

Orbital moiré and quadrupolar triple- q physics in a triangular latticeKazumasa Hattori *Department of Physics, Tokyo Metropolitan University, 1-1, Minami-osawa, Hachioji, Tokyo 192-0397, Japan*Takayuki Ishitobi *Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan*

Hirokazu Tsunetsugu

The Institute for Solid State Physics, The University of Tokyo, Kashiwanoha 5-1-5, Chiba 277-8581, Japan

(Received 4 August 2024; accepted 4 December 2024; published 26 December 2024)

We numerically study orders of planar type ($xy, x^2 - y^2$) quadrupoles on a triangular lattice with nearest-neighbor isotropic J and anisotropic K interactions. This type of quadrupole possesses unique single-ion anisotropy proportional to a third order of the quadrupole moments. This provides an unconventional mechanism of triple- q orders which does not exist for the degrees of freedom with odd parity under time-reversal operation such as magnetic dipoles. In addition to several single- q orders, we find various orders including incommensurate triple- q quasi-long-range orders with orbital moiré and a four-sublattice triple- q partial order. Our Monte Carlo simulations demonstrate that the phase transition to the latter triple- q state belongs to the universality class of the critical line of the Ashkin-Teller model in two dimensions close to the four-state Potts class. These results indicate a possibility of realizing unique quadrupole textures in simple triangular systems.

DOI: [10.1103/PhysRevResearch.6.L042068](https://doi.org/10.1103/PhysRevResearch.6.L042068)

Introduction—Superposition of multiple standing waves creates a moiré texture, and this is recognized in our daily life, e.g., when looking at overlapped fences or lace curtains swaying in the wind. It is known as notorious wavy patterns in photography and its post processing [1]. This visually fascinating texture is characterized by multiple wave vectors $\{\mathbf{q}_\ell\}$. Recent evolution of “moiré engineering” has been developed [2,3] for twisted graphene multilayers [4–6] and transition metal dichalcogenides [7–9], and this opens a way to manipulate their functionality and physics itself such as superconductivity [4], nonreciprocal phenomena [10], flat bands [11], and magnetoelectric effects [12,13].

Another realization of moiré structure uses topological texture of internal degrees of freedom. Representative examples are superfluid He [14], various types of superconductors [15–17], Bose-Einstein condensates in cold atoms [18–20], and exciton-polariton condensates [21–23]. This started in magnetic systems from the seminal work about quasi long-range orders in planar magnets [24], and this has opened a way toward topological physics. The discovery of skyrmion crystal in MnSi [25–27] triggered the current explosion of researches which aim at realizing moiré structure [28]. Such a spin moiré [29,30] is also realized by a crystallization of various topological defects [31–33]. Clarifying their prop-

erties is important for understanding the fundamental physics and also for developing the next-generation technology.

Spatial modulations of a moiré structure are characterized by a set of ordering vectors $\{\mathbf{q}_\ell\}$ ($\ell = 1, 2, \dots$). This requires a multiple- q order of order parameter, and control of the ordering vectors $\{\mathbf{q}_\ell\}$ is an important issue. Ferromagnets with noncentrosymmetric structure have Dzyaloshinskii-Moriya interaction [34] and it shifts the ordering vector $\mathbf{q}$ from 0 to finite incommensurate (IC) values. Such an IC ordering is also established by other mechanisms such as geometrical frustration [35], Fermi-surface effects in itinerant electron systems [36,37], and anisotropic interactions [38–40]. These varieties of possibility open a wider range of material research for realizing multiple- q orders [41].

Recently, several studies have tried realizing multiple- q orders of nonmagnetic degrees of freedom φ such as electric charge [42,43] and electric quadrupole [44–46]. These order parameters are time-reversal even, which differs from magnetic ones. This makes a crucial difference in their free energy from the magnetic counterpart [43,47–50], and, for example, the cubic terms $\varphi_\alpha \varphi_\beta \varphi_\gamma$ are allowed. Such terms may arise from a single-ion anisotropy and favor triple- q orders, since the term $\propto \varphi_\alpha(\mathbf{q}_1) \varphi_\beta(\mathbf{q}_2) \varphi_\gamma(\mathbf{q}_3) \delta_{\mathbf{q}_1 + \mathbf{q}_2 + \mathbf{q}_3, \mathbf{0}}$ lowers the free energy by properly choosing the phase and direction of each $\varphi(\mathbf{q}_\ell)$. In magnetic systems, such couplings are possible only when a finite magnetic field is applied [38]. Thus, nonmagnetic systems have a potential to exhibit multiple- q physics at zero magnetic field, and provide a good playground for the moiré engineering.

In this Letter, we propose a minimal model and demonstrate that nonmagnetic systems are useful for realizing moiré

textures. The model is defined for electric quadrupoles on a triangular lattice with nearest-neighbor interactions, and we have performed Monte Carlo simulations for its effective classical Hamiltonian. In addition to varieties of phases, we have found IC triple- q quasi long-range orders relevant to the two-dimensional Ashkin-Teller and Potts models with the partial ordering of quadrupole moments.

Model—We consider a triangular lattice with the hexagonal site symmetry of the point group D_6 or C_{6v} . In this case, the order parameter with the symmetry of Γ_5 (E_2) doublet is particularly interesting, as will be explained below. This is the two-component quadrupole (ϕ_u, ϕ_v) with the symmetry of $(2xy, x^2 - y^2)$, and we ignore magnetic sectors, which might be present in real materials, for simplicity. We use a classical approximation to represent this quadrupole moment by a unit vector pointing to the direction denoted by θ_r ,

$$\phi_r = (\phi_{u,r}, \phi_{v,r}) =: \hat{e}(\theta_r), \quad \hat{e}(\theta) \equiv (\cos \theta, \sin \theta), \quad (1)$$

where $\mathbf{r}$ is the site position. We will use $\hat{e}$ also for other vectors along with $\hat{e}_\ell \equiv \hat{e}(\ell\omega)$ for the special angle $\omega \equiv \frac{2\pi}{3}$. This classical description is valid when its finite temperature properties are discussed. These two-component quadrupoles possess a local cubic coupling as a single-ion anisotropy:

$$V = \lambda \sum_r (3\phi_{u,r}^2 - \phi_{v,r}^2) \phi_{v,r} = \lambda \sum_r \sin 3\theta_r. \quad (2)$$

Without loss of generality we can set $\lambda \geq 0$. Its microscopic origin is discussed in the Supplemental Material [51] for models with the doublet local ground state $E_2: \{2xy, x^2 - y^2\}$ and a singlet excited state $A_1: 3z^2 - r^2$.

