

Charge order through crystallization of Frenkel excitons: realization in kagome metals

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ABSTRACT

Charge order is a widely observed and representative example of spontaneous broken symmetries in quantum states of matter. Owing to the large intra-atomic Coulomb energy, the charge redistribution in such an order typically implies significant alteration of the electronic and lattice properties of materials. While the standard description of charge order, namely a “charge density wave” instability of the Fermi surface, has been broadly and successfully applied to good metals, its applicability to correlated ionic materials has been rather limited. Here, we propose an alternative general scenario of charge order - crystallization of long-lived Frenkel excitons - suitable for these ionic materials. We demonstrate this scenario on the recently discovered kagome superconductors and successfully reproduce all the characteristics of experimental observations on both local charge correlations and long-range ordering. The proposed generic scenario offers a long-sought understanding of charge order applicable to modern correlated functional materials.

Key points: Charge order, Charge-density wave, Frenkel exciton, Crystallization, Local correlation, Long-range order.

Introduction

The phenomenon of charge order (CO) has been widely observed in a variety of ionic materials (in which at least some atoms have a well-defined effective ionic valence), including manganates^{1,2}, nickelates^{3,4}, cobaltates^{5,6}, cuprates⁷⁻⁹, kagome metals¹⁰⁻¹², and transition metal dichalcogenides¹³⁻¹⁵. Due to the large energy scale of the Coulomb interaction, the charge redistribution and long-range ordering associated with the formation of CO are often found to strongly couple to observed physical properties such as transport^{16,17}, magnetism^{18,19}, and other symmetry-broken phases²⁰. Highlighting systems displaying superconductivity, besides the obvious competition in long-range ordering^{21,22}, the relationship between short-range charge and superconducting correlations remains an open question that attracts intensive research activities^{23,24}.

The standard description of charge order, namely the theory of charge density waves (CDWs), is based on interaction-induced Fermi surface instabilities²⁵⁻³⁴, and it naturally applies to good metals in which the kinetic energy dominates the electronic structure. In semimetals with low carrier densities and in semiconductors with small gaps, these instabilities can be further formulated through the formation of Wannier excitons³⁵⁻³⁹, which can even drive the system into an excitonic insulator phase^{35,40} under strong enough electron-hole binding energies. Nonetheless, applicability of these established mechanisms is rather limited for ionic materials, in which intra-atomic electronic interactions are comparable to or even stronger than inter-atomic kinetic processes. For example, these Fermi surface based mechanisms, in their conceptually reliable (weakly interacting) regime, require a sufficiently large density of states near the Fermi energy that many of these ionic materials lack. Furthermore, the necessary large kinetic energy scale in these theories leads to a rather rigid ordering wavevector corresponding to the ‘nesting’ of the Fermi surface that often does not match the experimentally observed ordering wavevector^{28,32,41-44}.

The limitations of the standard theoretical framework are becoming increasingly apparent with recent significant improvements in experimental capability. In NbSe₂, for instance, scanning tunneling microscopy reveals short-range charge correlations at temperatures far exceeding the transition temperature¹³. Such short-range correlations are further verified by X-ray diffraction⁴⁵ and inelastic X-ray scattering^{46,47}. Other signatures of short-range correlations, including phonon anomalies with broad peaks near the ordering wave vectors, have been observed in manganites⁴⁸ and cuprates⁴⁹. Moreover, the differential pair distribution function analysis of resonant X-ray scattering reveals such local lattice distortions even *prior to* the long-range coherence of charge density, magnetism, Kondo resonance, or superconductivity in many materials⁵⁰⁻⁵⁴. In TaS₂, Raman spectroscopy further confirms substantial lattice distortions⁵⁵, while ARPES studies reveal the persistence of the CDW

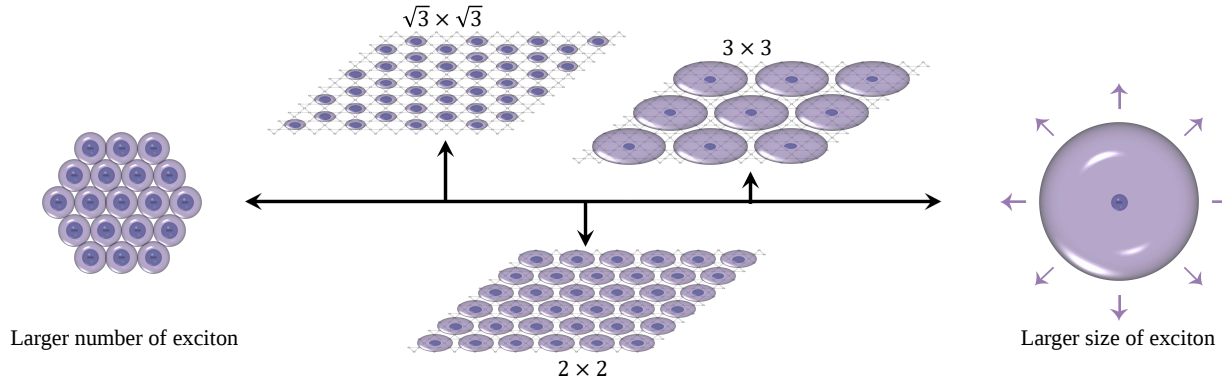


Figure 1. *Illustration of charge order via crystallization of Frenkel excitons.* Given atomic orbital energies, two mechanisms compete to determine the lattice of exciton crystal. Potential energy prefers higher density of small excitons (toward left), while excitons’ internal kinetic energy tends to enlarge the exciton (toward right). Constrained by the Pauli principle and repulsive interaction of excitons’ outer structure, a close-packed lattice results to optimize the competition.

energy gap above the transition temperature⁵⁶. These observations all point to the existence of some microscopic mechanism at a relatively high energy scale that drives the short-range charge correlation in these ionic materials way beyond the temperature scale of the long-range ordering, below which these short-range correlations become coherent across the macroscopic domains.

