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# 1 **Crystallizing Kagome artificial spin ice**

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15 Artificial spin ices are engineered arrays of dipolarly coupled nanobar magnets. They enable  
16 direct investigations of fascinating collective phenomena from their diverse microstates. However,  
17 experimental access to ground states in the geometrically frustrated systems has proven difficult,  
18 limiting studies and applications of novel properties and functionalities from the low energy states.  
19 Here, we introduce a convenient approach to control the competing dipolar interactions between  
20 the neighboring nanomagnets, allowing us to tailor the vertex degeneracy of the ground states. We  
21 achieve this by tuning the length of selected nanobar magnets in the spin ice lattice. We  
22 demonstrate the effectiveness of our method by realizing multiple low energy microstates in a  
23 Kagome artificial spin ice, particularly the hardly accessible long range ordered ground state – the  
24 spin crystal state. Our strategy can be directly applied to other artificial spin systems to achieve  
25 exotic phases and explore new emergent collective behaviors.

26 Artificial spin ices (ASIs) are exemplar material-by-design systems with intriguing physical  
27 phenomena such as geometrical frustration [1–6], monopole-like excitations [7–10], Coulomb  
28 phase [10,11] and phase transitions [12–14]. They lead to novel functionalities with great potential  
29 for applications, such as low-power data storage [15], encryption devices [16] and advanced  
30 computations [17–19]. As one of the simplest ASI structures, the Kagome ASI has attracted  
31 extensive attention [2,7–9,12,13,20–29], because it is highly frustrated and contains a rich phase  
32 diagram. Theoretical investigations suggest four thermal phases with reducing  
33 temperatures [13,21,28,30]: a high temperature paramagnetic state (PM phase); a spin liquid with  
34 correlated spins satisfying the Kagome ice rule (‘two in/one out’ or ‘two out/one in’) but with  
35 neither charge nor spin ordering (SL1 phase); a long-range ordered charge crystal in a disordered  
36 spin liquid state (SL2 phase); as well as a spin crystal state in which the spins have long-range  
37 ordering (LRO phase) as the lowest temperature state. These thermal phases were used to  
38 understand the temperature dependent magnetotransport results in honeycomb structures of  
39 nanowire networks (which is sometimes also called Kagome ASI, because the moments of the  
40 nanowires satisfy the Kagome ice rule), where the LRO spin crystal state may contribute to the  
41 topological Hall signals at the lowest temperatures [30]. However, previous investigations have  
42 shown that direct experimental access of the spin crystal states of a Kagome ASI is  
43 challenging [12,31]. With the exception of the magnetic writing approach [31], no LRO spin  
44 crystal state has been unambiguously visualized in a Kagome ASI consisting of fully disconnected  
45 nanobar magnets, hindering investigations of emergent phenomena and phase transitions from its  
46 low energy manifolds.

47 The collective properties of ASIs are directly associated with their lattice geometries, and are  
48 intimately governed by the competing dipolar interactions between their constituent elements, i.e.,  
49 the single-domain nanobar magnets [16,32–34]. The difficulty of obtaining the LRO state of a  
50 Kagome ASI originates from the extensive degeneracy of the ground state, resulting from the high  
51 frustration of the tri-leg vertices. The ground state of a Kagome ASI exhibits neither charge nor  
52 spin order when only nearest-neighbor interactions are considered [21,35]. Further-neighbor  
53 interactions induce charge and/or spin ordering [13,21,28]. However, these longer-distance  
54 interactions are much weaker than that from the nearest-neighbor. As a result, the properties of  
55 artificial spin ices are mostly dominated by the nearest-neighbor interactions. Recently, utilizing  
56 the micromagnetic nature of connected vertices in the honeycomb structure of nanowire networks,  
57 the LRO spin crystal phase of Kagome ices was realized for the first time, in which notches were  
58 introduced to reduce the vertex degeneracy [36]. More recently, asymmetric bridges were  
59 introduced to break the six-fold symmetry of Kagome ASI’s vertices, leading to a direct real-space  
60 imaging of the phase transitions [37]. However, these strategies are not applicable to Kagome ASIs  
61 consisting of disconnected nanobar magnets. In this letter, we develop a new method to tailor the  
62 vertex degeneracy and the ground states of a Kagome ASI of disconnected magnetic nanobars, and  
63 we directly present the phase transition from the SL1 liquid to LRO crystal state. Unlike the  
64 recently realized connected Kagome ASI structure [36,37], in which the coupling is dominated by  
65 short range exchange interaction, our disconnected ASIs maintain their long range dipolar  
66 interaction, therefore, allowing us to directly evaluate the critical role of the nearest and/or next  
67 nearest neighbor interactions, which is crucial for understanding the phase transitions between the

68 various low energy manifolds of ASIs.

