressure cycling (compression-decompression below T_{CDW}), the 2×2 order can only be restored after warming up above T_{CDW} and subsequent cooling, indicating an extreme fragility of 2×2 phase to pressure, thus the $\sqrt{3} \times \sqrt{3}$ phase is the ground state of FeGe at high p and a metastable phase at low p , fig. 1(g).

Given that the $\sqrt{3} \times \sqrt{3} \times 2$ PLD evolves from the 2×2 order, featuring an order-disorder transformation of dimerized Ge₁ [47], we model the $\sqrt{3} \times \sqrt{3} \times 2$ order following the Ising scenario of antiferromagnetic triangular lattices, fig. 3(a) (suppl. inf. Appendix D), where the strength and direction of interacting spins is controlled by the c_i coefficients in the eq. 1.

$$H = \sum_{\langle i,j \rangle:NN} c_1 \sigma_i \sigma_j + \sum_{\langle i,j \rangle:NNN} c_2 \sigma_i \sigma_j + \sum_{\langle i,j \rangle:4NN} c_3 \sigma_i \sigma_j + \sum_{\langle i,j \rangle:z-NN} c_4 \sigma_i \sigma_j + \sum_i h \sigma_i + E_0, \quad (1)$$

The c_i parameters were obtained *ab initio*, (see suppl. inf. and references [69–72] therein) and take the values $c_1 = 32.495$ meV, $c_2 = -17.68$ meV, $c_3 = 5.166$ meV and $c_4 = -14.232$ meV. The MC derived atomic configuration of the $\sqrt{3} \times \sqrt{3} \times 2$ order is plotted in fig. 3(b) with patches of *zig-zag* dimerization along the c -direction, fig. 3(d). The Fourier transform of such Ge₁ pattern [73, 74] is displayed in fig. 3(f) and nicely reproduces the experimental x-ray diffraction pattern.

On the other hand, the $\sqrt{3} \times \sqrt{3} \times 3$ order requires a modified Ising hamiltonian, since the dimers need to be considered as 2 distinct Ge₁ atoms, suppl. inf. Appendix D. Moreover, the model requires a c_5 coefficient in the eq. 1 that accounts for the second nearest neighbor interaction along the c -direction,

$$\sum_{\langle i,j \rangle:z-NNN} c_5 \sigma_i \sigma_j, \quad (2)$$

The MC simulation reproduces the atomic pattern displayed in fig. 3(c), where the Ge₁ dimerizes diagonally along the c -direction, fig. 3(e). Remarkably, the Fourier transform of such Ge₁ configuration, presented in fig. 3(g), shows Bragg spots at the $(\frac{1}{3} \frac{1}{3} \frac{1}{3})$ wavevector, again in good agreement with the experimental data at ~ 25 GPa, hence, the MC simulations capture in full detail the cascade of order-disorder transitions in FeGe under hydrostatic pressure.