An important clue for the identification of this microscopic mechanism is the observation that many materials exhibit multiple patterns of charge ordering under strain with similar volumes, such as 2×2 , 4×1 , and 3×3 ^{41,42}. This suggests the key is some short-range physics with characteristic *volume* that dictates the charge correlation, before the long-range order develops at lower energy scale^{50–54}. Such a variety of patterns is inconsistent with the Fermi surface nesting mechanism which should be insensitive to strain due to the Luttinger theorem^{57,58}. This indicates that such charge orders requires an alternative *general* volume-sensitive paradigm applicable at higher energy- and shorter length-scale in these ionic materials.

In this work, we propose a novel general mechanism for charge order that is applicable to ionic systems hosting strong short-range correlations: the crystallization of long-lived Frenkel excitons⁵⁹. We illustrate this mechanism in detail in the recently discovered kagome superconductors, CsV_3Sb_5 ⁶⁰, and then show that it also offers a natural explanation for the distinct charge orders in ScV_6Sn_6 . Finally, we compare the generic characteristics of this new Frenkel exciton mechanism with those of the standard Fermi surface instability, which suggests the broad applicability of our theory as a general paradigm in understanding various physical behaviors in a wide scope of ionic systems displaying charge order.

Charge order through crystallization of long-lived Frenkel excitons

We build our proposed mechanism starting with the following scenario. First, we consider ionic systems that are energetically unstable when the effective valence of ions is uniformly integer across the sample. That is, we consider systems that can lower their energy by allowing some degree of local charge transfer between neighboring ions, effectively generating Frenkel excitons to stabilize the system. Here, the exact microscopic mechanism, such as electronic or electron-lattice interactions, is unessential as long as its energy scale is large. Second, we focus on cases with charge fluctuations that are not strong enough to suppress the reversal of the charge transfer, namely systems with long-lived local Frenkel excitons having low annihilation rate.

Under these conditions, the system would spontaneously generate some number of Frenkel excitons according to the optimal balance between the potential and kinetic energies of the system. As illustrated in Fig. 1, in the limit where the potential energy of exciton formation dominates (toward left), the system prefers a large number of excitons. In the opposite limit where the kinetic energy of the shorter-time internal dynamics of excitons dominates (toward right), the system prefers excitons of larger size. In parallel, the internal electron-hole structure of the excitons implies that their outer regions are subject to the Pauli exclusion principle and Coulomb interaction, leading to an effective “hard core” for each exciton. Combined, all these considerations lead to a final exciton configuration that provides an optimal balance between a higher density of smaller excitons and a smaller number of larger excitons. Naturally, at low enough temperature such optimization can be reached through the formation of a close-packed periodic lattice of excitons. Such crystallization of long-lived Frenkel excitons therefore displays the charge order observed in the experiments.

Frenkel exciton crystallization is a fundamentally different mechanism for charge order compared to Fermi surface instabilities, and therefore one expects qualitatively distinct phenomenology depending on the driving mechanism. Below, we first illustrate this generic scenario using the kagome superconductor CsV_3Sb_5 as an example, and then discuss the distinguishing features of charge order driven by Frenkel exciton crystallization in contrast to those of the standard mechanism of Fermi surface instability.