69 In square ASIs, the six vertex configurations satisfying the spin ice-rule are divided into two  
70 types according to energy [1,2,10,11,14,17,18,22,24,29,38–44]. The lowest energy configuration  
71 is two-fold degenerate, leading to the formation of a long-range ordered ground state [1,38–41].  
72 The low degeneracy of 2 in a square ASI is induced by the non-equal interactions between the  
73 nanobar magnets at each vertex. Following this notion, we developed a method to achieve the LRO  
74 spin ice state of a regular Kagome ice by inducing non-equal interactions between the three  
75 nanobar magnets at a vertex, which reduces the ground state degeneracy of a Kagome vertex from  
76 6 to 2. As shown in Fig. 1(a), we increase the length of one of the three nanobar magnets ( $\alpha$ ) in  
77 each vertex, while maintaining the length of the other two magnets ( $\beta$ ) and the lattice constant (see  
78 Fig. S1 of Supplemental Material for the detailed arrangement of  $\alpha$  and  $\beta$  magnets in the  
79 lattice [45]). This breaks the three-fold rotation symmetry of the vertex. The interactions between  
80 the three nanobar magnets at each vertex are no longer equivalent. As shown in Fig. 1(b), the  
81 original six-fold degenerate vertices are divided into two groups of Type K-I and K-II  
82 configurations with different energies. We denote the interaction energy of the frustrated magnet  
83 pair between two  $\beta$  nanomagnets in Type K-I vertices as  $J_1$ , while that between  $\alpha$  and  $\beta$   
84 nanomagnets in Type K-II vertices as  $J_2$  [Fig. 1(b)]. Since each vertex satisfying the Kagome ice  
85 rule contains only one frustrated magnet pair,  $J_1$  and  $J_2$  also represent the energies of the Type K-  
86 I and K-II vertices, respectively. Because the endpoints of the lengthened  $\alpha$  nanomagnet are closer  
87 to the vertex center,  $J_1$  is lower than  $J_2$ , resulting in the two-fold degenerate Type K-I vertices to  
88 be the ground state configuration. As shown in Fig. 1(a), when all the vertices are satisfied to be  
89 in the Type K-I ground state configuration, a LRO spin crystal emerges. We can further tailor the  
90 energy difference between Type K-I and K-II vertices by varying the length ( $L_\alpha$ ) of the  $\alpha$

91 nanomagnet. Figure 1(c) presents the vertex energy evolution of Type K-I and K-II vertices as a  
92 function of  $L_\alpha$ , calculated from micromagnetic simulation using Mumax3 [46]. It shows that  $J_2$   
93 increases with  $L_\alpha$  while  $J_1$  remains constant. Thus, the energy difference  $J_2 - J_1$  between Type K-I  
94 and K-II vertices increases with  $L_\alpha$ . Therefore, varying the length of the  $\alpha$  nanomagnets allows us  
95 to regulate the effective temperature, similar to the effect of tuning the vertex notch in the  
96 connected honeycomb structures [36]. This enables us to tailor the phases in a fully disconnected  
97 Kagome ASI.

98 To experimentally validate this approach, we fabricated Kagome ASIs with Permalloy  
99 nanomagnets and with a series of  $L_\alpha$  values (220 nm, 270 nm, 320 nm, 370 nm, 420 nm and 440  
100 nm) for the  $\alpha$  nanomagnets (see SEM images in Fig. S2 of Supplemental Material [45]). The length  
101 ( $L_\beta$ ) of the  $\beta$  magnets is fixed at 220 nm, and the lattice constant is  $a = 640$  nm for all samples [Fig.  
102 1(a)]. The width and thickness of all the nanomagnets are 80 nm and 15 nm, respectively. Details  
103 of the sample fabrication process and parameters can be found in Supplemental Material [45]. A  
104 demagnetization procedure (see Supplemental Material [45]) lasting 72 hours was performed to  
105 obtain the low-energy states [11,47]. Figures 1(d) and 1(e) show the SEM images of the samples  
106 with  $L_\alpha = 220$  nm and 420 nm, respectively. The corresponding magnetic force microscopy (MFM)  
107 images are displayed in Figs. 1(f) and 1(g), respectively, which allow us to determine the magnetic  
108 moment (or spin) configurations [see arrows in Figs. 1(f) and 1(g)]. The results show that all the  
109 vertices in all measured samples satisfy the Kagome ice rule ('two in/one out' or 'two out/one in').  
110 This indicates our demagnetization procedure successfully brought the samples into the low energy  
111 ice rule manifold. The conventional Kagome ASI with  $L_\alpha = L_\beta$  [Fig. 1(d)] exhibits disordered spin  
112 and charge configurations [Fig. 1(f)], consistent with the frozen spin liquid SL1 state. In contrast,  
113 the modified Kagome ASI with  $L_\alpha > L_\beta$  (Fig. 1e) exhibits perfect spin and charge ordering [Fig.

114 1(g)], leading to a successful realization of the LRO spin crystal ground state.

115 The transition from SL1 phase to LRO spin crystal phase is demonstrated by the MFM images  
116 of samples with increasing  $L_a$  values in Figs 2(a-f). The corresponding maps of vertex distributions  
117 in Figs. 2(g-l) shows that domains of crystallization become larger with  $L_a$ . For the sample with  $L_a$   
118 = 220 nm (the conventional Kagome ASI), the Type K-I and K-II vertices are degenerate, leading  
119 to a disordered magnetic state with vertex populations of 33.75% and 66.25% for Type K-I and K-  
120 II vertices, respectively. This is consistent with the configurational (or random) populations of 1/3  
121 and 2/3 for Type K-I and K-II vertices, as expected for a spin liquid, and proves that our  
122 demagnetization procedure effectively brought the system into an effective thermal equilibrium  
123 state. When  $L_a > 220$  nm, the Type K-II vertices become an excited state. With increasing  $L_a$ , the  
124 energy difference between Type K-I and Type K-II vertices increases [Fig. 1(c)] and thus the  
125 population of Type K-I vertices gradually increases. As shown in Figs. 2(g-l), ordered domains of  
126 Type K-I vertices emerge and grow with increasing  $L_a$ . For the sample with large  $L_a$ , such as 440  
127 nm [Fig. 2(l)], 93% of the vertices are in the Type K-I ground state, and domain walls comprised  
128 of excited Type K-II vertices (red) are clearly visible. These results match nicely with our Monte  
129 Carlo simulations (Figs. S3 and S4 of Supplemental Material [45]) conducted using a thermal  
130 annealing protocol and considering only nearest-neighbor interactions (see Supplemental  
131 Material [45]). This suggests that the transition from SL1 phase to LRO spin crystal state is  
132 dominated by the nearest-neighbor interactions.

