Accessing low-energy magnetic microstates in symmetry-broken isolated square artificial spin ice vertices with magnetic field

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In artificial spin ice systems, an interplay of defects and dipolar interactions is expected to play important roles in stabilizing different collective magnetic states. In this work, we investigated the magnetization reversal of individual defective square artificial spin ice vertices where defects break four-fold rotational symmetry of the system. By varying the angle between the applied field and the geometrical axis of the vertices, we observe a change in energy landscape of the system resulting into the stabilization of collective low-energy magnetic states. We also observe that by changing the angle, it is possible to access different vertex configurations. Micromagnetic simulations are performed for varying angle as well as external field, the results of which are consistent with the experimental data.

I. INTRODUCTION

Artificial spin-ice (ASI) systems are lithographically patterned arrangements of interacting magnetic nanostructures that were introduced for investigating the effects of geometric frustration in a controlled manner [1–3]. These are 2D arrays of nanomagnets with strong shape anisotropy that mimics natural 3D spin ice materials [4–6]. Strong shape anisotropy makes the magnetic moments in these lithographically defined systems analogous to Ising spins. An intriguing aspect in ASI, which is attracting much interest, is the study of controlled defects and tunability of microstates in these systems. The impact of defects or disorder via modifying the lattice constant, shape of nanomagnets in such ASI systems remains a field of intense research [7–13]. Investigating individual vertices allows detailed understanding of magnetization reversal [15, 16]. As a natural extension of such ASI systems, creation of novel complex geometries such as sakthi [17, 18], tetris [19], etc. made it possible to study frustration in different kind of geometries. As such structures can be relatively easily created, therefore, a detailed understanding of the interplay of defects and dipolar interactions may be helpful to create newer designer materials. The square lattice of ASI can be considered as composed of two orthogonal sublattices of identical nanomagnets owing to their easy axes aligned along the [10] and [01] directions. Thus, square ASI geometry has four-fold symmetry and hence a deformation in the form of misalignment of the easy axis of one of the vertex nanoislands breaks the rotational symmetry of the system. As observed from simulations earlier [14], this may lead to a rich and nontrivial magnetization reversal due to external magnetic field. Due to the broken symmetry engineered by introducing misalignment, the system becomes energetically inequivalent under rotation in an applied magnetic field. Thus, it may be possible to access different energy landscapes by rotating the sample with respect to an external magnetic field. The magnetization reversal and effective anisotropy can be modified, allowing access to different microstates.

In order to achieve a detailed understand of the interplay of defect and dipolar interaction, in this work, we investigated the magnetization reversal for individual defective vertex where the defect is in the form of misalignment which is artificially created. Varying the angle of the applied field with respect to the geometrical axes of different sublattices offers a route to the generation of predictable microstates. In order to extract quantitative information about the energetics of the defective system under rotation and understand the magnetization reversal behavior in depth, we also performed micromagnetic simulation. Fig.1(a) shows the schematics of different types of microstates in square geometry in increasing energy (E_{type-I} <...... < E_{type-IV}). Under the dumbbell model, each individual macrospin can be represented by a positive and negative magnetic charge. For the magnetic configurations shown in type-I and type-II (Fig.1(a)) there are two positive and two negative charges at the vertex and therefore, the vertex is chargeless, whereas the higher-energy configuration type-III and type-IV is associated with a net charge at the vertex. Fig.1(b) shows the schematics of different edge loops viz. onion (i), horse-shoe (ii) and microvortex (iii) state. Calculations based on the macrospin model show that the energy hierarchy of these states follows E_{microvortex} < E_{horse–shoe} < E_{onion} [11, 14].
II. METHODS