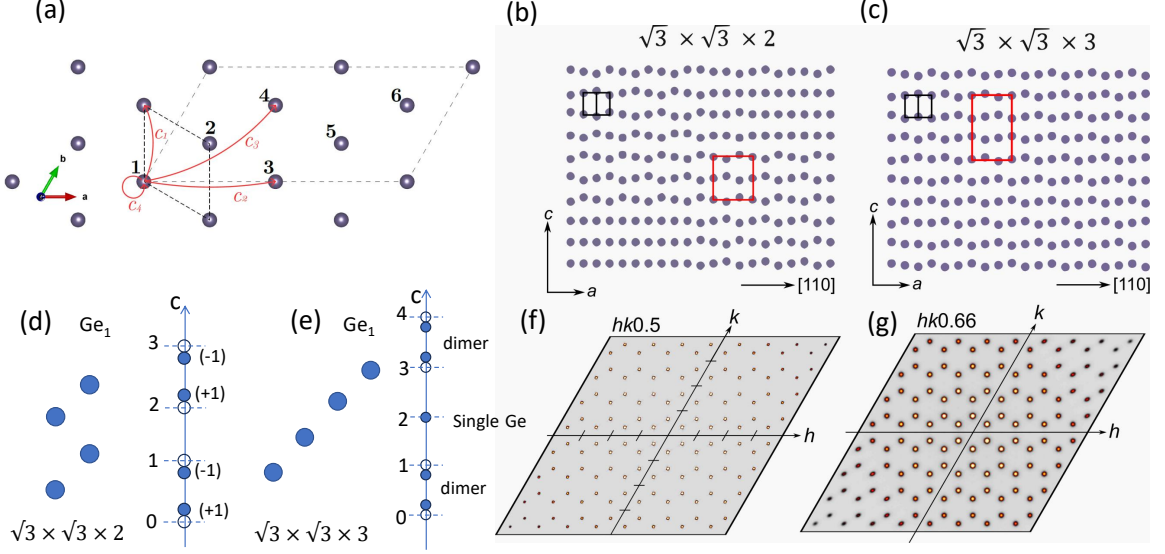


Figure 3. (a) Sketch of the model used for the MC simulations, where the interaction parameters of the Ising Hamiltonian are described as $c_{i's}$. (b) Atomic arrangement of the trigonal Ge_1 atoms derived from the MC simulations using the *ab initio* $c_{i's}$ for the $\sqrt{3} \times \sqrt{3} \times 2$ order. (c) Atomic configuration of the for the $\sqrt{3} \times \sqrt{3} \times 3$ order. The red squares highlight the two-fold and three-fold superlattice along the c -direction. (d) and (e) describe the dimerization arrangement along the c -direction for the for the $\sqrt{3} \times \sqrt{3} \times 2$ and for the $\sqrt{3} \times \sqrt{3} \times 3$ orders, respectively. +1 (-1) stands for the upward (downward) displacement of the Ge_1 . (f) and (g) Fourier transform of (b) and (c), respectively, showing the $\frac{1}{3} \frac{1}{3} \frac{1}{2}$ and $\frac{1}{3} \frac{1}{3} \frac{1}{3}$ superlattice peaks.

Microscopically, understanding the pressure dependence of the CDW in FeGe represents a challenge. As shown in fig. 4(a), the phonon spectra, computed using the frozen phonon method [75–83] remain stable within the harmonic approximation and most of the phonon modes harden with p (suppl. inf. fig. 8). However, the phonon at the H point, characterized by the irreducible representation (irrep) $H3$ [77], remains nearly constant in energy. This $H3$ phonon corresponds to the dimerization of triangular Ge_1 atoms of the $\sqrt{3} \times \sqrt{3}$ order, similarly as the lowest phonon at the L point (irrep $L2-$) at low pressure, fig. 4(a), (suppl. inf. fig. 8). We have carried out DFT calculations to investigate the total energy of different CDW structures at various pressures. As shown in fig. 4(b), the 2×2 CDW structure has lower energy, consistent with existing experimental results [50, 63]. However, as pressure increases, the total energy of the $\sqrt{3} \times \sqrt{3} \times 2$ superstructure decreases more rapidly and becomes the ground state above 18 GPa, in good agreement with our experimental observations. On the other hand, the $\sqrt{3} \times \sqrt{3} \times 3$ order presents higher energy at the DFT level, see fig. 4(b), in contrast to the experimental observations, hinting at the possible crucial role of the lattice anharmonicity at high p . The 2×2 to $\sqrt{3} \times \sqrt{3} \times 2$ phase transition can be theoretically described by the Landau-Ginzburg theory, which incorporates the coupling between the order parameters asso-

ciated with the two distinct CDW phases, as described in suppl. inf. Appendix E [84]. The phase diagram obtained from Landau-Ginzburg analysis is presented in fig. 4(c), where the two tuning parameters m_ϕ and m_ψ represent the masses of the respective order parameters: ϕ for 2×2 CDW and ψ for $\sqrt{3} \times \sqrt{3} \times 2$ CDW. This phase diagram illustrates that a phase transition occurs from a 2×2 order to a phase where both CDW orders coexist, and eventually to a $\sqrt{3} \times \sqrt{3} \times 2$ CDW phase at high p .