133 We can further elucidate the phase transition from liquid to crystal using magnetic structure  
134 factors [11, 47]. Maps of the magnetic spin structure factor are shown in Figs. 2(m-r). For the  
135 conventional Kagome ASI ( $L_a = 220$  nm), the magnetic spin structure factor map shows structured

136 diffusive pattern [Fig. 2(m)], consistent with previous results of SL1 phase [13,36]. With gradually  
137 increasing  $L_\alpha$ , we can clearly observe the emergence and enhancement of Bragg peaks [Figs. 2(o-  
138 r)]. These Bragg peaks show split-peak structures whose number reduces with increasing  $L_\alpha$ , as  
139 shown in the insets of Figs. 2(o-r). These split Bragg peaks originate from scattering between  
140 domains, as demonstrated by the structure factor maps of the artificial domain configurations in  
141 Fig. S5 of Supplemental Material [45]. The number of domains decreases when they merge,  
142 resulting in reduced number of split Bragg peaks. This suggests that the texture of Bragg peaks  
143 could be used as a qualitative parameter to investigate ordering and domain formation in ASIs,  
144 e.g., we could estimate the relative sizes of domains from the number of split Bragg peaks.

145 Previous investigations of conventional Kagome ASIs revealed local ordering of magnetic  
146 charges [22–24]. Figs. 3(a-f) are maps of magnetic charge configurations corresponding to the spin  
147 and/or vertex configurations in Figs. 2(g-l). We can see that the charge domains of the two-fold  
148 degenerate phases (green and yellow) grow with increasing  $L_\alpha$ . When comparing the charge  
149 distributions to the vertex or spin distributions, we can see that their profiles match very well with  
150 each other for the samples with large  $L_\alpha$  values [Figs. 2(i-l) and 3(c-f)], i.e., the charge ordering is  
151 embedded in the spin ordering for spin crystal state. However, the charge and vertex distributions  
152 deviate as the number of Type K-II vertices increases [Figs. 2(g) and 3(a)]. A similar behavior is  
153 observed when we compare the magnetic spin structure factor maps with the magnetic charge  
154 structure factor maps. For large  $L_\alpha$ , both spin [Fig. 2(o-r)] and charge [Fig. 3(i-l)] structure factor  
155 maps exhibit clear Bragg peaks with the same peak splitting [insets of Fig. 2(o-r) and Fig. 3(i-l)].  
156 However, for small  $L_\alpha$  values, e.g., for the conventional Kagome ASI with  $L_\alpha = L_\beta = 220$  nm, the  
157 spin structure factor map is structured but diffusive [Fig. 2(m) and 2(n)], while in contrast, the

158 charge structure factor maps display clear Bragg peaks, although they are broad [Fig. 3(g) and  
159 3(h)]. This indicates the emergence of charge ordering in the spin liquid state. As mentioned earlier,  
160 charge ordering should not appear when there are only nearest-neighbor interactions and the six-  
161 fold symmetry is not broken [21,35]. Our Monte Carlo simulations with only nearest-neighbor  
162 interactions show that there are no Bragg peaks in the magnetic charge structure factor map for  
163 conventional Kagome ASI (Fig. S6g of Supplemental Material [45]). This suggests that further-  
164 neighbor interactions beyond nearest-neighbors play a role in the liquid state. On the other hand,  
165 the consistency between experiment [Fig. 3(i-l)] and simulation (Fig. S6(i-l) of Supplemental  
166 Material [45]) for samples with large  $L_a$  unambiguously suggests the LRO spin crystal phase can  
167 be established with only nearest-neighbor interactions.

168 We have shown that the local interactions of an ASI have significant impact on their collective  
169 behavior and ultimately affect the properties of the entire system. The length of selective nanobar  
170 magnets can be used as a convenient knob to tune the local coupling strength, which enables us to  
171 access various low-energy manifolds and phase transitions in a fully disconnected ASI. We proved  
172 that although the next-nearest neighbor interaction plays a notable role through the magnetic  
173 charge structure factor maps, the crystallization of the Kagome ice structure with reduced vertex  
174 degeneracies is dominated only by the nearest neighbor interaction. This method could be used to  
175 manipulate the frustration in ASIs for attaining even more exotic ground state phases (see Figs. S7  
176 of Supplemental Material [45]). It would also allow us to realize new magnetic configurations that  
177 are hard to access with magnetization method, e.g., to design novel ASIs with composite ground  
178 states in which different low energy states co-exist in the same sample, as illustrated by the hybrid  
179 ground state of both liquid and crystal in Fig. S8 of Supplemental Material [45]. This would allow