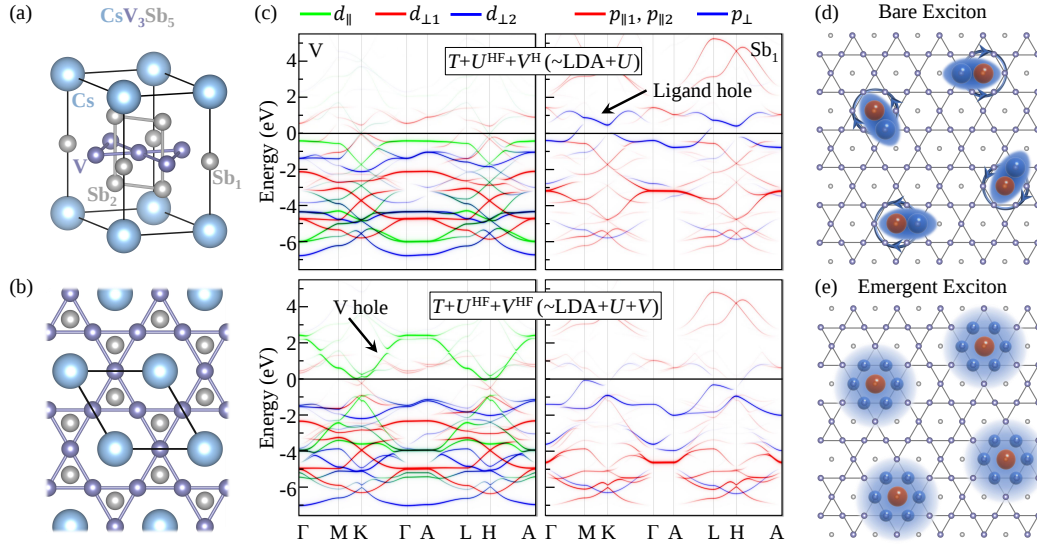


Figure 2. Formation of long-lived Frenkel excitons. (a) and (b) illustrate the lattice structure of CsV₃Sb₅. (c) The LDA+ U band structure of the majority spin in the upper panel gives a uniform charge distribution, corresponding to an effective $|d^3 d^3 d^3 \bar{L}\rangle$ configuration. Improving it with additional inter-atomic interaction V results in charge transfer from the in-plane V- $d_{\parallel}$ orbital to the Sb₁- $p_{\perp}$ orbital. The resulting $|d^2 d^3 d^3 \rangle$ configuration corresponds to the formation of a *bare* Frenkel exciton as a tightly bound particle-hole pair whose recombination is forbidden by their distinct z -parity, as illustrated by an *instantaneous* snap shot in (d). (e) Upon absorbing the short-time dynamics of the holes, the *emergent* long-lived Frenkel excitons would contain a electron in the center and a hole-cloud around it in the same plane.

Demonstration using kagome superconductor CsV₃Sb₅

Charge order has recently been discovered in kagome metals AV₃Sb₅ ($A = \text{K, Rb, Cs}$)^{60,61}, whose structure consists of an unusual combination of honeycomb, triangular, and kagome sublattices hosting Dirac cones in the band dispersion. These materials became the subject of intense research interest with the discovery of superconductivity^{62–64}, and multiple superconducting phases were later found under pressure^{65–67}. Furthermore, these materials display anomalous Hall and Nernst effects^{68–72} and anomalous transport and magnetic behaviors⁷³ which cast doubt on the applicability of Fermi liquid theory and consequently on the Fermi surface instability mechanism for the description of their emergent phenomena.

Focusing on their unconventional charge order, long-range order occurs ranging from 80 to 100 K in the AV₃Sb₅ family^{61,74–78}. Most experiments find the charge ordering phase transition to be likely first-order^{79–81}. Interestingly, the charge order is found to be accompanied by a simultaneously broken rotational symmetry^{82–89} and sometimes time-reversal symmetry^{10,90–98}. Most notably, alongside the prominent 2×2 lattice reconstruction, a clear signature of 4×1 order is also observed^{83,92,93,99}, which is inconsistent with the nesting wavevector of the Fermi surface of the system. This family therefore serves as a good candidate to explore our proposed scenario of Frenkel exciton crystallization.

Existence of long-lived excitons

Using CsV₃Sb₅ as a representative example¹⁰⁰, we start by examining whether the conditions for long-lived Frenkel excitons required for our proposed mechanism are satisfied. In a previous LDA+ U study¹⁰¹ with realistic intra-atomic interaction strength $U = 0.6 \text{ Ry}$ and $J = 0.06 \text{ Ry}$, it was found that the out-of-plane ligands Sb₂ [c.f. Fig. 2(a)(b) and SI¹⁰⁰] develop covalent bonds across the planes and contribute to *massless* Dirac-type carriers. On the other hand, the in-plane ligands (surrounded by six V ions) are strongly ionic, each with a hole in their $p_{\perp}$ orbitals. Indeed, as shown in the upper panels of Fig. 2(c), the presence of a ligand hole, $\bar{L}$, is made clear by the half-filled blue Sb₁ $p_{\perp}$ band in both spin-majority and minority (c.f. SI¹⁰⁰) channels of the in-plane Sb ions in the right panel. The spin-majority channel of V shown in Fig. 2(c) also indicates that *locally* V ions are in an *effective* $\text{V}^{2+} d^3$ spin-3/2 configuration with fully spin-polarized effective t_{2g} orbitals, as expected from the Hund's rules. Given that correlations, such as those driving exciton formation, are most pronounced in massive orbitals, the relevant low-energy degrees of freedom in these materials are therefore the V t_{2g} orbitals and Sb₁ $p_{\perp}$ orbitals, in the $|d^3 d^3 d^3 \bar{L}\rangle$ configuration.

