

Skyrmion lattice annihilation by point defects in the multiferroic Cu_2OSeO_3

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First-principles-based effective Hamiltonian simulations are used to reveal the hidden connection between various topological defects, namely point defects and skyrmions, in copper oxide selenite (Cu_2OSeO_3). Using this approach, we show that (i) Cu_2OSeO_3 hosts different topological defects with a nonzero local topological charge in various regions of the magnetic-field–temperature phase diagram (H, T), (ii) point defects exert external pressure on skyrmions leading to their gradual annihilation as temperature increases, and (iii) this annihilation of skyrmions is a result of the melting of the lattice into a glassylike state of spins that connects with the paramagnetic phase.

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Introduction. Topological defects arise in a wide variety of condensed matter systems, and due to their nature related to the topological properties, interest in the field of spintronics has surged. The potential of devices based on topological defects, especially skyrmions, has gained the attention of the condensed matter community in the last decade. A direct result of topological defects' topological stability is their robustness in the presence of continuous mechanical deformations and defects, leading to extensive studies of the topological solitons in magnetic materials, including vortices, hedgehogs, and skyrmions. Skyrmions were first proposed by Skyrme in 1962 in the field of particle physics to describe a solution of the nonlinear field in dense nuclear matter [1]. After that, the concept of skyrmions has been found in various condensed matter systems, such as liquid crystals [2], quantum Hall systems [3], and magnetic materials. Magnetic skyrmions are the primary example of topological spin textures. They are characterized by a topological invariant called the skyrmion number \mathcal{Q}_{sk} , which describes the local configuration of how the vector field of magnetic dipoles whirls in the plane. The magnetic skyrmions were expected to exist within magnetic materials with strong Dzyaloshinskii-Moriya interaction (DMI) [4,5]. They were first discovered in the metallic noncentrosymmetric chiral cubic MnSi [6]. This started the hunt for skyrmions in other B20 crystals such as FeGe [7] and $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ [8], and they were found in both bulk material and thin films [9]. These spin textures, called skyrmions, are nontrivial topological configurations that can be described as circular spin textures with an up spin on the edge of the circle and a down spin in the center. In between, there is a smooth transition with topological features. Furthermore, magnetic skyrmions were found in multiferroic copper oxide selenite (Cu_2OSeO_3) with the cubic space group $P2_13$ as for B20 magnets [10]. Skyrmions in bulk Cu_2OSeO_3 and bulk B20 magnets exist in a narrow pocket of

the magnetic-field–temperature phase diagram; however, isolated skyrmions are hard to stabilize, and thus in Cu_2OSeO_3 the magnetic skyrmions are arranged in a hexagonal two-dimensional (2D) lattice called the skyrmion lattice (SkL) phase.

There are several methods to characterize topological defects; in the case of 2D solitons such as vortices and skyrmions, the skyrmion number is defined as [11]

$$\mathcal{Q}_{sk} = \frac{1}{4\pi} \int_{\mathbb{R}^2} \mathbf{m} \cdot \left(\frac{\partial \mathbf{m}}{\partial x} \times \frac{\partial \mathbf{m}}{\partial y} \right) dx dy, \quad \mathbf{m} = \frac{\mathbf{M}}{|\mathbf{M}|}, \quad (1)$$

where \mathbf{m} is the normalized local magnetization.

This Research Letter aims to quantify the topological properties of different solitons in Cu_2OSeO_3 from first principles. We define the different significant topological quantities and calculate the charge of every soliton on the phase diagram of Cu_2OSeO_3 (skyrmion lattice phase, vortex lattice, and helical state). We also seek to isolate topological entities such as hedgehogs and antihedgehogs (point defect in three dimensions) and study their interaction with skyrmions. We numerically access the temperature evolution of the density n_{\pm} of topological point defects (sum of the density of hedgehogs and antihedgehogs) and the spin configurations at different temperatures and magnetic fields.