180 us to investigate phase transitions between these new types of low energy states. Furthermore,  
181 kinetics becomes topologically protected in the SL2 phase and in the LRO state [48], and our  
182 structural modifications can be employed to fine-tune different kinetic regimes in thermal  
183 realizations or under field inversions. In addition, this method is also applicable to other types of  
184 artificial spin ices, leading to new opportunities to explore more exotic collective phenomena, such  
185 as novel phases and phase transitions. It could also be combined with the other structure  
186 modification strategies, such as lattice transformation [49,50], to design new ASIs with  
187 controllable degeneracies. This method, maintaining the dipolar coupling between disconnected  
188 nanomagnets, could result in different spin dynamic properties and magnonic applications [51,52]  
189 than those in connected systems [36,37]. Moreover, our approach avoids complex analysis of the  
190 micromagnetic structures such as the domains/domain-walls in the vertices of connected systems,  
191 and thus offers a simpler Kagome model which can connect with a variety of systems outside of  
192 magnetism, such as metal organic frameworks [53] and mechanical metamaterials [54, 55]. Last  
193 but not least, the observed Bragg peaks' splitting induced by spin scattering between domains  
194 could lead to an alternative way to investigate domain/domain wall formation in ASIs.

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196

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### Figure captions

198 **Fig.1.** Tunable Kagome artificial spin ice. (a) Design of tunable Kagome artificial spin ice with  
199 extended length  $L_\alpha$  of  $\alpha$  magnets and fixed length  $L_\beta$  of  $\beta$  nanomagnets. (b) Six low energy  
200 vertex configurations satisfying the Kagome ice rule are separated into two groups based  
201 on energies.  $J_1$  and  $J_2$  are the coupling strengths of frustrated magnet pairs. (c) Evolution of  
202 the vertex energies as a function of  $L_\alpha$ . (d) and (e) SEM images of Kagome artificial spin  
203 ices with  $L_\alpha = 220$  nm (d) and  $L_\alpha = 420$  nm (e), respectively. Scale bar, 500 nm. (f) and (g)  
204 MFM images corresponding to (d) and (e), respectively. Arrows indicate spin  
205 configurations. Scale bar, 500 nm.

206 **Fig.2.** Transition from liquid to crystal. (a)-(f) MFM images for samples with various  $L_\alpha$  values,  
207 respectively. Scale bar, 2  $\mu\text{m}$ . (g)-(l) spin configurations and vertex distributions extracted  
208 from (a)-(f). Type K-I and K-II vertices are shown in blue and red, respectively. (m)-(r)  
209 corresponding maps of magnetic structure factors calculated respectively from the spin  
210 configurations in (g)-(l). Insets of (o)-(r) show expanded views of the split Bragg peaks.

211 **Fig.3.** Magnetic charge ordering. (a)-(f) magnetic charge distributions corresponding to MFM  
212 images in Figs. 2(a)-2(f) respectively. Green and yellow denote two phases of magnetic  
213 charge ordering. Scale bar, 2  $\mu\text{m}$ . (g)-(l) maps of magnetic charge structure factors  
214 associated with (a)-(f). Insets show expanded views of the split Bragg peaks.