To properly describe the binding of Frenkel excitons, we improve on the LDA+ U calculation by including the nearest

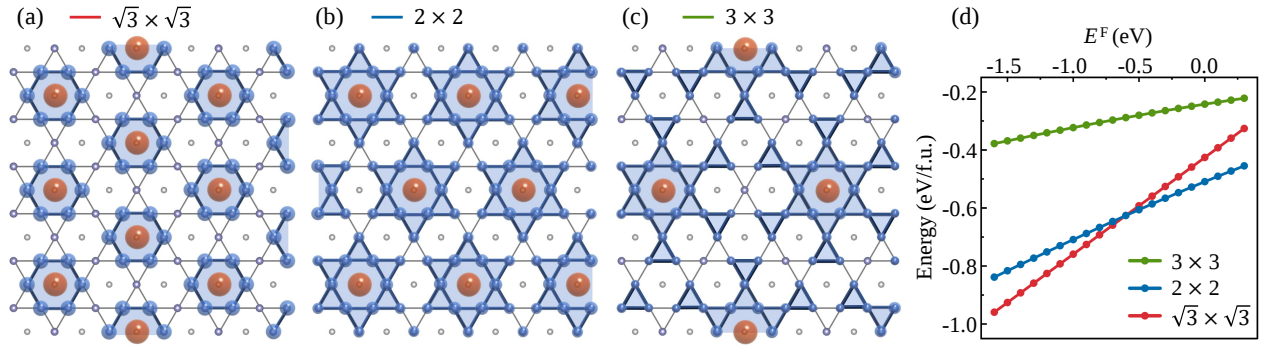


Figure 3. *Energetic optimization of exciton crystallization.* Consider three close-packed lattices of rotational symmetric emergent Frenkel excitons with different size (a) $\sqrt{3} \times \sqrt{3}$, (b) 2×2 , and (c) 3×3 under various strength of binding energy with fixed orbital energy and kinetic strengths. (d) The total energy per formula unit (f.u.) indicates that the $\sqrt{3} \times \sqrt{3}$ lattice is energetically favored at strong binding (lower formation energy E^F), while with weaker binding ($E^F > -0.7$ eV) the 2×2 lattice takes over to benefit from the kinetic energy at lower density.

neighbor (self-interaction-free) Coulomb repulsion V :

$$H^{\text{eff}} = \sum_{i,m,v} \epsilon_m c_{imv}^\dagger c_{imv} + \sum_{i,i',m,m',v} t_{im,i'm'} c_{imv}^\dagger c_{i'm'v} + \frac{1}{2} \sum_{i,m,m',m'',v,v'} U_{mm'm''} c_{imv}^\dagger c_{im'v}^\dagger c_{im''v'} c_{im'v} + \frac{1}{2} \sum_{i,i',m,m',v,v'} V_{im,i'm'} c_{imv}^\dagger c_{i'm'v}^\dagger c_{i'm'v'} c_{imv}. \quad (1)$$

in addition to the one-body orbital energy ϵ , kinetic hopping strength t , and intra-atomic interaction U among the V- d orbitals, denoted by creation $c_{imv}^\dagger$ and annihilation c_{imv} operators of orbitals m and spin v within unit cell i .

The lower panels of Fig. 2(c) display the resulting orbital-projected one-body spectral functions obtained from a self-consistent Hartree-Fock calculation under a realistic inter-atomic interaction $V = 0.07$ Ry. The results, which are unfolded¹⁰² to the original unit cell from a 2×2 supercell that allows the occurrence of zero to four Frenkel excitons, reveal that the original $|d^3 d^3 d^3 \underline{L}\rangle$ configuration is not energetically the most stable, consistently with the first condition of our scenario. Instead, relocating a ligand hole from the blue band to the green band of V ions, effectively a $|d^2 d^3 d^3\rangle$ ionic configuration, is energetically favorable. From a chemical point of view, this is unsurprising given that Sb with a full p -shell should have some energetic advantage. Relative to the uniform $|d^3 d^3 d^3 \underline{L}\rangle$ reference state, this state corresponds to the spontaneous occurrence of tightly bound Frenkel excitons, with the (electron, hole)-pair residing in neighboring (Sb_1 - $p_\perp$, V- $d_\parallel$) orbitals shown in Fig. 2(d).

Importantly, annihilation of this *bare* Frenkel exciton via charge fluctuations that recombine the electron and hole is *forbidden* by symmetry. Specifically, the Sb_1 - $p_\perp$ orbital that hosts the electron has odd z -parity and thus has *no* charge motion to the even z -parity V- $d_\parallel$ orbital that hosts the hole. As a consequence, this particular exciton exhibits exceptional longevity, satisfying the second condition of our scenario. Once thermally generated, these bare Frenkel excitons cannot undergo dynamical number fluctuations, a crucial characteristic for stabilizing long-lived phenomena such as charge order.

For such long-lived phenomena, it is physically more intuitive and convenient to absorb the rapid dynamics into the internal structure of larger *emergent* Frenkel excitons associated with longer timescales. In this example, the electrons in the Sb_1 - $p_\perp$ orbital have restricted out-of-plane kinetics, given the highly ionic Cs ions right above and below them. Similarly, the dominant hopping for holes in V- $d_\parallel$ are also in-plane due to its spatial distribution. Furthermore, the in-plane kinetic processes of the V- $d_\parallel$ orbitals (~ 600 meV) are more than an order of magnitude more efficient than those (~ 20 meV) of the Sb_1 - $p_\perp$ orbitals. Consequently, as illustrated in Fig. 2(d), a hole in the former would rapidly orbit around a localized electron in the latter. As shown below, at longer timescales the emergent long-lived Frenkel exciton would contain an electron at the Sb_1 site and a hole cloud around it in the same plane, as illustrated in Fig. 2(e). Such emergent Frenkel excitons align perfectly with the experimental real-space pattern of charge profile and the local density of states¹⁰.