The sample used for this study is an ASI vertex with closed edges. A defect in the form of misaligned island at the vertex is created which breaks the rotational symmetry. The details are shown in the schematic diagram in Fig. 1(c). The angle between the long axis of the misaligned island and the [10] axis is chosen as 20°. For this defective square ASI, stadium shaped nanoislands of dimensions 300 nm × 100 nm were patterned on SiO_2/Si substrate using electron-beam lithography. Thin film of Ti(5 nm)/Ni_{80}Fe_{20}(25 nm)/Al(5 nm) was deposited using e-beam deposition system. Finally, the lift-off processing was used to define the magnetic nanoislands. Ti layer of 5 nm is used for better adhesion of the Ni_{80}Fe_{20} film on the substrate whereas Al is used as top layer to prevent oxidation of Ni_{80}Fe_{20}. The center-to-center distance between each nanoisland is 450 nm. For MFM imaging in presence of magnetic field, a commercial variable field module (Asylum Research) equipped with a rotatable permanent magnet was used. The external magnetic field was applied in-plane along [01] direction. The angle dependent studies were performed by controllably rotating the sample counter clockwise (CCW) in the fixed field as shown in Fig. 1(c). We employed a low moment tip (magnetic moment 3 × 10^{-14} emu) to avoid any tip induced changes in the magnetic state of the nanostructures. The distance between the sample and the MFM tip was optimized in the range of 80-100 nm to avoid superposition of topography on the magnetic domain images. For the measurements, we initially fully magnetize the system along [01] and then sweep the magnetic field in the opposite direction ([01]) to image the magnetic structure in mid-transition magnetization states. The field sweep rate of 0.004 T/min was used through out the measurements. The measurements were performed at discrete fields (see below). The single domain character of the nanoislands was confirmed from MFM images where each island appears as a dumbbell of bright and dark contrasts corresponding to the head and tail of a macrospin characterizing these nanoisland. The experiments were inspired by our previously reported micromagnetic simulation work [14].

Micromagnetic simulations were performed using finite difference based Object Oriented Micro Magnetic Framework (OOMMF) [20] software at T = 0 K. For the simulations, the exact structures of the fabricated samples as obtained from scanning electron microscopy (SEM) images were used. The nanoislands have strong shape anisotropy ($K_{sh} \sim 7 \times 10^4$ Jm^{-3}) and hence we neglect the magnetocrystalline anisotropy of Ni_{80}Fe_{20}. The exchange stiffness was taken to be $A = 13 \text{pJ/m}$ [21], and the saturation magnetization was taken to be $M_s = 8.6 \times 10^5$ A/m,

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**Figure 1:** (a) Illustration of 16 possible spin states at a vertex of two-dimensional square artificial spin ice system. Degenerate states are grouped into four different types of increasingly higher energies. The different colored circles at the vertex for type-III states represent the different net magnetic charges at the vertex. (b) Schematic of (i) onion state (ii) horse-shoe state, and (iii) microvortex states [11, 14]. (c)-(e) Schematics of rotation of sample with fixed magnetic field direction.
Figure 2: (a) SEM image of the fabricated structure at rotation angle $\theta \sim 0^\circ$. MFM images taken at (b) saturation, (c) near remanence and (d)-(h) intermediate fields after corresponding magnetic switchings. The arrows in the magnetic images show the orientation of magnetizations in the nanomagnets at different fields and are for guide to the eyes. (i) Simulated hysteresis loop showing 3 switchings. Micromagnetic state (j) at remanence and (k)-(n) at intermediate field.

(as estimated from our SQUID measurements). The magnetization reversal was studied while sweeping the field between $\pm 200$ mT at $T = 0$ K. To understand the switching behavior of the interacting nanomagnets and compare with our experimental data, we calculated the exact micromagnetic states of the system at every 2 mT during the reversal.

III. RESULTS

First we discuss the magnetic field dependent MFM data for the sample with field configuration as shown in the schematics in Fig.1(c). Fig.2(a) shows the SEM image of the fabricated structure consisting of 12 nanoislands with a misaligned island at the vertex (marked from 1 to 12). The field is applied along [0 $\bar{1}$] direction. The system was initialized with the magnetization saturated parallel to an external field of 250 mT as shown in Fig.2(a). The MFM data at the saturated state is shown in Fig.2(b) where the arrows indicate the direction of magnetization of the nanoislands. During reversal of the external field, the static equilibrium configuration at remanence evolves into a two-in/two-out magnetic state at the vertex with four onion states in the loops as shown in Fig.2(c). The MFM data for $-10 < \mu_0 H < -60$ mT do not show any significant change in the magnetic state of the nanoisland indicating no magnetization switching has taken place in the field range. As shown in Fig.2(d), the first magnetic reversal is observed at $\mu_0 H = -62.5$ mT at which two diagonal islands (marked as 1 and 6) switch by forming horse-shoe states in the respective loops which has lower energy than the onion state. The switchings described above indicates an indirect coupling between the nanoislands 1 and 6. Similar indirect couplings of nanoisland and simultaneous switchings at a certain field were also observed in earlier reported micromagnetic simulations [11]. Next switching is observed at $\mu_0 H = -66$ mT where the nanoisland marked as 3 switches. Due to this switching, the two-in/two-out state at the vertex changes to one-in/three-out state (see Fig.2(e)). According to the dumbbell model, proposed by Castelnovo et al., [6] a magnetic dipole can be assumed to represent magnetic charges of $+Q_m$ and $-Q_m$. Thus, this reversal leads to an ice-rule-violating defect with a formation of net charge $\sum Q = -2Q_m$ at the vertex...
Figure 3: (a) SEM image of the fabricated structure for rotation angle $\theta \sim 45^\circ$. MFM images at (b) saturation, (c) near remanence and (d)-(h) intermediate fields after corresponding magnetic switchings. (i) Simulated hysteresis loop showing multiple switchings. Micromagnetic state (j) at remanence and (k)-(l) at intermediate field.