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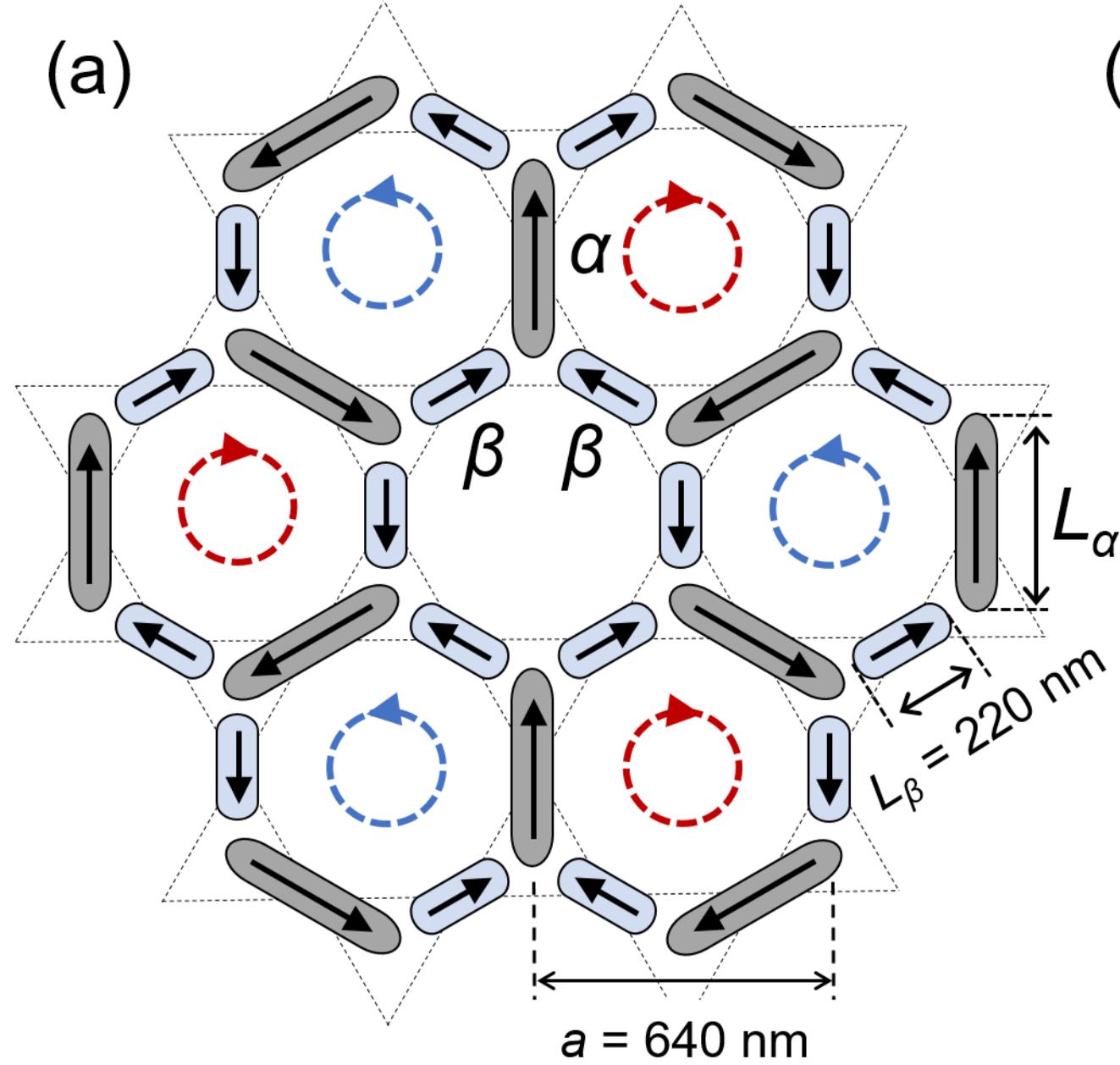
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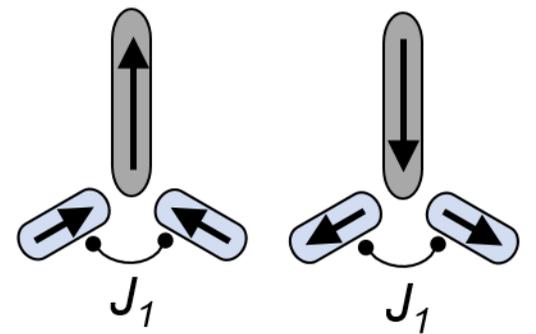
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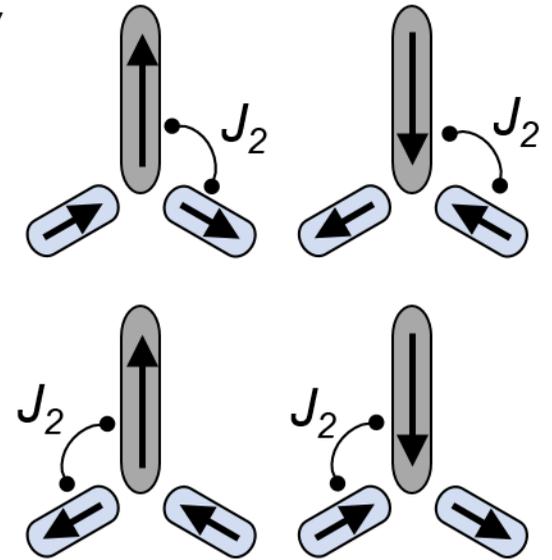
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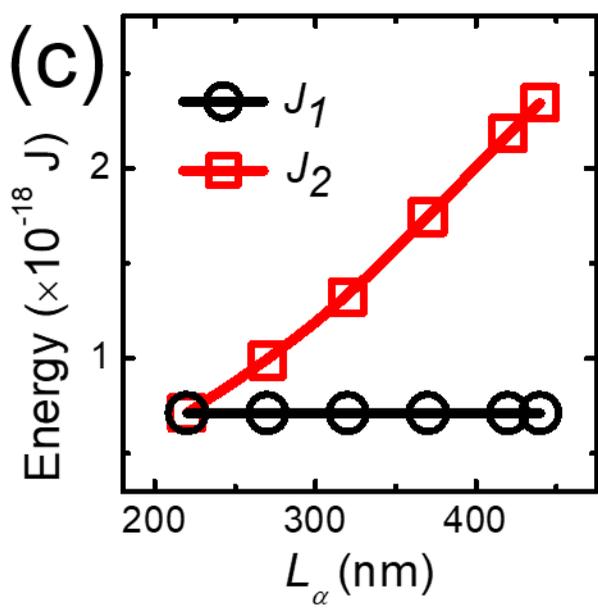
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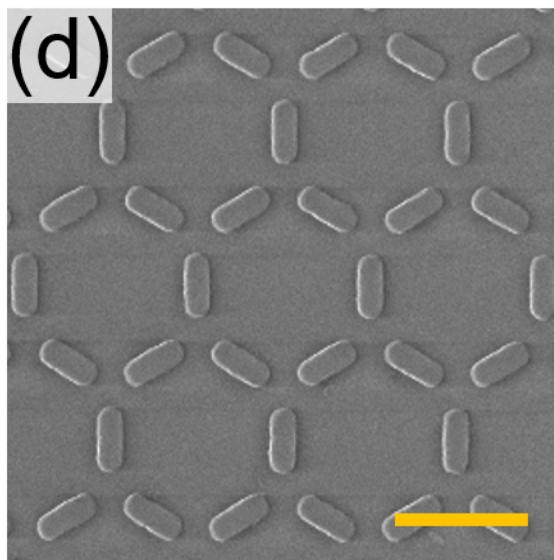