Methods. We use first-principles density functional theory (DFT) calculations with the Vienna *ab initio* simulation package (VASP) [12] to parametrize our effective Hamiltonian model. The pseudopotentials adopt the projector augmented-wave method, with the generalized gradient approximation [13] used as the exchange-correlation functional, and the U correction was added for the copper ions to account for the strong correlation properties of $3d$ electrons [14,15]. Monte Carlo simulations were performed to study the behavior at finite temperatures, which relies on the Metropolis algorithm [16,17] inside an $84 \times 84 \times 84$ cubic supercell with periodic boundary conditions and contains $\sim 10^7$ spins. We used 5×10^5 sweeps to equilibrate the system and then 5×10^5 sweeps to get the various statistical averages. The calculations begin at a high temperature from a random

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configuration, and the temperature is then decreased in small steps to get well-converged results. We note that the effective Hamiltonian employed to simulate the properties of

Cu_2OSeO_3 [18] yields the same characteristic phase diagram reported in previous experimental studies [10,19]. The energy of the magnetic effective Hamiltonian is written as

$$E_{\text{total}}(\{\mathbf{S}\}) = \sum_{i<j} [J_w^{\text{FM}} \mathbf{S}_i \cdot \mathbf{S}_j + J_s^{\text{AFM}} \mathbf{S}_i \cdot \mathbf{S}_j + J_s^{\text{FM}} \mathbf{S}_i \cdot \mathbf{S}_j + J_w^{\text{AFM}} \mathbf{S}_i \cdot \mathbf{S}_j + J_O^{\text{AFM}} \mathbf{S}_i \cdot \mathbf{S}_j] + \sum_{i<j} [\mathbf{D}_w^{\text{FM}} \cdot \mathbf{S}_i \times \mathbf{S}_j + \mathbf{D}_s^{\text{AFM}} \cdot \mathbf{S}_i \times \mathbf{S}_j + \mathbf{D}_s^{\text{FM}} \cdot \mathbf{S}_i \times \mathbf{S}_j + \mathbf{D}_w^{\text{AFM}} \cdot \mathbf{S}_i \times \mathbf{S}_j + \mathbf{D}_O^{\text{AFM}} \cdot \mathbf{S}_i \times \mathbf{S}_j] + \sum_i g_s \frac{\mu_B}{\hbar} \mathbf{S}_i \cdot \mathbf{B}_{\text{ext}}, \quad (2)$$

where J_s (J_w) are the different ferromagnetic and antiferromagnetic strong (weak) Heisenberg interactions in our system, with \mathbf{D} being the equivalent Dzyaloshinskii-Moriya interaction (DMI) vector, and J_O is the only superexchange interaction (long-range coupling mediated by the bridging oxo ligand) with its corresponding DMI vector \mathbf{D}_O . The values of the different interactions are the same as in our previous study [18].

Topological quantities. Following the approach by Berg and Lüscher [20] (see also Ref. [21]), we define the skyrmion number in the case of lattice systems. For a plane of four spins, the topological charge is given by the sum over the ensemble of oriented triangles Δ_{ijk} . We calculate first the local charge q_{Δ} of the triangle Δ_{ijk} with respect to the chosen orientation. The skyrmion number can be extracted from the local charge by summing on the whole space (in the case of 2D solitons as skyrmions and vortices, we sum over the 2D surface), and the lattice-based formula for the skyrmion number is (see Supplemental Material [22] for details)

$$Q_{sk} = \frac{1}{4\pi} \sum_{\Delta=(ijk)} q_{\Delta}. \quad (3)$$

In order to quantify 3D point defects (hedgehogs and antihedgehogs) with charge Q_{\pm} , we use the procedure of Motrunich and Vishwanath [23]. The hedgehog (antihedgehog) topological charge Q_+ (Q_-) enclosed by the unit cube containing six squares can be written as follows:

$$Q_{\pm} = \frac{1}{2\pi} \sum_{\square=1}^6 \mathbf{F}_{\square}, \quad (4)$$

where \mathbf{F}_{\square} is the flux of a cube face. This charge is guaranteed to be an integer from the definition and ensures that the net topological charge (the supercell sum of topological charges associated with unit cells) always equals zero in the considered system with periodic boundary conditions (details of the derivation of the charge can be found in the Supplemental Material [22]).

Results. We first analyze the topological skyrmion number for the zero-field case, where we have a helical state at lower temperatures and then a transition to the fluctuation-disordered (FD) phase that exists in the temperature range 37.5–42.25 K [18]. We find, as shown in Fig. 1(a), that the helical state is, as expected, topologically trivial and is characterized with $Q_{sk} = 0$. This, in particular, means that the DMI itself does not create the net topological charge, and it needs the magnetic field to break the symmetry in agreement with the work of Hou *et al.* [24]. However, as we enter the FD

region of temperatures, the strong spin fluctuations make the skyrmion number fluctuate around the trivial state.