Now, let us introduce exchange couplings for the quadrupole moments. Each nearest-neighbor pair ϕ_r and ϕ_{r+d} interacts via two types of exchange couplings: an isotropic J and an anisotropic K one, as

$$\mathcal{H}_{\text{ex}} = \sum_{r,d} \sum_{ij} [(J - K)\delta_{ij} + 2K d_i d_j] \phi_{i,r+d} \phi_{j,r}. \quad (3)$$

Here, $\mathbf{r}$ runs over the sites on the triangular lattice. $\mathbf{d}$ runs over a half of the nearest neighbors $\mathbf{d} \equiv (d_u, d_v) \in \{\hat{e}_n\}_{n=0}^2$. Then, the Hamiltonian is the sum of the two terms $\mathcal{H} = \mathcal{H}_{\text{ex}} + V$. Note that the orbital degrees of freedom are nothing but anisotropic charge distribution, thus they usually possess anisotropy in their Hamiltonian. See Refs. [52–54], for example. This contrasts with the magnetic systems where the single site anisotropy originates from small spin-orbit couplings.

Ordering vectors—Let us first examine the single- q spiral orders $\phi_r \propto \text{Re } \phi(\mathbf{q}^*) \exp(\pm i \mathbf{q}^* \cdot \mathbf{r})$, and determine the value of ordering vector $\mathbf{q}^*$ [51]. One can easily find that the order is ferroic ($\mathbf{q}^* = \mathbf{0}$) when $2J < -|K|$. When $-|K| < 2J < 5|K|$, the order is commensurate antiferroic (AF) and $\mathbf{q}^*$ locates at one of the edge centers (the M points) in the Brillouin zone (BZ): $\mathbf{Q}_\ell = \frac{2\pi}{\sqrt{3}} \hat{e}[(\frac{1}{4} - \ell)\omega]$ ($\ell = 1, 2, 3$). An interesting part is the IC state for the region of $5|K| < 2J$. As $|K|$ decreases, $\mathbf{q}^*$ leaves $\mathbf{Q}_\ell$ and moves along the BZ edge, $\mathbf{q}_\ell^* = \mathbf{Q}_\ell + \Delta_M \hat{e}_{4-\ell}$ with $\cos(\Delta_M/2) = (2J + |K|)/(4J - 4|K|)$, and it reaches the BZ corner [K or K' point; $\mathbf{q}_K = 2\omega \hat{e}_0$ and $\mathbf{q}_{K'} = 2\omega \hat{e}(\frac{5}{2}\omega)$] when $K=0$. As shown in Figs. 1(a) and 1(b), the lowest-energy eigenmodes $\phi(\mathbf{q}_\ell^*)$ differ between distinct edges, but

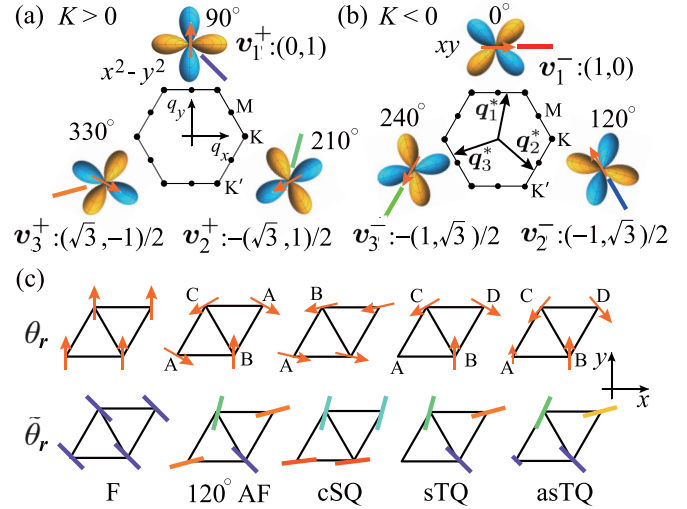


FIG. 1. Lowest-energy eigenmode $\phi(\mathbf{q}^*)$ of $\mathcal{H}_{\text{ex}}$ for $\mathbf{q}_\ell^*$ on the BZ edges for (a) $K > 0$ and (b) $K < 0$. Within each edge, the eigenmode does not change and the quadrupole moment is illustrated together with the corresponding pseudospin (arrow for angle θ) and nematic director (bar for angle $\bar{\theta} = -\frac{1}{2}\theta$). (c) Representative configurations of θ_r and $\bar{\theta}_r$ in commensurate orders: ferroic (F), 120° antiferroic (120° AF), canted single- q (cSQ), symmetric triple- q (sTQ), and asymmetric triple- q (asTQ). A–D are the sublattice indices

remain unchanged on each edge irrespective of Δ_M : $\phi(\mathbf{q}_\ell^*) \propto \mathbf{v}_\ell^+ = \hat{e}[(\ell - \frac{1}{4})\omega]$ or $\mathbf{v}_\ell^- = \hat{e}_{\ell-1}$.

Ferroic, AF 120°, and triple- q orders—Now that $\mathbf{q}_\ell^*$'s are determined, the classical solution of ϕ_r is readily obtained. However, the cubic potential (2) causes nontrivial modifications. Before their discussion, let us analyze simpler parts, i.e., the ferroic (F) order for $2J < -|K|$ and the 120° AF order for $5|K| < 2J$. See Fig. 1(c). Owing to the cubic term (2), these phases have no continuous degeneracy, and the moment's direction is locked at $\theta_r = (n_r + \frac{3}{4})\omega$ with $n_r \in \{0, 1, 2\}$.

For $0 < 2J < 5K$, the simplest candidate is the symmetric triple- q (sTQ) state. The three ordering vectors locate at $\mathbf{Q}_\ell$'s with an identical amplitude $\phi_\ell \equiv |\phi(\mathbf{Q}_\ell)|$ for all ℓ . Note that $\phi(\mathbf{Q}_\ell)$'s are all real since $-\mathbf{Q}_\ell \equiv \mathbf{Q}_\ell$. The $\mathbf{v}_\ell^+$ mode has a lower energy of $\mathcal{H}_{\text{ex}}$ than the $\mathbf{v}_\ell^-$ mode, and this comprises the triple- q configuration

$$\phi_r = \sum_{\ell=1,2,3} \phi_\ell \cos(\mathbf{Q}_\ell \cdot \mathbf{r}) \sigma_\ell \mathbf{v}_\ell^+, \quad \sigma_\ell \in \{+1, -1\}. \quad (4)$$

Substituting this with $\phi_\ell = \frac{1}{2}\phi > 0$ in Eq. (2), one finds $V = -\frac{3}{4}N\lambda\phi^3\sigma_1\sigma_2\sigma_3$, where N is the number of sites. This term is minimized when all three or otherwise one of σ_ℓ 's is +1. This energy gain readily guarantees that the first-order transition occurs at a temperature higher than that of a continuous transition to the single- q state in the simple Landau analysis. This sTQ state indeed has a four-sublattice *partial order* with $\{\phi_r\} = \{\mathbf{0}, \phi \mathbf{v}_1^+, \phi \mathbf{v}_2^+, \phi \mathbf{v}_3^+\}$, as shown in Fig. 1(c). The four sets of $\{\sigma_\ell\}$ correspond to domains with the disordered site on different sublattices.