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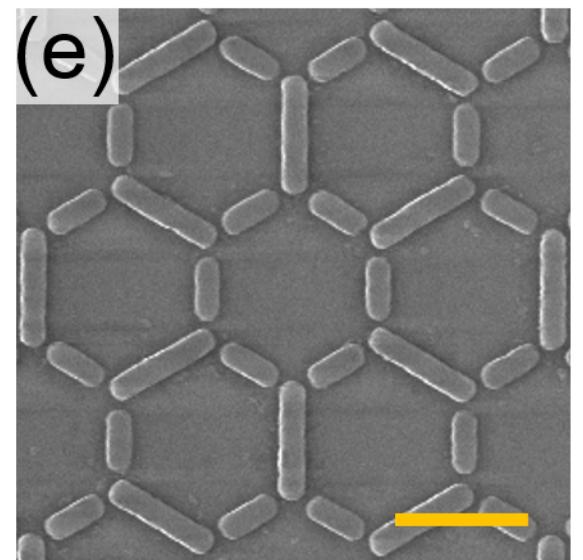
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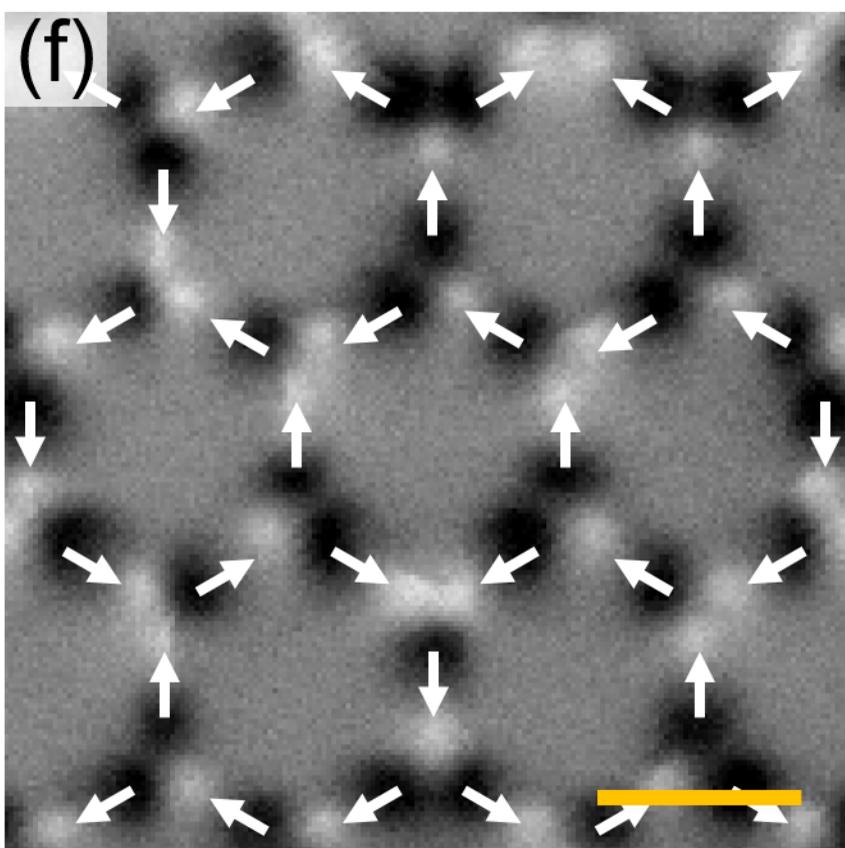
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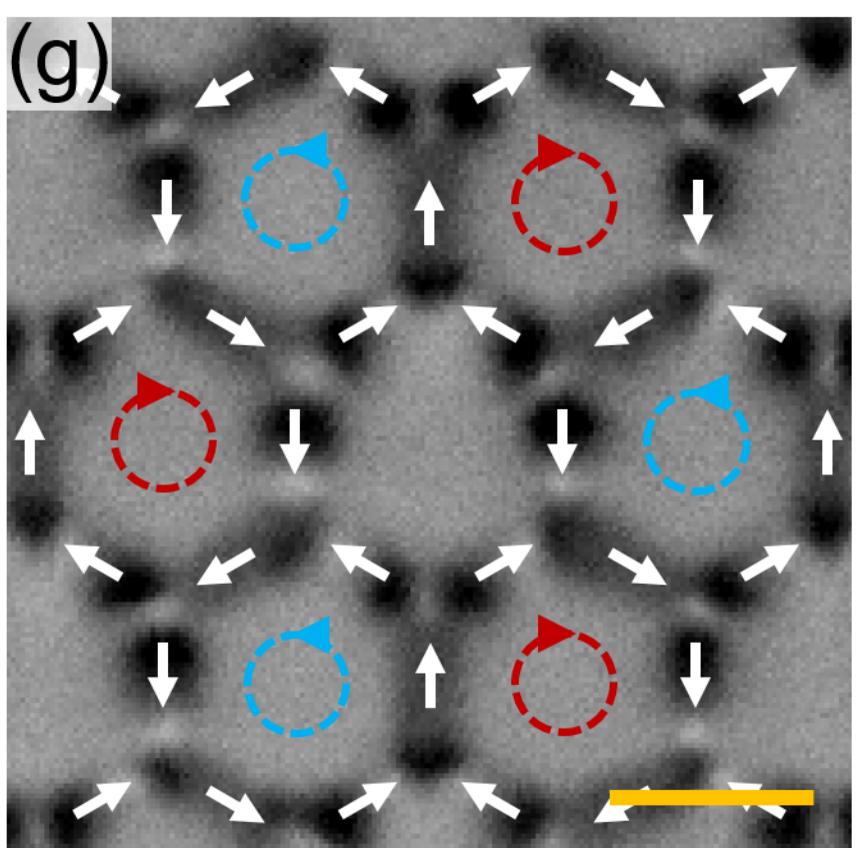
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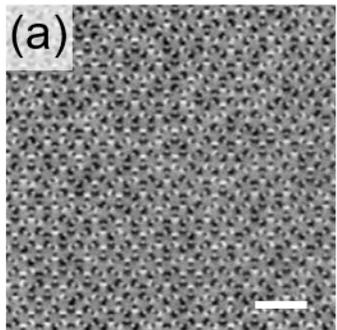
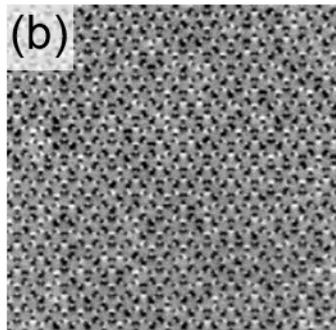
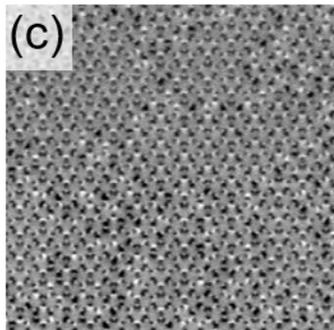
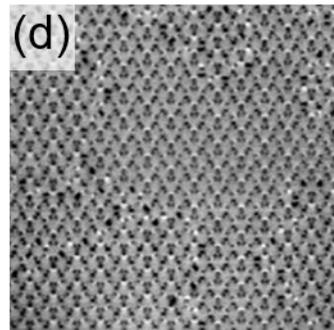
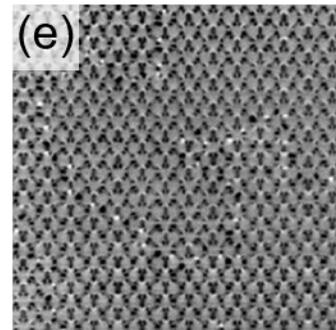