We analyze the skyrmion number for (111) planes in an applied magnetic field $\mathbf{B}_{\text{ext}} = 40$ mT along the (111) direction. We find that the skyrmion number stays zero for lower temperatures where the trivial conical phase is stable. It

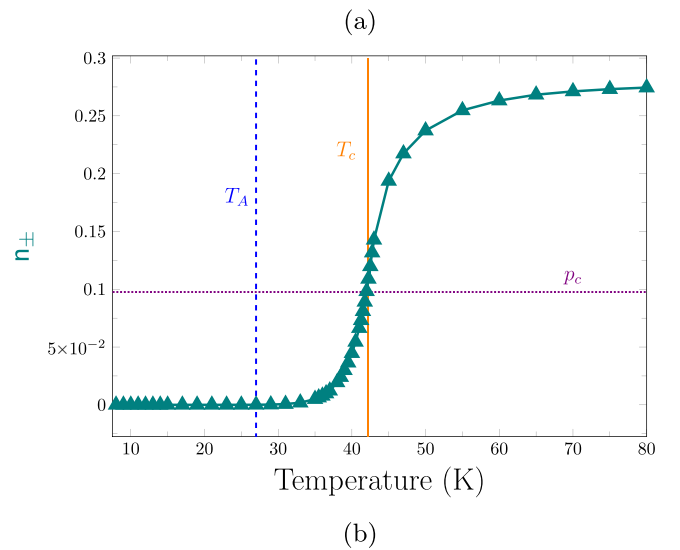
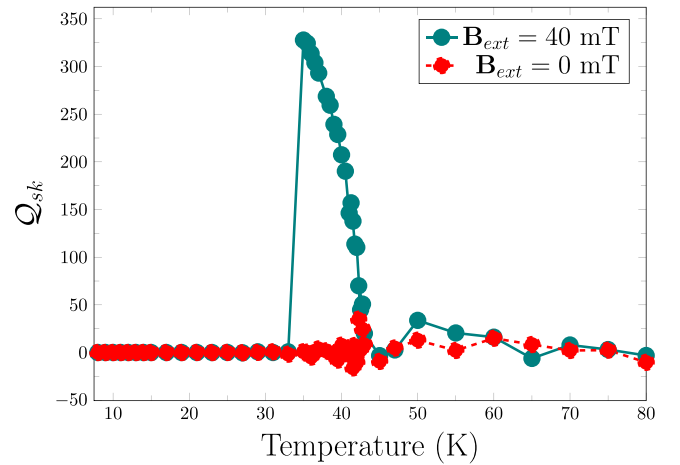


FIG. 1. (a) Dashed line, skyrmion number Q_{sk} along the conical-SkL-FD-paramagnetic transition; solid line, skyrmion number Q_{sk} along the helical-FD-paramagnetic transition. (b) Density of the cells with point defects n_{\pm} as a function of temperature, where p_c is the percolation threshold of a Rubik's cube environment and T_A is the point defects' activation temperature.

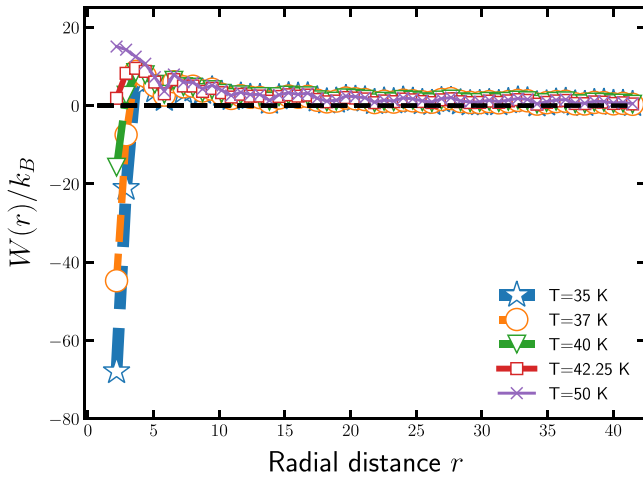
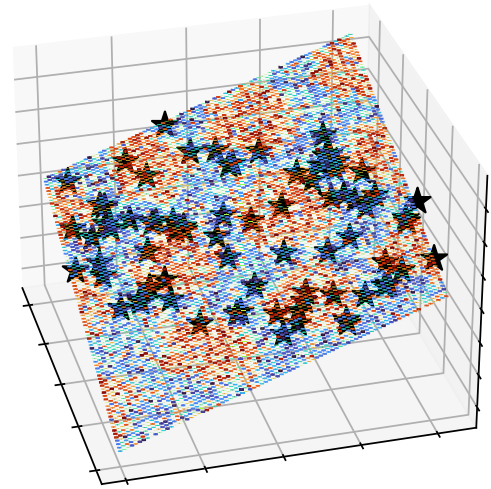