For $5K < 2J < 0$, one may expect a similar sTQ state, but the situation is quite different. Employing Eq. (4) with $\mathbf{v}_\ell^+ \rightarrow$

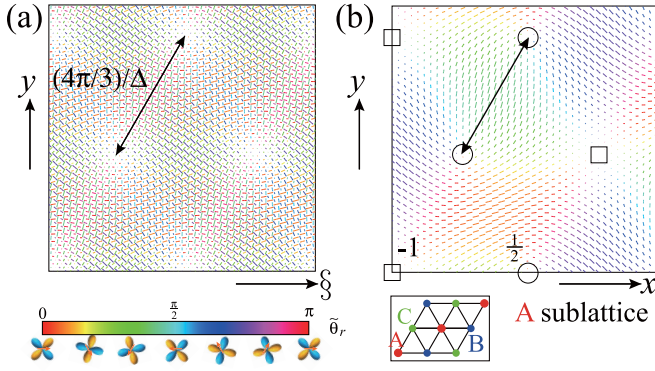


FIG. 2. (a) Director angle configuration $\tilde{\theta}_r$ in the triple- $\mathbf{q}$ state with $\mathbf{q}_\ell^* \sim \mathbf{q}_{K'} - \Delta \hat{\mathbf{e}}_{4-\ell}$ with $\Delta = \frac{\pi}{24}$. (b) Half-vortices ($\circ$) form a honeycomb lattice, and vortices ($\square$) are positioned at the center of each hexagonal unit.

$\mathbf{v}_\ell^-$ results in $V = 0$. Thus, the sTQ state is not stable for $K < 0$ in contrast to the case of $K > 0$.

Parasitic ferro moments—The cubic term (2) affects the order parameters significantly. Using the eigenmodes $\mathbf{v}_\ell^+$ for $K > 0$, we find that V contains the following term:

$$-\frac{3}{2}\lambda \mathbf{D} \cdot \boldsymbol{\phi}(\mathbf{0}), \quad \mathbf{D} = (\sqrt{3}(\phi_1^2 - \phi_2^2), 2\phi_1^2 - \phi_2^2 - \phi_3^2). \quad (5)$$

Once the symmetry $\phi_1 = \phi_2 = \phi_3$ is broken, $\mathbf{D}$ is activated, and this induces the ferro moment $\boldsymbol{\phi}(\mathbf{0})$ [49,50]. This is natural since in the sTQ state $\boldsymbol{\phi}_r$'s are disordered at a quarter of the sites and thus this state is not stable at $T = 0$. Such an asymmetric triple- $\mathbf{q}$ (asTQ) state is illustrated in Fig. 1(c) for the case of $\phi_1 > \phi_2 = \phi_3$, and $\boldsymbol{\phi}(\mathbf{0}) \parallel \hat{\mathbf{e}}(\frac{\pi}{2})$. In general, the $\mathbf{v}_\ell^+$ and $\mathbf{v}_\ell^-$ components hybridize, and this rotates $\boldsymbol{\phi}_r$. See $\boldsymbol{\phi}_r$ in the C and D sublattices in Fig. 1(c).

Even for single- $\mathbf{q}$ states, such a ferroic moment emerges. Consider the single- $\mathbf{q}$ state for $K < 0$ with $\mathbf{q}^* = \mathbf{Q}_\ell$. The AF mode $\boldsymbol{\phi}(\mathbf{Q}_\ell) \equiv \phi_\ell \hat{\mathbf{e}}(\theta_\ell)$ can couple with the ferro mode $\boldsymbol{\phi}(\mathbf{0}) = \phi_0 \hat{\mathbf{e}}(\theta_0)$, and this yields $V \sim 3N\lambda\phi_\ell^2\phi_0 \sin(2\theta_\ell + \theta_0)$. This shows that the ferro component $\boldsymbol{\phi}(\mathbf{0})$ points to the direction $\theta_0 = \frac{3}{2}\pi - 2\theta_\ell$, and thus, the canted single- $\mathbf{q}$ (cSQ) state is stabilized as shown in Fig. 1(c). For $K < 0$, $\theta_\ell = (\ell - 1)\omega$, and thus, $\boldsymbol{\phi}(\mathbf{0}) \parallel \hat{\mathbf{e}}[(\frac{3}{4} + \ell)\omega]$.

IC triple- $\mathbf{q}$ —Now, we study the triple- $\mathbf{q}$ states for $K > 0$ with the ordering vectors $\{\mathbf{q}_\ell^*\}_{\ell=1}^3$ on the BZ edges. The cubic term is important, as it stabilizes the IC triple- $\mathbf{q}$ (IC-TQ) states. Since $-\mathbf{q}_\ell^* \neq \mathbf{q}_\ell^*$, the factor $\sigma_\ell \cos(\mathbf{Q}_\ell \cdot \mathbf{r})$ in Eq. (4) should be replaced by $\cos(\mathbf{q}_\ell^* \cdot \mathbf{r} + \eta_\ell)$ with a general phase η_ℓ . Then, $V = -\frac{3}{4}N\lambda\phi_1\phi_2\phi_3 \cos(\eta_1 + \eta_2 + \eta_3)$, and this is minimized when $\eta_1 + \eta_2 + \eta_3 \equiv 0$. This energy gain guarantees that the IC-TQ state is more stable than the single- $\mathbf{q}$ state for sufficiently large λ .

Such triple- $\mathbf{q}$ configurations can be visualized as *orbital moiré*. Figure 2(a) shows an example of “nematic” director angles $\tilde{\theta}_r \equiv -\frac{1}{2}\theta_r \pmod{\pi}$. They point to the direction of the orbital node: $\tilde{\theta}_r = 0$ for the xy orbital and $\frac{3}{4}\pi$ for $x^2 - y^2$. When $\mathbf{q}_\ell^*$'s are near the K or K' point, the configuration is characterized by a set of sublattice directors $\tilde{\theta}_{r \in X}$ ($X = A, B$, or C). Then, we introduce their vector vortex charges $\mathbf{n} = (n_A, n_B, n_C)$

defined by [55]

$$n_X(\mathbf{r}) \equiv \oint_{r+r' \in X} \frac{d\mathbf{r}'}{2\pi} \cdot \frac{\partial}{\partial \mathbf{r}'} \tilde{\theta}_{r+r'}, \quad (6)$$

where the integral is defined for a closed loop encircling the site $\mathbf{r}$. For the data in Fig. 2, the ordering vectors are chosen as $\mathbf{q}_\ell^* = \mathbf{q}_{K'} - \Delta \hat{\mathbf{e}}_{4-\ell}$ with $\Delta = \frac{\pi}{24}$. The $\tilde{\theta}_r$ configuration in Fig. 2(a) shows a triangular lattice of vortices and half-vortices with the approximate periodicity $4\pi/(3\Delta)$. Their vector vortex charges are either $\mathbf{n}_1 = (-1, \frac{1}{2}, \frac{1}{2})$, $\mathbf{n}_2 = (\frac{1}{2}, -1, \frac{1}{2})$, or $\mathbf{n}_3 = (\frac{1}{2}, \frac{1}{2}, -1)$. Figure 2(b) shows the director configuration in the A sublattice. One can easily see that half-vortices ($n_A = \frac{1}{2}$) form a honeycomb structure, with vortices ($n_A = -1$) being located at the center of each hexagon. Similarly, when $\mathbf{q}_\ell^*$'s are close to the M points, the moiré texture is characterized by four-sublattice vortices [51]. These moiré textures have cores where $|\boldsymbol{\phi}_r| \simeq 0$. These cores are energetically unfavored at low temperatures, similar to the disordered sites in the sTQ state. They are common characteristics of the triple- $\mathbf{q}$ states at finite temperatures [35].