Crystallization for optimal energy

Given that both conditions of our proposed scenario are fulfilled in CsV_3Sb_5 , we proceed to determine the optimal balance between the kinetic energy that favors larger excitons and the potential energy that favors a larger number of excitons. Recall that it is the exclusive (or repulsive) charge correlation between the outer structure of the excitons that forces these two mechanisms to compete, and therefore one must incorporate this correlation in the overall energy optimization. Without this key correlation, as in Hartree-Fock mean-field treatments, as soon as charge transfer is energetically favored, the exciton population would grow to the maximum.

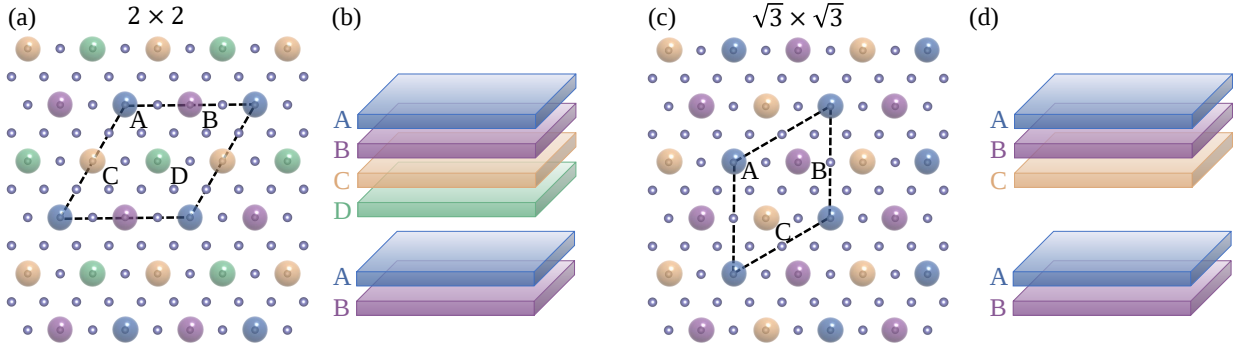


Figure 4. z-axis stacking in the kagome superconductors. (a)(b) show the ABCD $2 \times 2 \times 4$, or imperfectly $2 \times 2 \times 2$ in CsV_3Sb_5 and (c)(d) show the ABC $\sqrt{3} \times \sqrt{3} \times 3$, or imperfectly AB $\sqrt{3} \times \sqrt{3} \times 2$ in ScV_6Sn_6 .

To this end, we simulate the exclusive charge correlation by restricting the internal dynamics of the hole in each exciton to be within a region $\Omega(j)$ centered around an electron $p_j^\dagger$ in the Sb_1 - $p_\perp$ orbital at site j :

$$H = \sum_{\{i,m;l',m'\} \in \Omega(j)} t_{im,l'm'} d_{im}^\dagger d_{l'm'} + \left(\varepsilon \sum_{\{i,m\} \in \Omega(j)} d_{im}^\dagger d_{im} - V^B \sum_{\{i,m\} \in \text{NN}(j)} d_{im}^\dagger p_j^\dagger p_j d_{im} \right), \quad (2)$$

such that the exclusive charge correlation with each other is respected when forming a close-packed lattice for energy optimization. Here, the LDA+ U extracted parameter ε denotes the orbital energy difference between Sb_1 - $p_\perp$ and V - $d_\parallel$ orbitals, and $t_{im,l'm'}$ is the internal kinetic hopping of a *hole*, $d_{im}^\dagger$, between V - $d_\parallel$ orbitals at different locations m of unit cell i . We omit spin indices for simplicity. To illustrate the physical trends, we vary the dominant effective binding energy, V^B , between the nearest-neighboring electron in $p_j^\dagger$ and hole in $d_{im}^\dagger$, such that the formation energy of the nearest neighboring “bare” excitons is $E^F = \varepsilon - V^B$. Below, we further fix $\varepsilon = 0.3$ eV and $t_{im,l'm'}$ from Eq. (1)¹⁰⁰ in order to examine the V^B -dependence of ground-state energies per formula unit within $V^B \in [0, 2]$ eV. The remaining weak external dynamics of the emergent larger exciton will be constrained by the close-packing of crystallization.

The considered cases are $\sqrt{3} \times \sqrt{3}$, 2×2 , and 3×3 shown in Figs. 3(a)-(c), corresponding to exciton densities of 3^{-1} , 4^{-1} , and 9^{-1} per formula unit, respectively. Figure 3(d) then gives the resulting energy comparison for three close-packed lattices of rotation-symmetric emergent Frenkel excitons with different sizes. While higher exciton densities lead to greater potential energy gains, this advantage is offset by reduced kinetic energy benefits. This trade-off becomes particularly evident in the 3×3 lattice, where the low density of large excitons results in both inefficient kinetic energy gain and insufficient binding-associated potential energy, rendering this configuration energetically unfavorable. In contrast, the 2×2 and $\sqrt{3} \times \sqrt{3}$ lattices, with their higher densities of smaller excitons, achieve a more optimal balance between internal kinetic energy and overall potential energy. Specifically, under a strong effective binding, V^B (lower E^F), the higher density of smaller excitons in the $\sqrt{3} \times \sqrt{3}$ lattice is energetically optimal. However, for weaker binding, $E^F > -0.7$ eV, it is more advantageous to have slightly lower density of larger excitons to benefit from the kinetic energy and form a 2×2 exciton lattice, in agreement with the experimentally observed 2×2 charge order¹⁰.