Micromagnetic simulations of the structure show that the remanent state of the vertex is of two-in/two-out type-II spin ice state as shown in Fig.2(i). The hysteresis in magnetization observed from simulations performed at every 2 mT field shows 3 sharp jumps (Fig.2(i)). The first jump corresponds to the switching of the islands 5 and 6 as shown in Fig.2(k). Note here that due to the simulations carried out at every 2 mT, an uncertainty of 2 mT in the switching fields is to be considered. At $\mu_0 H = -64$ mT as shown in Fig.2(k). The third jump corresponds to switching of island 2 at $\mu_0 H = -90$ mT (not shown). As the reverse field is further increased, the magnetization of island 9 for which the external field is along hard axis, changes its orientation thereby turning the vertex in to type-II state at $\mu_0 H = -112$ mT (see Fig.2(m) and (n)). We note here that during reversal process, we observe bending of average local magnetization at the edges of magnetic nanoislands which may have effect in magnetic interaction between the nanoislands. The observed switching behavior is reproducible while sweeping the field in the opposite di-
rection. Thus, the micromagnetic simulations reproduces the field-dependent different magnetic states of the vertex as observed in our MFM images (see Table 1). However, the exact switching patterns differ in experiments and simulations which we speculate as due to changes in the local magnetization such as bending due to tip-induced effect. This may result in to a different magnetostatic coupling between the experimental nanoislands than observed in simulations. Additional effects due to minor difference in the angle $\theta$ between experiment and simulations can not be ruled out.