Monte Carlo simulation—We have performed classical Monte Carlo (MC) simulations to examine the effects of thermal fluctuations on the triple- $\mathbf{q}$ orders discussed above. We have used hexagonal clusters with the edge length $L \leq 96$, and the maximum one has $N = 27937$ sites. The periodic boundary conditions are applied for the three pairs of opposite edges. We use the exchange MC algorithm [56,57] combined with single-spin flip updates for efficient simulations. At each temperature T , we calculate observables by averaging over $M = 10^6$ MC steps (MCS) after thermalization of typically 10^5 MCS. We parametrize $(J, K) = \bar{J}(\cos \alpha, \sin \alpha)$ with $\bar{J} = 1$.

Let $\{\boldsymbol{\phi}_r^{(m)}\}$ denote the snapshot at the m th MCS and define their Fourier component by $\phi_\sigma^{(m)}(\mathbf{q}_\ell) \equiv N^{-1} \sum_r \mathbf{v}_\ell^\sigma \cdot \boldsymbol{\phi}_r^{(m)} e^{-i\mathbf{q}_\ell \cdot \mathbf{r}}$ ($\sigma = \pm$) for those $\mathbf{q}_\ell$'s on the ℓ th BZ edge. Since $\phi_\sigma^{(m)}(\mathbf{q}_\ell)$'s may have arbitrary phases, we define the single- $\mathbf{q}$ order parameter as $\mathcal{S}^\sigma(\mathbf{q}_\ell) := M^{-1} \sum_m |\phi_\sigma^{(m)}(\mathbf{q}_\ell)|^2 =: \langle |\phi_\sigma(\mathbf{q}_\ell)|^2 \rangle$, while that for the triple- $\mathbf{q}$ order is defined as $\mathcal{T}^\sigma(\{\mathbf{q}_\ell\}_{\ell=1}^3) \equiv \langle \phi_\sigma(\mathbf{q}_1) \phi_\sigma(\mathbf{q}_2) \phi_\sigma(\mathbf{q}_3) \rangle$ with $\mathbf{q}_1 + \mathbf{q}_2 + \mathbf{q}_3 \equiv \mathbf{0}$.

Figure 3(a) shows the T - α phase diagram for $\lambda = 1$. Similar results have been obtained for $\lambda \gtrsim 0.5$ [51]. There appear six ordered phases in total. Their transition temperature is mostly determined as the temperature where the Binder ratio of the order parameter coincides for $L = 32$ and 48 . For first-order transitions, it is identified to the temperature at which the order parameter jumps upon lowering T . Near $\alpha = 0$, the 120° AF order is stable and its transition from the high- T disordered phase is first order. This is fully consistent with the results of the analysis based on the Landau theory [51]. The presence of six 120° AF domains suggests that the transition is described by the six-state Potts model, which is known to exhibit a first-order transition [58]. For the F and cSQ states, the finite-size scaling analysis shows that the transition is of the three-state Potts universality class [51]. For the sTQ, we find that the universality class of the transition belongs to that on the critical line of the Ashkin-Teller model [59]. This is apparent in both triple- $\mathbf{q}$ $\mathcal{T}^+(\{\mathbf{Q}_\ell\}_{\ell=1}^3)$ and single- $\mathbf{q}$ $\mathcal{S}^+(\mathbf{Q}_\ell)$ order parameters as shown in Fig. 3(b) for $\alpha = 0.375\pi$. The critical exponents determined are $\nu \simeq 0.71$ and $\beta = \nu/8$, where the latter relation holds along the critical line. As α varies, ν

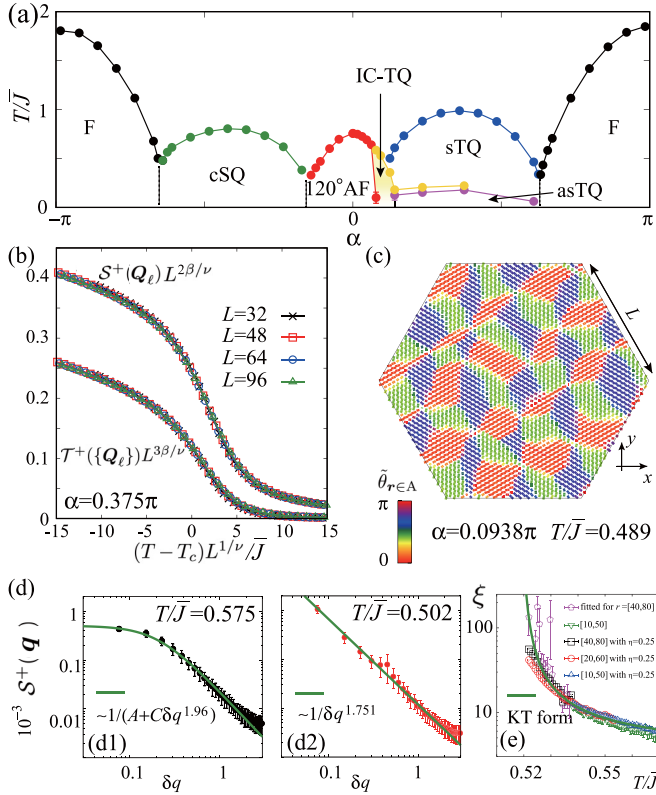


FIG. 3. (a) T - α phase diagram. The low- T region of the IC-TQ state, situated between the 120° and the sTQ states, is expected to be the 120° AF one, because $\mathbf{q}^*$ moves to $\mathbf{q}_{K,K'}$ as T decreases. (b) Finite-size scaling analysis of $S^+(\mathbf{Q}_\ell)$ and $T^+(\{\mathbf{Q}_\ell\})$ for $\alpha = 0.375\pi$ with the choice $(\nu, \beta) = (0.71, 0.089)$, where the transition temperature is assumed $T_c/\bar{J} \simeq 0.9853$. (c) Snapshot $\tilde{\theta}_r$ in the A sublattice for $L = 50$, $\alpha = 0.0938\pi$, and $T/\bar{J} = 0.489$, where the IC-TQ configuration with $\mathbf{q}_\ell^* = \mathbf{q}_{K'} - \frac{4\pi}{3} \frac{3}{50} \hat{\mathbf{e}}_{4-\ell}$ is realized. (d) $S^+(\mathbf{q})$ vs $\delta\mathbf{q} = \mathbf{q} - \mathbf{q}^*$ for (d1) $T > T^*$ and (d2) $T < T^*$ for $L = 96$, where the data along two directions $\delta\mathbf{q} \parallel \hat{\mathbf{e}}(\ell\omega)$ and $\parallel \hat{\mathbf{e}}[(\ell - \frac{1}{4})\omega]$ are averaged. The data are fitted in the region of $0.5 \leq |\delta\mathbf{q}| \leq 2.0$ by a function shown in the figure. (e) T dependence of correlation length ξ for $L = 96$ obtained by fitting $\chi_{uu}(r\hat{\mathbf{e}}_0) = c_1 \cos[(c_2 + \omega)r]r^{-c_3} e^{-r/\xi}$. The fitting range is $10 \leq r \leq 50$ (blue, green) for higher T , while $20 \leq r \leq 60$ (red) and $40 \leq r \leq 80$ (black, pink) for lower T . The green curve is the KT form $\xi = a_1 \exp[a_2/\sqrt{T - T^*}]$, with $a_{1,2}$ being a constant. Data fitted with fixed $c_3 = 1/4$ (expected in the KT form) and those with c_3 being a free parameter are similar, while the low- T data for the latter have large error bars (pink).