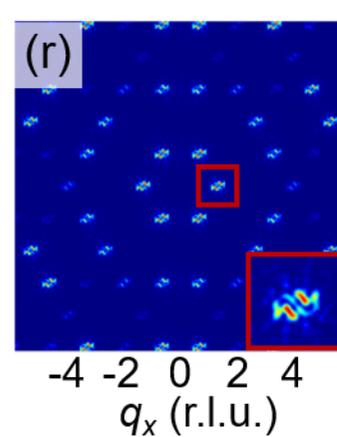
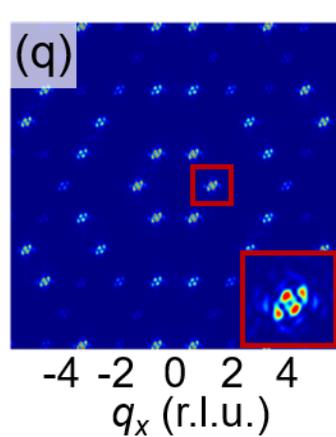
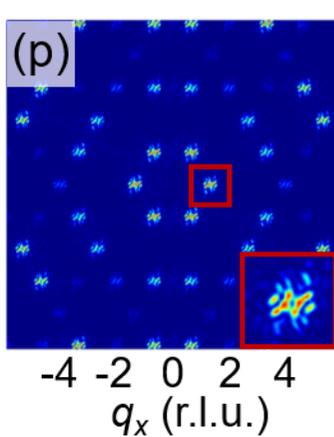
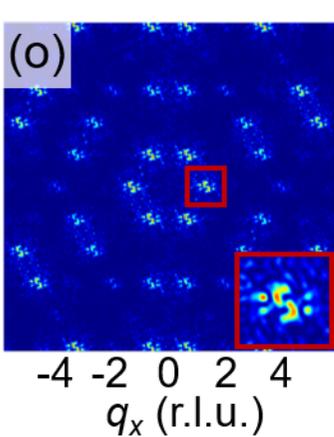
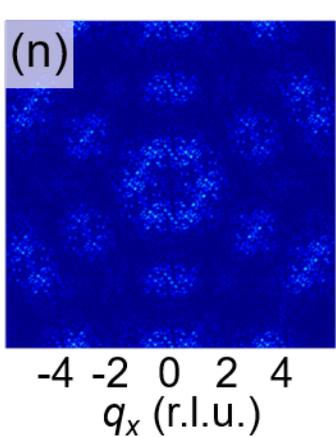
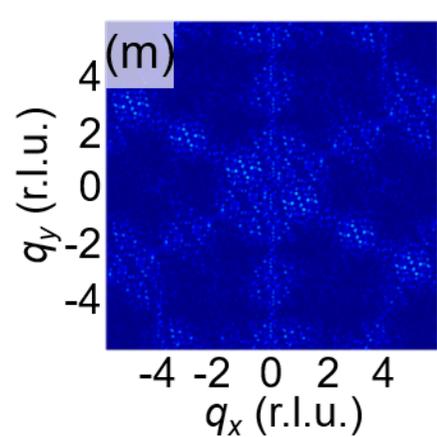
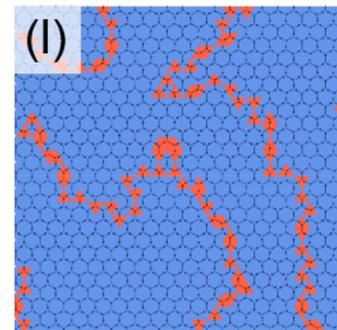
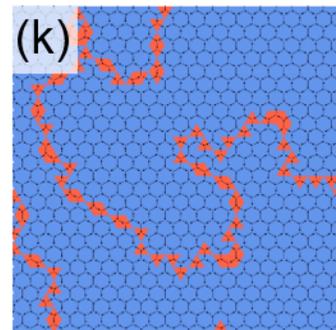
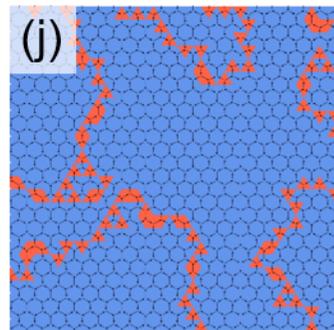
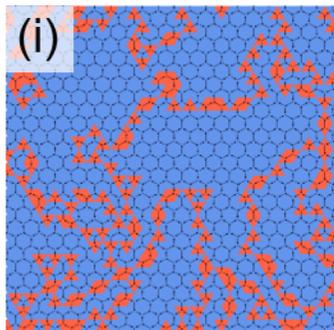
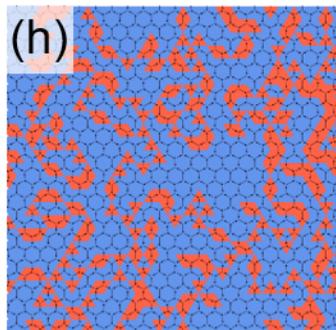
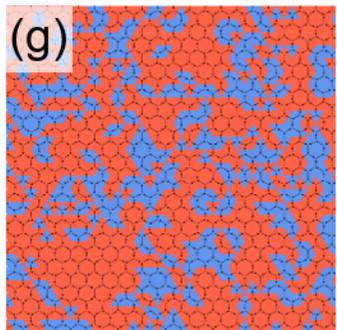
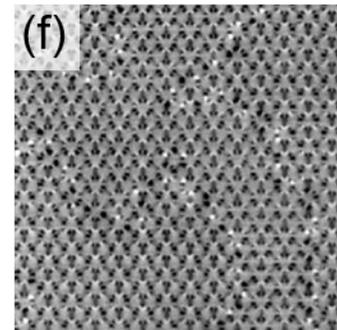