In order to investigate how the applied field direction influences the interisland magnetostatic coupling and resulting magnetization reversal of the system, we changed $\theta$. The sample is rotated CCW by $\theta \sim 45^\circ$ as shown in the schematics in Fig. 3(d). Profound changes are observed in reversal mechanism resulting in new microstates which were not observed earlier. At this configuration, the defective island’s anisotropy axis and external applied field are oriented at an angle $\alpha_1 \sim 25^\circ$ (see Fig. 3(a)). Thus, a large component of external field is directed along the easy axis of the misaligned nanoisland. The other nanoislands are oriented at $\alpha_2 \sim 45^\circ$ with respect to the field direction (see Fig. 3). The system was saturated parallel to an external field of 250 mT. The MFM image recorded at this field is shown in Fig. 3(b). As the field is reversed, the magnetization of each islands relaxes along their easy axis with a type-II configuration at the remanence. A near remanence image at $-10$ mT is shown in Fig. 3(c). For the field less than $\mu_0 H = -63$ mT, no switching was observed. As the field approaches $\mu_0 H = -64$ mT, the nanoisland 2 switches as shown in Fig. 3(d). This converts an onion-type loop to a horse-shoe type loop. Investigating magnetic state at every $\sim 1$mT, the next switching is observed at $\mu_0 H = -67$ mT, at which islands 3, 8 and 9 switch simultaneously (see Fig. 3(e)), thereby converting two onion and one horse-shoe into three microvortex loops. With these switchings, the chain of nanoislands consisting of islands 2, 3, 9 and 8 all have their major component of magnetization oriented along the external field direction. Together with other islands, viz., 5, 10, 4 and 11, they form a larger loop which is energetically favorable. The microvortex state consists of sublattices with opposite magnetizations only. Thus, the three microvortices lead to reduction in the no. of head-to-head (tail-to-tail) configurations and hence are energetically more favourable. Interestingly, these switchings also lead to the creation of a type-I state at the vertex. Considering that the square ASI vertex of type-I state of lowest possible energy was never observed during magnetization reversal for the case of $\theta = 0^\circ$, this is a remarkable observation. This demonstrates a possible way of achieving the ground state of an ASI-vertex. As the field is further increased to $\mu_0 H = -72$ mT, six islands (1, 4, 5, 6, 10 and 11) switch together and eventually type-II state is generated again at the vertex as illustrated in Fig. 3(f). It is clearly evident that the rotation of the system with respect to the field angle increases the component of applied field along the easy axis of different nanoislands resulting in multiple switching and coupling of different nanoislands. The next switching occurs at $\mu_0 H = -76.5$ mT and $-82.5$ mT, at which island 12 (see Fig. 3(g)) and island 7 (see Fig. 3(h)) switch respectively. The corresponding simulated hysteresis loop, as shown in Fig. 3(i), depcits the gradual rotation of magnetization of the nanoislands followed by six sharp jumps. We show only 3 micro-magnetic states here (Fig. 3(j)–(l)). Fig. 3(j) illustrates the micromagnetic state at remanence with four onion states with a type-II state at the vertex. As the field is increased in the reverse direction, again in this case, we observe the edges of the stadium shaped nanomagnets exhibiting curling of the local magnetization directions (shown in Fig. 3(k)). Fig. 3(l) shows micromagnetic state captured just after the first switching of island 3 which takes place at $\mu_0 H = -62$ mT. The curl or bending of local magnetization at the edges of some nanoislands are clearly observed here. In addition to these details of local magnetization within the individual nanoislands, importantly, we observe that the magnetization switching of the island 3 converts the vertex of type-II to a type-I. Here again, we find that the exact sequence of the switching of the nanoislands does not match with our experiments however the simulation results are consistent with the experimental observation of the evolution of the vertex state as a function of applied field. Thus, our experimental and simulation results for this relative field configuration suggests an increased stability of the system at this configuration resulting in to the creation of the lower energy type-I state at the vertex. The initial and final reversal of magnetization is observed at a greater field value ($\mu_0 H = -64$ mT and $\mu_0 H = -82.5$ mT) for this orientation of $\theta \sim 45^\circ$. The data show that the microstates of the system are strongly influenced by the geometric arrangement of the islands with respect to the applied field.

We next discuss our results for rotation angle $\theta \sim 90^\circ$. The same measurement field protocol was followed as discussed in the above 2 cases. At this configuration, the external field makes an angle $\alpha \sim 20^\circ$ with the easy axis of misaligned nanoisland, while other nanoislands are oriented along or perpendicular to the applied field (see Fig 4(a)). An MFM image of the system at saturation ($\mu_0 H = 250$ mT) is shown in Fig 4(b). In this case also, we observe that the vertex at the remanence is of type-II. This is also observed at $\mu_0 H = -10$ mT (Fig 4(c)) where no switching has taken place so far. An interesting switching behavior is observed at $\mu_0 H = -62$ mT. While scanning at this field, we observe that the local stray field of the MFM tip induces a switching of magnetization of island 9 thereby creating a three-in/one-out type-III state at the vertex (see Fig 4(d)). This tip induced switching during scanning is observed as white-white patches at the ends of island 9 which
otherwise exhibits black-white patches as can be seen in Fig.4(c). Similar color contrasts at both ends indicate that the switching occurred as the scanning progressed over the single-domain nanoisland. Interestingly, as the scanning progresses further upwards, another tip induced switching takes place for the island 10. This switching converts the vertex from type-III to type-II which is chargeless. The two tip-induced switchings are indicated by yellow arrows in Fig.4(d). Thus, these abrupt switchings convert a type-III state (charged state) to type-II (chargeless state) during the same scan (see Fig.4(e)). It is important to note here that the same magnetic tip as well as scan height were used for the cases, viz., for $\theta = 0^\circ$ or $45^\circ$ as discussed above. But no such tip-induced switching was observed for those two cases. This reversal changes all the onion states to lower energy horse-shoe states as shown in Fig.4(e). The second switch is observed at $\mu_0 H = -66 \text{mT}$ at which islands 11 and 12 switch simultaneously as shown in Fig.4(f). At $\mu_0 H = -69 \text{mT}$, island 7 switches thereby forming three onion states and one horse-shoe state (see Fig.4(g)). The fourth switch corresponds to switching of island 8 at $\mu_0 H = -72 \text{mT}$ which restores onion states again in all the loops (see Fig.4(h)).