changes slightly, but its value is close to that for the four-state Potts universality class corresponding to the end point of the critical line: $\nu = \frac{2}{3}$ and $\beta = \frac{1}{12}$ [58,60]. These results are related to the fourfold degeneracy of the disordered site position. A similar situation has been discussed in the literature for such an example of N_2 molecules adsorbed on Kr-plated graphite [61,62]. Between the sTQ and 120° AF states, the IC-TQ appears at finite T . However, it becomes unstable at lower T 's since a finite density of disordered sites has no contribution to energy gain. Note that the “incommensurate” is just nominal and means $\mathbf{q}^*$ is neither on the K(K') nor M point along the edge of the BZ.

IC-TQ and orbital moiré—In the IC-TQ state, the ordering vector $\mathbf{q}^*$ moves along the BZ edge with lowering T , and this is similar to the case of the Devil's staircase in frustrated magnets [63]. The varying $\mathbf{q}^*$ is triggered by forming dislocations of the vortex lattice [51]. Figure 3(c) shows a snapshot sublattice configuration of $\tilde{\theta}_r$ averaged over 1000 MCS. It is well described by a hexagonal moiré texture with its sublattice vortices $n_A = -1$ and $\frac{1}{2}$ (Fig. 2). The short-time averaging makes the vortex cores clearer than in a single snapshot. For $\alpha \lesssim \tan^{-1}\frac{2}{5} \simeq \alpha_c$, the 120° AF order seems to appear at the lowest temperature. Near $\alpha = \alpha_c$, the MC sampling is not sufficient due to the presence of many local minima related to the IC configurations. We expect $\mathbf{q}^*$ approach $\mathbf{q}_{K,K'}$ and the IC-TQ states are unstable at lower temperatures. These IC-TQ states intervene between the sTQ and the asTQ states for $\alpha \gtrsim \alpha_c$. However, we should be aware that the vector $\mathbf{q}^*$ depends on L and the boundary conditions for these IC-TQ states. Recalling that the phases η_ℓ 's correspond to the choice of the origin of IC oscillation [29], it is natural to expect their fluctuations to destroy the IC long-range order [24,64–66]. Indeed, one evidence of such a quasi-long-range order is the power-law behavior of the correlation function of $\phi(\mathbf{q})$ shown in Fig. 3(d), where the mass term A in $S^+(\mathbf{q}) \sim (A + C|\mathbf{q} - \mathbf{q}^*|^2)^{-1}$ vanishes below the transition temperature $T^*/\bar{J} \simeq 0.518$. The dependence on $\delta\mathbf{q} = \mathbf{q} - \mathbf{q}^*$ is obtained by fitting the data for $0.5 < |\delta\mathbf{q}| < 2.0$ near T^* , resulting in $S^+(\mathbf{q}) \sim |\delta\mathbf{q}|^{-\zeta}$ with $\zeta \simeq 1.6 \sim 2.0$ [51]. These are not far from the value $\zeta = 2 - 1/4 = 1.75$ at the Kosterlitz-Thouless (KT) transition [24]. The real space correlation $\chi_{ij}(\mathbf{r}) = \langle \phi_{i,r} \phi_{j,0} \rangle$ shows an exponential decay above T^* . Figure 3(e) shows that its correlation length ξ is well fitted by the exponential divergence typical in the case of the KT transition as $\xi \propto \exp[\text{const.}/\sqrt{T - T^*}]$ [51]. In addition, the data of T^+ indicate that all the configurations of $\{\eta_\ell\}$ are realized, which leads to a relation $\langle \phi'_+(\mathbf{q}_1) \phi'_+(\mathbf{q}_2) \phi'_+(\mathbf{q}_3) \rangle = -\langle \phi'_+(\mathbf{q}_1) \phi''_+(\mathbf{q}_2) \phi''_+(\mathbf{q}_3) \rangle$. Here, ϕ' (ϕ'') denotes the real (imaginary) part of ϕ . In this sense the phases $\{\eta_\ell\}$ are not locked [51].

Discussion—Let us discuss how to detect the predicted quadrupolar orders. Below, we assume that the system has a weak three-dimensionality with a uniform stacking of ϕ_r along the z axis and the IC-TQ states have true long-range orders. The most direct and powerful method effective for the quadrupole orders is the resonant x-ray scattering experiments [67,68]. The sTQ order is identified by observing disordered sites, and one can use nuclear magnetic or quadrupole resonance for that purpose if appropriate nuclei exist, as has been discussed for UNi_4B [46]. One can also detect free quadrupole moments at the disordered sites by ultrasonic experiments. When a quadrupole order parameter spatially modulates [69], an electric polarization field $\mathbf{P}(\mathbf{r})$ is induced in proportion to $\boldsymbol{\pi}(\mathbf{r}) \equiv (\partial_y \phi_{u,r} + \partial_x \phi_{v,r}, \partial_x \phi_{u,r} - \partial_y \phi_{v,r}, 0)$ and it is detectable. See a recent observation of polar skyrmions in $\text{PbTiO}_3/\text{SrTiO}_3$ [70].

When the system has itinerant electrons, they feel a moiré of ordered quadrupoles. This should be reflected in the band structure, and moiré minibands have been recently observed in twisted bilayer WSe_2 by the resonant inelastic light scattering experiment [71]. Since the IC-TQ order lacks the local inversion symmetry, one expects that the spin-orbit coupling

is activated, and it transcribes the orbital texture into a moiré of magnetizations $\mathbf{M}(\mathbf{r})$. It is induced by electric current $\mathbf{j}$ via the interaction energy

$$\Delta E = g_1(\mathbf{M}_q \times \boldsymbol{\pi}_{-q})_z j_z + g_2 M_{zq}(\boldsymbol{\pi}_{-q} \times \mathbf{j})_z, \quad (7)$$

where g_1 and g_2 are constants. $\boldsymbol{\pi}_q$ and $\mathbf{M}_q$ are the Fourier transform of $\boldsymbol{\pi}(\mathbf{r})$ and $\mathbf{M}(\mathbf{r})$, respectively. In this Letter we have not considered magnetic degrees of freedom such as magnetic dipole $M_z(\mathbf{r})$ perpendicular to the triangular plane, and octupole moments $\sim M_x(\mathbf{r})[M_x^2(\mathbf{r}) - 3M_y^2(\mathbf{r})]$ or $M_y(\mathbf{r})[M_y^2(\mathbf{r}) - 3M_x^2(\mathbf{r})]$. If they exist, they may order around the cores of quadrupole vortices, and the overall configuration is a hybrid quadrupole-dipole/octupole skyrmion(-like) lattice. Such a dipole-quadrupole skyrmion has been proposed for a spin-1 system with an elevated SU(3) symmetry [72]. See also recent analyses [73–77]. Entropy release of the magnetic degrees of freedom is large at the sites with disordered quadrupole and it helps such exotic phases to persist even at low temperatures, as demonstrated for multiple- q multipole orders [44,46,78,79]. Searching for such states is one of the fascinating directions for the future.