Once the optimal 2-dimensional lattice of the emergent Frenkel excitons is determined via optimizing the in-plane physics, the lower-energy out-of-plane stacking of weakly coupled 2-dimensional layers naturally follows the classical stacking of planes of charged hard spheres. As shown in Fig. 4(a)(b), for a 2×2 lattice of emergent Frenkel excitons, the optimal stacking (with lowest Coulomb energy) would be a 4-period ABCD stacking with excitons centered around one of the four available Sb_1 sites in each layer, corresponding to a $2 \times 2 \times 4$ charge order, in agreement with the experimental observations^{78,103,104}. Alternatively, the imperfect 2-period ABAB ones (with only slightly higher inter-layer Coulomb energy), may also occur as observed experimentally^{74,76,79,105}.

Generic features of charge order driven by Frenkel exciton crystallization

Having illustrated the newly proposed mechanism of Frenkel exciton crystallization as the driving mechanism for charge order in CsV_3Sb_5 , we next explore the universal qualitative characteristics of charge order resulting from this mechanism. Strikingly, these features, which differ fundamentally from those associated with conventional Fermi surface instabilities, show excellent agreement with various anomalous experimental findings in strongly correlated materials exhibiting charge order.

The scenario of Frenkel exciton crystallization starts with high-energy short-range local charge correlations, which develop into coherent long-range order at lower energy scales. The dominant physics in this scenario, namely strong local electron/lattice correlations, is fundamentally distinct from that of the standard Fermi surface instability mechanism^{10,74,106–109},

Table 1. Comparison between Fermi surface instability and Frenkel exciton crystallization mechanisms for charge order.

	<i>Fermi surface instability</i>	<i>Frenkel exciton crystallization</i>
<i>Building block</i>	Long-range coherence	Short-range correlation
<i>Key factor</i>	Density of state at E_F	Coulomb energy
<i>Dominant physics in</i>	Momentum	Position
<i>Sensitive to</i>	Periodicity (wavevector)	Supercell size (density)
<i>Typical period</i>	π/k_F (Incommensurate)	Commensurate / Discommensurate
<i>Density modulation</i>	Harmonic	Atomic
<i>Local spatial symmetry</i>	Typically symmetric	Possibly lower symmetry
<i>Spin texture</i>	No	Possible
<i>Leading fluctuation</i>	Amplitude (pairing)	Phase (coherence)
<i>At $T \approx T_{CDW}$</i>	Local order vanishing	Exciton crystal melting
<i>Phase transition</i>	Second-order	(weakly) First- / Second-order
<i>Phonon frequency near T_{CDW}</i>	Softens to zero	Possibly remains finite
<i>At $T \gg T_{CDW}$</i>	Uniform density	Local deformation remains

characterized by coherent kinetics perturbed by weak interactions or electron-phonon coupling^{110,111}. Therefore, one expects its characteristics, just like the so-called strong coupling scenario³⁴, to be qualitatively distinct from the standard lore. Below, we systematically examine these unique features across energy scales, with key representative properties summarized in Tab. 1.

First, the Frenkel exciton crystallization scenario accepts remarkable flexibility in both spatial patterns and ordering wavevectors. The internal kinetic energy gain of emergent Frenkel excitons depends primarily on the number of internal kinetic processes, making it more sensitive to exciton size than to exciton shape. This size-dependence leads to similar energies for states with comparable exciton densities, regardless of their different lattice configurations. Therefore, unlike the rather robust nesting wavevectors associated with instabilities of the Fermi surface, emergent excitons may crystallize in multiple lattice structures of similar energies corresponding to distinct ordering wavevectors. It is therefore more likely for the Frenkel exciton crystallization scenario to develop charge order with lower rotational symmetry in both the local lattice deformation and the ordering wavevectors¹¹², as frequently observed in kagome metals (KV₃Sb₅¹¹³, RbV₃Sb₅⁹², CsV₃Sb₅^{93,99}), and cuprates (Bi_{2-y}Pb_ySr_{2-z}La_zCuO_{6+x}¹¹⁴, Ca_{2-x}Na_xCuO₂Cl₂¹¹⁵, La_{2-x}Sr_xCuO₄¹¹⁶, YBa₂Cu₃O_y¹¹⁶). Similarly, a change in the ordered excitonic lattice structure can be easily triggered by applied pressure, strain, or fields, as observed in transition metal dichalcogenides (NbSe₂^{41,42}, TiSe₂¹¹⁷, TaS₂¹¹⁸, VSe₂¹¹⁹), or even under lower temperature, as in nickelates (Ba_{1-x}SrNi₂As₂¹²⁰), kagome superconductors (LaRu₃Si₂¹²), transition metal dichalcogenides (NbTe₂¹²¹, TaS₂¹¹⁸), and manganates (Bi_{1-x}Sr_{x-y}Ca_yMnO₃¹²²).