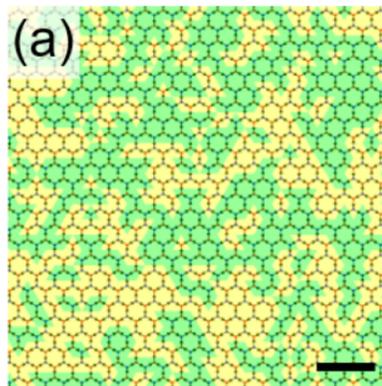
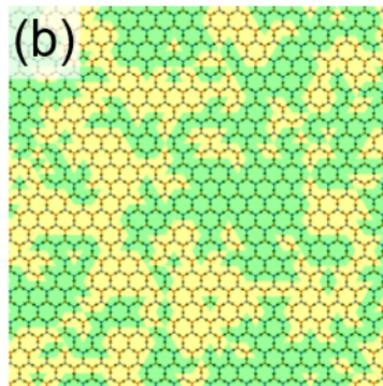
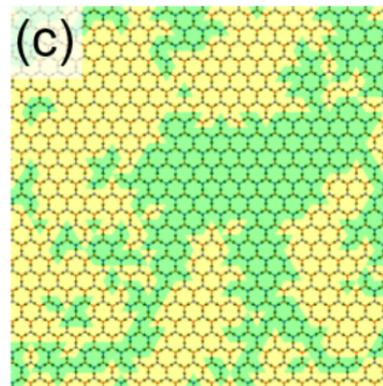
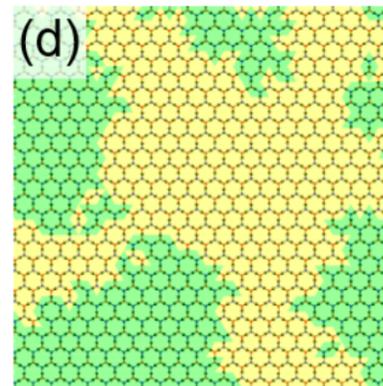
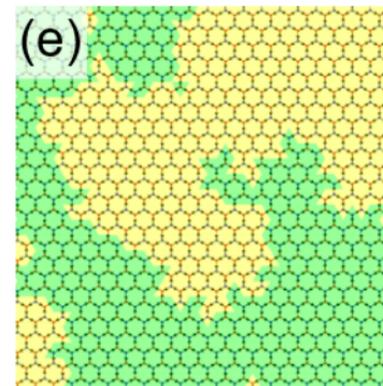
(f)



(g)



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