Searching materials exhibiting the orbital moiré is interesting. To the best of our knowledge, no such materials have

been known so far. Promising local degrees of freedom are a set of partially filled $\{2xy, x^2 - y^2\}$ type- d electrons with the excited $3z^2 - r^2$ orbital or $\{yz, zx\}$ dominated non-Kramers doublet [80] in d^2 configurations in triangular systems [51]. Several non-Kramers f -electron systems are also interesting: an orbital triple- q order was reported for the stacked triangular-lattice compound UPd₃ [81] similar to the sTQ, while IC orbital orders were reported for the cubic compound PrPb₃ [69]. We hope for discovery of compounds exhibiting various types of triple- q physics through material search, paying special attention to the above-mentioned local degrees of freedom in triangular systems.

Summary—We have proposed a minimal quadrupole model which realizes orbital moiré texture on the triangular lattice. This model exhibits various triple- q orders, and they are stabilized by the cubic anisotropy that is forbidden in magnetic systems due to time-reversal symmetry. We have shown the emergence of incommensurate triple- q states that exhibits an orbital moiré texture with quasi-long-range order. We expect our results pave a basis for the future investigation for nonmagnetic multiple- q and moiré physics.

Acknowledgment. This work was supported by JSPS KAKENHI (Grants No. JP21H01031, No. JP23K20824, and No. JP23H04869).

- [1] I. Amidror, *The Theory of the Moiré Phenomenon* (Springer Science & Business Media, London, 2009), Vol. 1.
- [2] Y. Xiao, J. Liu, and L. Fu, Moiré is more: Access to new properties of two-dimensional layered materials, *Matter* **3**, 1142 (2020).
- [3] D. M. Kennes, M. Claassen, L. Xian, A. Georges, A. J. Millis, J. Hone, C. R. Dean, D. N. Basov, A. N. Pasupathy, and A. Rubio, Moiré heterostructures as a condensed-matter quantum simulator, *Nat. Phys.* **17**, 155 (2021).
- [4] M. Yankowitz, S. Chen, H. Polshyn, Y. Zhang, K. Watanabe, T. Taniguchi, D. Graf, A. F. Young, and C. R. Dean, Tuning superconductivity in twisted bilayer graphene, *Science* **363**, 1059 (2019).
- [5] G. Chen, A. L. Sharpe, P. Gallagher, I. T. Rosen, E. J. Fox, L. Jiang, B. Lyu, H. Li, K. Watanabe, T. Taniguchi, J. Jung, Z. Shi, D. Goldhaber-Gordon, Y. Zhang, and F. Wang, Signatures of tunable superconductivity in a trilayer graphene moiré superlattice, *Nature (London)* **572**, 215 (2019).
- [6] W. Wang, G. Zhou, W. Lin, Z. Feng, Y. Wang, M. Liang, Z. Zhang, M. Wu, L. Liu, K. Watanabe, T. Taniguchi, W. Yang, G. Zhang, K. Liu, J. Gao, Y. Liu, X. C. Xie, Z. Song, and X. Lu, Correlated charge density wave insulators in chirally twisted triple bilayer graphene, *Phys. Rev. Lett.* **132**, 246501 (2024).
- [7] H. Li, J. Wu, Z. Yin, and H. Zhang, Preparation and applications of mechanically exfoliated single-layer and multilayer MoS₂ and WSe₂ nanosheets, *Acc. Chem. Res.* **47**, 1067 (2014).
- [8] K. Hippalgaonkar, Y. Wang, Y. Ye, D. Y. Qiu, H. Zhu, Y. Wang, J. Moore, S. G. Louie, and X. Zhang, High thermoelectric power factor in two-dimensional crystals of MoS₂, *Phys. Rev. B* **95**, 115407 (2017).
- [9] M.-L. Lin, Q.-H. Tan, J.-B. Wu, X.-S. Chen, J.-H. Wang, Y.-H. Pan, X. Zhang, X. Cong, J. Zhang, W. Ji, P.-A. Hu, K.-H. Liu, and P.-H. Tan, Moiré phonons in twisted bilayer MoS₂, *ACS Nano* **12**, 8770 (2018).
- [10] J.-X. Lin, P. Siriviboon, H. D. Scammell, S. Liu, D. Rhodes, K. Watanabe, T. Taniguchi, J. Hone, M. S. Scheurer, and J. I. A. Li, Zero-field superconducting diode effect in small-twist-angle trilayer graphene, *Nat. Phys.* **18**, 1221 (2022).
- [11] M. M. Al Ezzi, J. Hu, A. Ariando, F. Guinea, and S. Adam, Topological flat bands in graphene super-moiré lattices, *Phys. Rev. Lett.* **132**, 126401 (2024).
- [12] W.-Y. He, D. Goldhaber-Gordon, and K. T. Law, Giant orbital magnetoelectric effect and current-induced magnetization switching in twisted bilayer graphene, *Nat. Commun.* **11**, 1650 (2020).
- [13] B. Xie, R. Peng, S. Zhang, and J. Liu, Alternating twisted multilayer graphene: generic partition rules, double flat bands, and orbital magnetoelectric effect, *NPJ Comput. Mater.* **8**, 110 (2022).
- [14] V. P. Mineev, Half-quantum vortices, *Low Temp. Phys.* **39**, 818 (2013).
- [15] D. A. Ivanov, Non-Abelian statistics of half-quantum vortices in p -wave superconductors, *Phys. Rev. Lett.* **86**, 268 (2001).
- [16] J. Garaud and E. Babaev, Skyrmionic state and stable half-quantum vortices in chiral p -wave superconductors, *Phys. Rev. B* **86**, 060514(R) (2012).
- [17] A. A. Zyuzin, J. Garaud, and E. Babaev, Nematic skyrmions in odd-parity superconductors, *Phys. Rev. Lett.* **119**, 167001 (2017).
- [18] J. R. Abo-Shaeer, C. Raman, J. M. Vogels, and W. Ketterle, Observation of vortex lattices in Bose-Einstein condensates, *Science* **292**, 476 (2001).
- [19] H. Takeuchi, Quantum elliptic vortex in a nematic-spin Bose-Einstein condensate, *Phys. Rev. Lett.* **126**, 195302 (2021).