Further insights into the interesting switching behavior is gained by performing micromagnetic simulations for this field configuration given by $\theta = 90^\circ$. The simulated hysteresis loop shows that the magnetization reversal of the system in this case takes place via four sharp jumps indicative of four switchings (see Fig.4(i)). A closer look at the micromagnetic states near these switching fields shows that magnetization of island 10 orient in a zig zag way at $-60 \text{mT}$, most likely due to a strong competition between the Zeeman and the anisotropy energy at this configuration. It is plausible to assume that the energy barrier is reduced as a result of for the metastable char-

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**Figure 4:** (a) SEM image of the fabricated structure at rotation angle $\theta \sim 90^\circ$. MFM images taken at (b) saturation, (c) near remanence, (d)-(h) intermediate fields after corresponding magnetic switchings. Two tip-induced switchings are indicated by dotted arrows in (d). (i) Simulated hysteresis loop showing four switchings. Corresponding micromagnetic state at (j) saturation and (k)-(o) at intermediate field.
Table I: Table summarizes the magnetic states at intermediate fields corresponding to different rotation angles. (*) refers to vertex state observed for all other fields.

| Angle of rotation \(\theta\) | Magnetic states at intermediate field |
|---------------------------|-------------------------------------|
| Experimental              | Simulation                          |
| 0°                        | type-II* and type-III (66 mT–69.5 mT) | type-II* and type-III (82 mT–100 mT) |
| 30°                       | -                                   | type-II*                              |
| 45°                       | type-I (67 mT–72 mT) and type-II*    | type-I (62 mT–66 mT) and type-II*     |
| 60°                       | -                                   | type-I (62 mT–66 mT) and type-II*     |
| 90°                       | type-I*                             | type-I*                               |
| 135°                      | -                                   | type-II* and type-II (74 mT–142 mT)   |
| 160°                      | -                                   | type-II*, type-III (100 mT–200 mT) and vortex states (18 mT to -122 mT) |
| 180°                      | -                                   | type-II*, type-III (100 mT–200 mT) and vortex states (18 mT to -122 mT) |

Next, we discuss the energetics of the system as it evolves with the variation of \(\theta\). Fig. 5(a) shows the total energy \(E_{tot}\) of the system as a function of \(\theta\) for six different values of the external field, viz., \(\mu_0 H = 0\) mT \((E_0)\), \(\mu_0 H = 50\) mT \((E_{50})\), \(\mu_0 H = 70\) mT \((E_{70})\), \(\mu_0 H = 100\) mT \((E_{100})\), \(\mu_0 H = 120\) mT \((E_{120})\) and \(\mu_0 H = 200\) mT \((E_{sat})\) respectively. \(E_0\) is the energy of the system at remanence which expectedly, remains almost constant for all \(\theta\). At higher fields altered dipolar interactions among the nanomagnets leads to variations in the net energy as \(\theta\) changes. This behavior of energy remains qualitatively similar for all fields which are less than the switching fields. Note that the switching fields for all the configuration as observed from simulations are in the range of 60 mT - 85 mT (see Table I). It is evident from Fig. 5(a) that as the field increases beyond the switching field, the shape of the energy profile manifests itself in a very clear form. For clarity, only three energy profiles in this higher field range, viz., \(E_{100}, E_{120}\) and \(E_{sat}\) are plotted. At these high fields, the energy becomes negative for all values of \(\theta\). As shown in Fig. 5(a), the profiles at these fields show that the rotation of the system leads to an asymmetric energy landscape with a global minima referring to the lowest energy point on the entire energy landscape at \(\theta \approx 45°\) and a local minima at \(\theta \approx 135°\).

The energy profiles for fields within the switching field regime indicate multiple features reflecting the changes in energy due to the switchings of nanomagnets in this field regime. A typical example of such an energy profile in this field regime is shown by plotting \(E_{70}(\theta)\).