The rotation of the in-plane propagation vector and the tripling of the c -axis with p , although confirmed by DFT and MC simulations, is a remarkable result, which deserves further analysis. The calculated band structures of antiferromagnetic FeGe at $p = 0$ and 15 GPa are shown in (suppl. inf. fig. 6) fig. 4(d). Roughly, as pressure increases, the van Hove singularities at the M and L points shift away from the Fermi energy E_F , while the electron pockets at the K point expand with p . This expansion does not enhance the Fermi surface nesting at different K points with respect to the low p phase, as shown by the non-interacting charge susceptibility plot, $\chi_{\mathbf{q}}$ (suppl. inf. fig. 7). Nevertheless, the peak in $\chi_{\mathbf{q}}$ at K suggests that charge correlations might be central in the emergence of the $\sqrt{3} \times \sqrt{3}$ CDW order. We would like to finish with a brief discussion about the weakening of the CDW intensities of the $\sqrt{3} \times \sqrt{3} \times 2$ order below T_{CDW} . One possibility might be related to the local deformations induced by the large dimerization of the Ge_1 atoms that could act

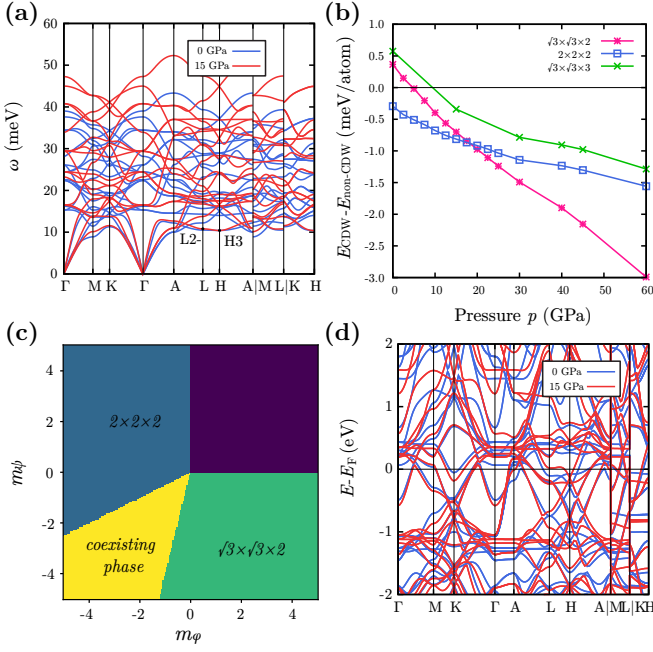


Figure 4. (a) Phonon spectra at 0 and 15 GPa. Note that the phonon spectra are calculated in the antiferromagnetic phase and plotted in the paramagnetic BZ. (b) The difference of total energy between the CDW structure and non-CDW structure in the antiferromagnetic phase versus p for $\sqrt{3} \times \sqrt{3}$ and 2×2 orders. (c) Phase diagram derived from the Landau-Ginzburg free energy. (d) Band structure of antiferromagnetic FeGe at 0 and 15 GPa.

as strong pinning centers of the mobile carriers leading to the formation of standing waves, i.e., Friedel oscillations (FOs) [85–87]. The modulations observed might simply be due to FOs around local sources of disorder that screen the local distortions and stabilize the charge fluctuations near T_{CDW} at $2k_F$. Below T_{CDW} , the quasi-long-range order might suppress the disorder and gives rise to a drop of the $\sqrt{3} \times \sqrt{3}$ intensity [88], as we have observed in fig. 3.

In summary, we have carried out a high-pressure x-ray diffraction and DFT study in the antiferromagnetic kagome metal FeGe that reveals quasi-long-range $\sqrt{3} \times \sqrt{3}$ order with propagation vectors $\mathbf{q}^* = (\frac{1}{3}, \frac{1}{3}, \frac{1}{2})$ and $\mathbf{q}^\dagger = (\frac{1}{3}, \frac{1}{3}, \frac{1}{3})$, that compete with the low-pressure 2×2 ordered phase. The $\sqrt{3} \times \sqrt{3}$ orders are described by a large

dimerization of the trigonal Ge_1 atoms in the kagome plane and modeled by Monte Carlo simulations based on the Ising hamiltonian of frustrated triangular lattices and demonstrate the strong tunability of the fragile ground state of FeGe under pressure.

Note added: During the completion of this manuscript, we became aware of related results reporting a possible high pressure $\sqrt{3} \times \sqrt{3} \times 6$ phase in annealed FeGe crystals by Xikai Wen et al. [89].

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