- [20] L. Chen, Y. Zhang, and H. Pu, Spin-nematic vortex states in cold atoms, *Phys. Rev. Lett.* **125**, 195303 (2020).
- [21] Y. G. Rubo, Half vortices in exciton polariton condensates, *Phys. Rev. Lett.* **99**, 106401 (2007).
- [22] K. G. Lagoudakis, T. Ostatnický, A. V. Kavokin, Y. G. Rubo, R. André, and B. Deveaud-Plédran, Observation of half-quantum vortices in an exciton-polariton condensate, *Science* **326**, 974 (2009).
- [23] H. Flayac, I. A. Shelykh, D. D. Solnyshkov, and G. Malpuech, Topological stability of the half-vortices in spinor exciton-polariton condensates, *Phys. Rev. B* **81**, 045318 (2010).
- [24] J. M. Kosterlitz and D. J. Thouless, Ordering, metastability and phase transitions in two-dimensional systems, *J. Phys. C* **6**, 1181 (1973).
- [25] U. K. Rössler, A. N. Bogdanov, and C. Pfleiderer, Spontaneous skyrmion ground states in magnetic metals, *Nature (London)* **442**, 797 (2006).
- [26] S. Mühlbauer, B. Binz, F. Jonietz, C. Pfleiderer, A. Rosch, A. Neubauer, R. Georgii, and P. Böni, Skyrmion lattice in a chiral magnet, *Science* **323**, 915 (2009).
- [27] A. Tonomura, X. Yu, K. Yanagisawa, T. Matsuda, Y. Onose, N. Kanazawa, H. S. Park, and Y. Tokura, Real-space observation of skyrmion lattice in helimagnet MnSi thin samples, *Nano Lett.* **12**, 1673 (2012).
- [28] Y. Tokura and N. Kanazawa, Magnetic skyrmion materials, *Chem. Rev.* **121**, 2857 (2021).
- [29] K. Shimizu, S. Okumura, Y. Kato, and Y. Motome, Spin moiré engineering of topological magnetism and emergent electromagnetic fields, *Phys. Rev. B* **103**, 184421 (2021).
- [30] K. Shimizu, S. Okumura, Y. Kato, and Y. Motome, Phase shift, ellipticity, angle, and topological number in skyrmion lattices, *J. Phys.: Conf. Ser.* **2164**, 012069 (2022).
- [31] N. Kanazawa, J.-H. Kim, D. S. Inosov, J. S. White, N. Egetenmeyer, J. L. Gavilano, S. Ishiwata, Y. Onose, T. Arima, B. Keimer, and Y. Tokura, Possible skyrmion-lattice ground state in the B20 chiral-lattice magnet MnGe as seen via small-angle neutron scattering, *Phys. Rev. B* **86**, 134425 (2012).
- [32] S. Ishiwata, T. Nakajima, J. H. Kim, D. S. Inosov, N. Kanazawa, J. S. White, J. L. Gavilano, R. Georgii, K. M. Seemann, G. Brandl, P. Manuel, D. D. Khalyavin, S. Seki, Y. Tokunaga, M. Kinoshita, Y. W. Long, Y. Kaneko, Y. Taguchi, T. Arima, and B. Keimer *et al.*, Emergent topological spin structures in the centrosymmetric cubic perovskite SrFeO₃, *Phys. Rev. B* **101**, 134406 (2020).
- [33] B. Göbel, I. Mertig, and O. A. Tretiakov, Beyond skyrmions: Review and perspectives of alternative magnetic quasiparticles, *Phys. Rep.* **895**, 1 (2021).
- [34] A. Fert, M. Chshiev, A. Thiaville, and H. Yang, From early theories of Dzyaloshinskii–Moriya interactions in metallic systems to today’s novel roads, *J. Phys. Soc. Jpn.* **92**, 081001 (2023).
- [35] T. Okubo, S. Chung, and H. Kawamura, Multiple- q states and the skyrmion lattice of the triangular-lattice Heisenberg antiferromagnet under magnetic fields, *Phys. Rev. Lett.* **108**, 017206 (2012).
- [36] Y. Akagi, M. Udagawa, and Y. Motome, Hidden multiple-spin interactions as an origin of spin scalar chiral order in frustrated Kondo lattice models, *Phys. Rev. Lett.* **108**, 096401 (2012).
- [37] S. Hayami and Y. Motome, Topological spin crystals by itinerant frustration, *J. Phys.: Condens. Matter* **33**, 443001 (2021).
- [38] Z. Wang, Y. Su, S.-Z. Lin, and C. D. Batista, Meron, skyrmion, and vortex crystals in centrosymmetric tetragonal magnets, *Phys. Rev. B* **103**, 104408 (2021).
- [39] R. Yambe and S. Hayami, Effective spin model in momentum space: Toward a systematic understanding of multiple- Q instability by momentum-resolved anisotropic exchange interactions, *Phys. Rev. B* **106**, 174437 (2022).
- [40] R. Yambe and S. Hayami, Anisotropic spin model and multiple- Q states in cubic systems, *Phys. Rev. B* **107**, 174408 (2023).
- [41] M. Hirschberger, S. Hayami, and Y. Tokura, Nanometric skyrmion lattice from anisotropic exchange interactions in a centrosymmetric host, *New J. Phys.* **23**, 023039 (2021).
- [42] B. R. Ortiz, L. C. Gomes, J. R. Morey, M. Winiarski, M. Bordelon, J. S. Mangum, I. W. H. Oswald, J. A. Rodriguez-Rivera, J. R. Neilson, S. D. Wilson, E. Ertekin, T. M. McQueen, and E. S. Toberer, New kagome prototype materials: discovery of KV₃Sb₅, RbV₃Sb₅, and CsV₃Sb₅, *Phys. Rev. Mat.* **3**, 094407 (2019).
- [43] M. M. Denner, R. Thomale, and T. Neupert, Analysis of charge order in the kagome metal AV₃Sb₅A = K,Rb,Cs, *Phys. Rev. Lett.* **127**, 217601 (2021).
- [44] T. Ishitobi and K. Hattori, Triple- q quadrupole-octupole order scenario for PrV₂Al₂₀, *Phys. Rev. B* **104**, L241110 (2021).
- [45] T. Yanagisawa, H. Matsumori, H. Saito, H. Hidaka, H. Amitsuka, S. Nakamura, S. Awaji, D. I. Gorbunov, S. Zherlitsyn, J. Wosnitza, K. Uhlířová, M. Vališka, and V. Sechovský, Electric quadrupolar contributions in the magnetic phases of UNi₄B, *Phys. Rev. Lett.* **126**, 157201 (2021).
- [46] T. Ishitobi and K. Hattori, Triple- Q partial magnetic orders induced by quadrupolar interactions: Triforce order scenario for UNi₄B, *Phys. Rev. B* **107**, 104413 (2023).
- [47] M. B. Walker, C. Kappler, K. A. McEwen, U. Steigenberger, and K. N. Clausen, Triple- q quadrupolar order in UPd₃, *J. Phys.: Condens. Matter* **6**, 7365 (1994).
- [48] A. V. Nikolaev and K. H. Michel, Quantum charge density fluctuations and the phase transition in Ce, *Eur. Phys. J. B* **9**, 619 (1999).
- [49] H. Tsunetsugu, T. Ishitobi, and K. Hattori, Quadrupole orders on the fcc lattice, *J. Phys. Soc. Jpn.* **90**, 043701 (2021).
- [50] K. Hattori, T. Ishitobi, and H. Tsunetsugu, Quadrupole partial orders and triple- q states on the face-centered cubic lattice, *Phys. Rev. B* **107**, 205126 (2023).
- [51] See Supplemental material at <http://link.aps.org/supplemental/10.1103/PhysRevResearch.6.L042068> for the details of the model information, the Landau analysis, and the Monte-Carlo data not shown in the maintext.
- [52] F. Vernay, K. Penc, P. Fazekas, and F. Mila, Orbital degeneracy as a source of frustration in LiNiO₂, *Phys. Rev. B* **70**, 014428 (2004).
- [53] G. Khaliullin, D. Churchill, P. P. Stavropoulos, and H.-Y. Kee, Exchange interactions, Jahn-Teller coupling, and multipole orders in pseudospin one-half d^2 Mott insulators, *Phys. Rev. Res.* **3**, 033163 (2021).
- [54] Changle Liu, Yao-Dong Li, and Gang Chen, Selective measurements of intertwined multipolar orders: Non-Kramers doublets on a triangular lattice, *Phys. Rev. B* **98**, 045119 (2018).