Such variations in energy can be understood by invoking Stoner-Wohlfarth model for the dipolar coupled system for which we consider effective anisotropy \((K_{eff})\), effective magnetic moment \((m_{eff})\) etc. of the system consisting of 12 similar nanoislands. In that case, the total energy \(E_{tot}\) of the system in presence of field is given as sum of anisotropy energy \((E_a)\), dipolar interaction energy \((E_d)\), and Zeeman energy \((E_z)\) of the system, i.e.,

\[
E_{tot} = E_a + E_d + E_z
\]

where, \(E_z = \mu_0 m_{eff} H cos\beta\), \(H\) the external field and \(\beta\) is the angle between effective magnetic moment \(m_{eff}\) and applied field \(H\). The first term in eqn. 1 remains constant as \(\theta\) is varied. On the other hand, the second term, \(E_d\) shows clear dependence on \(\theta\) in presence of field as depicted in Fig. 5(b) which shows the variations of \(E_d\) for \(\mu_0 H = 100\) mT, 120 mT and 200 mT, respectively. It is evident that corresponding dipolar energies also show global minima at \(\theta \approx 45°\) at these fields. The third term in eqn. (1) depends on \(\beta\) which dominates at higher fields and in turn depends on \(\theta\) as discussed below.

To understand the energy profile at magnetic fields higher than the switching fields, we evaluate \(\beta\) for varying \(\theta\) for our system. From the magnetization of individual nanoislands in the coupled system for a given \(\theta\) and \(\mu_0 H\) as obtained from the simulations, we determine the corresponding direction of \(m_{eff}(\theta)\) for different fields applied in the direction of [01]. Fig. 5(c) shows the orientation of \(m_{eff}\) for four different \(\theta\) values calculated.
for $\mu_0 H = -100$ mT. The results for the three fields are tabulated in Table II.

We find that for $\theta = 45^\circ$, $m_{\text{eff}}$ orients along the direction of the applied field (i.e., $\beta = 0^\circ$) which is the condition for the minimum energy state. Manifested of this is observed as a global minimum in the profiles of $E_d$ and in turn $E_{\text{tot}}$. In the micromagnetic behavior, interestingly, we observe the vertex to acquire the ground state (type-I) configuration at this angle of rotation which most likely is related to the global minimum of the energy. We note further that the micro-vortex type edge loops which have the lowest energy are also observed for this angle. Thus, these results clearly demonstrate that the angle between external field and the geometric axis of an ASI vertex can be considered as an important parameter to explore the energetics of the system.

| $\theta$ | $\beta(-100 \text{ mT})$ | $\beta(-120 \text{ mT})$ | $\beta(-200 \text{ mT})$ |
|----------|------------------|------------------|------------------|
| $0^\circ$ | $-10^\circ$ | $-5^\circ$ | $10^\circ$ |
| $30^\circ$ | $9^\circ$ | $8^\circ$ | $6^\circ$ |
| $45^\circ$ | $0^\circ$ | $0^\circ$ | $0^\circ$ |
| $60^\circ$ | $-9^\circ$ | $0^\circ$ | $-4^\circ$ |
| $90^\circ$ | $-25^\circ$ | $-17^\circ$ | $10^\circ$ |
| $135^\circ$ | $0^\circ$ | $-2^\circ$ | $0^\circ$ |
| $160^\circ$ | $-14^\circ$ | $-11^\circ$ | $-2^\circ$ |
| $180^\circ$ | $-16^\circ$ | $-12^\circ$ | $0^\circ$ |

Table II: Table summarizes values of angle $\beta$ at applied field $\mu_0 H = -100$ mT, $-120$ mT and $-200$ mT respectively, for different rotation angles.

IV. CONCLUSIONS

Our results show that the ASI vertex structure with broken rotational symmetry presents an interesting system for experimental exploration. The broken symmetry engineered by introducing misalignment leads to an energetically inequivalent system under rotation in an applied magnetic field and allows easy access to different energy
landscapes. By rotating the samples in an applied field, we are able to stabilize different vertex configurations, from magnetically chargeless type-I to charged type-III states. Our results suggest the role of intricate interplay of defect and dipolar interactions in predictably stabilizing different vertex states which may be of interest to explore electronic transport behavior in such clearly defined vertices.

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