Second, given the rapid decrease of the *additional* internal kinetic energy gain upon further increasing the exciton size, this scenario typically disfavors very large emergent Frenkel excitons. That is, the corresponding charge order typically avoids long-period modulations (small wavevectors.) Similarly, in the quantum (low-temperature) limit, charge-order commensurate with the atomic lattice is typically energetically favored. Also note that when accumulation of stress reaches a critical value, a discommensuration¹²³ would take place to adjust the overall periodicity while still respecting the commensurate local structure. Correspondingly, over the length scale of the ordered period, the spatial density profile is defined mostly by atomic orbitals, rather than by harmonic waves as in charge density waves. Such locally commensurate structure has, in fact, been overwhelmingly observed in correlated materials, such as the transition metal dichalcogenides (TaS₂¹¹⁸, TaSe₂¹²⁴, VSe₂¹²⁴, NbSe₂¹²⁵), manganates (Bi_{1-x}Sr_{x-y}Ca_yMnO₃¹²², La_{1-x}Ca_xMnO₃², Pr_{1-x}Ca_xMnO₃¹²⁶), nickelates (La_{1-x}Sr_xNiO₂¹²⁷), cuprates (YBa₂Cu₃O_y¹²⁸), kagome metals (KV₃Sb₅⁶¹, RbV₃Sb₅¹²⁹, CsV₃Sb₅^{129,130}, ScV₆Sn₆¹³¹), iridium alloys (IrTe₂¹³², Lu₅Ir₄Si₁₀¹³³, Lu₂Ir₃Si₅¹³⁴), and cobaltates (CoO¹³⁵, Na_xCoO₂⁶).

Third, since the dominant physics of the Frenkel exciton crystallization scenario are the interaction-driven local correlations, for charge-ordered states with low itinerant carrier densities the Fermi surface nesting would play a small role in either driving or affecting the charge order. Such disrespect to nesting wavevectors has in fact been observed in many ionic materials⁴³, for example the transition metal dichalcogenides (NbSe₂⁴¹⁻⁴⁴, TaSe₂⁴⁴, TiSe₂²⁸, Cu_xNbS₂⁴⁴). On the other hand, it is known that in the presence of large itinerant carrier densities in ordered states, long-range quantum fluctuations can disrupt the long-range phase coherence of the order¹³⁶, leading towards a quantum unordered phase^{136,137}. In that case, a rekindled long-range order¹³⁸ may re-emerge in the presence of chemical or structural disorder, or another ordering wavevector of similar exciton density can take over. From this consideration, a reasonably nested normal-state Fermi surface compatible with the period of the exciton crystal, *rather than* driving the order, can still be very helpful in allowing the long-range coherence for the order. This coherence perspective of nesting is perhaps more relevant than the traditional nesting picture in ionic materials,

such as transition metal dichalcogenides (TaTe₂¹³⁹, NbSe₂⁴⁴), kagome metals (CsV₃Sb₅¹⁰⁹), nickel pnictide superconductors [Ba(Ni_{1-x}Co_x)₂As₂¹⁴⁰], and cuprates (Bi₂Sr_{2-x}La_xCuO_{6+δ}¹⁴¹).

Fourth, since the formation of local emergent Frenkel excitons is driven by higher-energy local interactions, it is natural that these excitons carry a spin-1 texture, namely opposite spins for the electron and hole components. In that case, the crystallization of such emergent Frenkel excitons would *simultaneously* order in the spin channel (a *single* order parameter having both charge and spin texture.) For example, along the direction of an anti-ferromagnetic order for excitons, the magnetic order would have a period associated with the charge order, as observed in many ionic materials, such as cuprates (La_{2-x}Sr_xCuO₄¹⁴²), nickelates (hole-doped La₂NiO₄¹⁴³), iron-based superconductors (FeSe¹⁴⁴, KFe_{0.8}Ag_{1.2}Te₂¹⁴⁵ and Ba_{1-x}Na_xFe₂As₂¹⁴⁶), topological semimetals (GdSb_xTe_{2-x-δ}¹⁴⁷), transition metal dichalcogenides (Fe_xNbS₂¹⁵), and kagome magnets (FeGe^{11,148}).

Fifth, the rather large charge redistribution associated with the emergent Frenkel excitons would typically dictate a heavy dressing by local lattice distortions, much stronger than in the scenario of Fermi surface instabilities. Even though such strong local deformations can be masked in the ordered component by long-range fluctuations, a significant difference in local bond length can still be observed through pair distribution function analysis of diffraction, nuclear quadrupole resonance, or X-ray absorption fine structure. Such “contradiction” between probes of long- and short-wavelength has in fact been observed in transition metal dichalcogenides (NbSe₂^{13,45-47}, TaS₂^{50,55}, NbTe₄⁵²), manganites (La_{1.2}Sr_{1.8}Mn₂O₇⁴⁸, Pr_{0.5}Sr_{0.5}MnO₃⁵⁴), cuprates (La_{2-x}Sr_xCuO₄⁴⁹), Pt-intermetallics (UPt₂Si₂⁵¹, CePt₂Si₂⁵¹, LaPt₂Si₂⁵¹, LaPt_{2-x}Ge_{2+x}⁵¹), and high-entropy alloys (MoNbTaW⁵³).