- [55] H. T. Ueda, Y. Akagi, and N. Shannon, Quantum solitons with emergent interactions in a model of cold atoms on the triangular lattice, *Phys. Rev. A* **93**, 021606(R) (2016).
- [56] R. H. Swendsen and J. S. Wang, Replica monte carlo simulation of spin glasses, *Phys. Rev. Lett.* **57**, 2607 (1986).
- [57] K. Hukushima and K. Nemoto, Exchange Monte Carlo method and application to spin glass simulations, *J. Phys. Soc. Jpn.* **65**, 1604 (1996).
- [58] F. Y. Wu, The Potts model, *Rev. Mod. Phys.* **54**, 235 (1982).
- [59] B. Nienhuis, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowitz (Academic, London, 1987), Vol. 11.
- [60] R. J. Creswick and S.-Y. Kim, Critical exponents of the four-state Potts model, *J. Phys. A* **30**, 8785 (1997).
- [61] R. K. P. Zia and D. J. Wallace, Critical behaviour of the continuous n -component Potts model, *J. Phys. A* **8**, 1495 (1975).
- [62] E. Domany, M. Schick, and J. S. Walker, Classification of order-disorder transitions in common adsorbed systems: Realization of the four-state Potts model, *Phys. Rev. Lett.* **38**, 1148 (1977).
- [63] P. Bak and J. von Boehm, Ising model with solitons, phasons, and “the devil’s staircase,” *Phys. Rev. B* **21**, 5297 (1980).
- [64] D. R. Nelson and B. I. Halperin, Dislocation-mediated melting in two dimensions, *Phys. Rev. B* **19**, 2457 (1979).
- [65] S. N. Coppersmith, D. S. Fisher, B. I. Halperin, P. A. Lee, and W. F. Brinkman, Dislocations and the commensurate-incommensurate transition in two dimensions, *Phys. Rev. B* **25**, 349 (1982).
- [66] D. F. Agterberg, M. Geracie, and H. Tsunetsugu, Conventional and charge-six superfluids from melting hexagonal Fulde-Ferrell-Larkin-Ovchinnikov phases in two dimensions, *Phys. Rev. B* **84**, 014513 (2011).
- [67] L. J. P. Ament, M. van Veenendaal, T. P. Devereaux, J. P. Hill, and J. van den Brink, Resonant inelastic x-ray scattering studies of elementary excitations, *Rev. Mod. Phys.* **83**, 705 (2011).
- [68] F. d. Groot, Multiplet effects in x-ray spectroscopy, *Coord. Chem. Rev.* **249**, 31 (2005).
- [69] T. Onimaru, T. Sakakibara, N. Aso, H. Yoshizawa, H. S. Suzuki, and T. Takeuchi, Observation of modulated quadrupolar structures in PrPb_3 , *Phys. Rev. Lett.* **94**, 197201 (2005).
- [70] M. R. McCarter *et al.*, Structural chirality of polar skyrmions probed by resonant elastic x-ray scattering, *Phys. Rev. Lett.* **129**, 247601 (2022).
- [71] N. Saigal, L. Klebl, H. Lambers, S. Bahmanyar, V. Antić, D. M. Kennes, T. O. Wehling, and U. Wurstbauer, Collective charge excitations between moiré minibands in twisted WSe_2 bilayers probed with resonant inelastic light scattering, *Phys. Rev. Lett.* **133**, 046902 (2024).
- [72] N. A. Mikushina and A. S. Moskvina, Dipole and quadrupole skyrmions in $S = 1$ (pseudo)spin systems, *Phys. Lett. A* **302**, 8 (2002).
- [73] Y. Akagi, Y. Amari, N. Sawado, and Y. Shnir, Isolated skyrmions in the CP^2 nonlinear sigma model with a Dzyaloshinskii-Moriya type interaction, *Phys. Rev. D* **103**, 065008 (2021).
- [74] Y. Amari, Y. Akagi, S. B. Gudnason, M. Nitta, and Y. Shnir, CP^2 skyrmion crystals in an $\text{SU}(3)$ magnet with a generalized Dzyaloshinskii-Moriya interaction, *Phys. Rev. B* **106**, L100406 (2022).
- [75] K. Remund, R. Pohle, Y. Akagi, J. Romhányi, and N. Shannon, Semi-classical simulation of spin-1 magnets, *Phys. Rev. Res.* **4**, 033106 (2022).
- [76] H. Zhang, Z. Wang, D. Dahlbom, K. Barros, and C. D. Batista, CP^2 skyrmions and skyrmion crystals in realistic quantum magnets, *Nat. Commun.* **14**, 3626 (2023).
- [77] S. Hayami, Hybrid skyrmion and antiskyrmion phases in polar C_v systems, *Phys. Rev. B* **109**, 054422 (2024).
- [78] S. Hayami and K. Hattori, Multiple- q dipole-quadrupole instability in spin-1 triangular-lattice systems, *J. Phys. Soc. Jpn.* **92**, 124709 (2023).
- [79] H. Zhang and S.-Z. Lin, Multipolar skyrmion crystals in non-Kramers doublet systems, *Phys. Rev. Lett.* **133**, 196702 (2024).
- [80] A. Paramekanti, D. D. Maharaj, and B. D. Gaulin, Octupolar order in d -orbital Mott insulators, *Phys. Rev. B* **101**, 054439 (2020).
- [81] K. A. McEwen, U. Steigenberger, K. N. Clausen, Y. J. Bi, M. B. Walker, and C. Kappler, Neutron scattering study of the antiferroquadrupolar structure and order parameter of UPd_3 , *Physica B Condens. Matter* **213–214**, 128 (1995).