Finally, since the formation of bare and emergent excitons is driven by high-energy (above-eV scale) physics, it should be much more robust than the low-energy (~ 10 meV-scale) long-range order. Therefore, the local correlations (and the associated lattice deformations) should persist way above the transition temperature of long-range charge order, as frequently observed in transition metal dichalcogenides (NbSe₂¹³) and cuprates (La_{2-x}Sr_xCuO₄⁴⁹). Correspondingly, the loss of long-range order in this scenario is associated with the melting of the exciton crystal (phase fluctuations) accompanied by exciton density fluctuations, in great contrast to the rapid vanishing of particle-hole pairs (amplitude fluctuations) associated with the Fermi surface instability mechanism. Therefore, the finite-temperature phase transition of the former can often have a first-order nature, possibly with finite phonon frequencies, while it is typically second-order in the latter, with phonon softening to zero frequency. This naturally explains the observation in kagome metals (CsV₃Sb₅⁷⁹⁻⁸¹, ScV₆Sn₆¹³¹, FeGe¹⁴⁸), iridium alloys (IrTe₂¹³², Lu₅Ir₄Si₁₀¹³³, Lu₂Ir₃Si₅¹³⁴), and transition metal dichalcogenides (NbSe₂⁴⁶, TaSe₂⁴⁶).

Recall that within the renormalization group framework^{149,150} of phases and their transitions, a system can only flow to *one* particular stable fixed point that captures all the essential characteristics of the phase. As summarized in Tab. 1, the characteristics of this scenario are qualitatively distinct from those of the Fermi surface instability scenario (via either Fermi surface nesting or electron-phonon coupling). In other words, the two scenarios correspond to different fixed points. Therefore, for materials such as kagome metals whose characteristics fit this scenario well, the latter offers a qualitatively incorrect physical picture, and its resulting ordering wavevector, even if in agreement with experiment, carries little physical significance.

Resolving the puzzling ordering wavevector in ScV₆Sn₆

The scenario of Frenkel exciton crystallization offers a natural resolution to the puzzling periodicity of the observed charge order in the bilayer kagome ScV₆Sn₆, another member of the same V-based kagome superconductor family as CsV₃Sb₅ discussed above. On the one hand, similar to the latter, nearly all observed physical properties¹⁵¹ of ScV₆Sn₆ coincide with the above generic characteristics. On the other, while the nearly nested Fermi surface of ScV₆Sn₆ resembles very much that of CsV₃Sb₅, the observed¹⁵² in-plane charge ordering wavevector, $\sqrt{3} \times \sqrt{3}$, is drastically distinct from the more nesting-compatible 2×2 wavevector observed in CsV₃Sb₅.

Given the above-mentioned strong local lattice distortion associated with their formation, naturally the Frenkel excitons in the bilayer structure of ScV₆Sn₆ have a strong tendency toward interlayer binding for an energetically more compatible structure between layers. Such a bound interlayer “bi-exciton” would have an enhanced effective binding (lower E^F) and a reduced kinetic strength t in Eq. (2). As demonstrated in Fig. 3(d), both of these effects would favor a smaller exciton size, from 2×2 (blue) to $\sqrt{3} \times \sqrt{3}$ (red), thus offering a qualitative explanation for the puzzling nesting-incompatible ordering wavevector.

The proposed exciton crystallization also intuitively explains the other puzzle on the variable out-of-plane period of the observed charge correlation, namely a $\sqrt{3} \times \sqrt{3} \times 2$ short-range order coexisting with a $\sqrt{3} \times \sqrt{3} \times 3$ long-range order¹⁵¹⁻¹⁵⁵. Similar to the case of CsV₃Sb₅, the out-of-plane stacking of the bi-exciton layers simply follows a lower-energy closed packing. Specifically, as shown in Fig. 4(c)(d), in contrast to the four allowed sites for stacking in the 2×2 excitonic lattice in panel (a)(b), the $\sqrt{3} \times \sqrt{3}$ excitonic lattice has three allowed sites. In turn, the chiral ABC stacking, consistent with the observed $\sqrt{3} \times \sqrt{3} \times 3$ period, would have the lowest-energy. As in the case of CsV₃Sb₅, slightly higher-energy ABAB stacking is also locally stable, thus the presence of the observed short-range order.

Conclusions

In summary, to address the long-standing issue of unconventional charge order in functional materials, we propose an alternative general scenario - crystallization of long-lived Frenkel excitons - applicable to ionic systems hosting strong short-range correlation. We demonstrate this mechanism via the recently discovered kagome superconductors, CsV_3Sb_5 , which naturally offers an intuitive explanation for the distinct charge-order patterns in ScV_6Sn_6 . Furthermore, we compare the generic characteristics of this mechanism against those of the standard picture of Fermi surface instability, which indicates a broad applicability of this mechanism as a unifying paradigm for understanding diverse charge-ordered phenomena in a wide range of ionic systems. The proposed framework thus offers a long-sought theoretical complement to modern descriptions on unconventional charge order in correlated functional materials.

Technical Review

Please see supplementary for details.

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