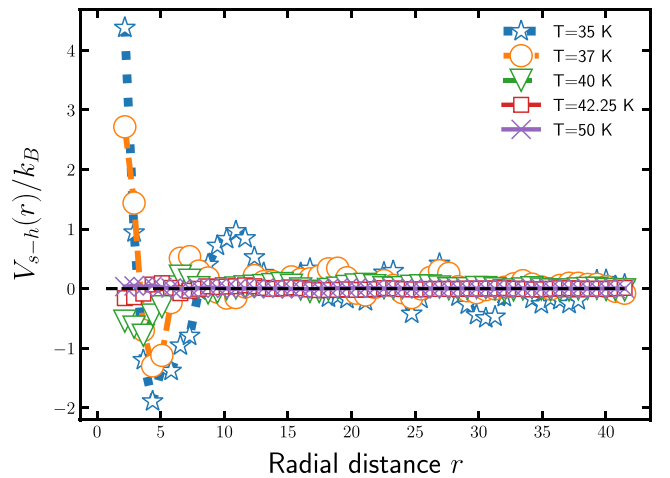
FIG. 2. The pairwise potential between point defects $W(r)$ for various temperatures ($T_c = 42.25$ K), where we observe the changing nature from an attractive potential for $T < T_c$ to a repulsive potential for $T \geq T_c$.

increases steeply to the maximum as we enter the skyrmion lattice phase and then decreases smoothly with a slower rate and small fluctuations due to the FD regime [19]. In the paramagnetic phase ($T > T_c = 42.25$ K), we have fluctuation of the skyrmion number around the value of zero; such behavior was previously found in ultrathin films of Pd/Fe/Ir(111) [25]. Let us now define the density n_{\pm} of topological defects as the ratio of cells containing hedgehogs or antihedgehogs to the supercell volume L^3 with $L = 84$. Figure 1(b) shows the thermal evolution of the density of hedgehog-antihedgehog pairs. The density n_{\pm} is found to decrease smoothly with decreasing temperature until an activation temperature T_A where the density is precisely zero. It is striking to realize that the critical temperature T_c is the inflection temperature of the hedgehogs' density, which also coincides with the site-percolation threshold of a Rubik's cube neighborhood p_c . The site-percolation threshold p_c is that of a regular lattice with a neighborhood extending to the third-nearest neighbors (Rubik's-cube-like) $p_c = 0.0976$ [26]. This threshold can be seen as the critical probability above which we have the appearance of a single cluster of topological defects spanning over the whole supercell ("infinite cluster"). This behavior of point defects was previously found in relaxor ferroelectric $\text{Pb}(\text{Sc}_{0.5}\text{Nb}_{0.5})\text{O}_3$ (PSN) and $\text{Ba}(\text{Zr}_{0.5}\text{Ti}_{0.5})\text{O}_3$ (BZT) systems [27] and in Heisenberg ferromagnets [28,29]. The nonvanishing value of the point defects' density under T_c indicates the coexistence of hedgehog-antihedgehog pairs alongside the skyrmion lattice phase in the case of an applied 40 mT magnetic field. This means that if the density of hedgehog-antihedgehog pairs $n_{\pm} > p_c$, the appearance of a long-range spin configuration is hindered by the infinite cluster of topological point defects.

We compute the radial distribution function $g(r)$, used to extract the mean-force potential between defects $W(r) = -k_B T \ln(g(r))$ as well as using the K -means clustering method to explore the evolution of point defect clusters. Figure 2 shows the evolution of the mean-force potential between defects $W(r)$ as a function of temperature. We observe that



(a)



(b)

FIG. 3. (a) Point defects' (black stars) distribution in a (111) plane in the presence of the skyrmion lattice phase ($\mathbf{B}_{\text{ext}} = 40$ mT and $T = 37$ K) where point defects are clustered in the space between skyrmions (in red). (b) The interaction potential between skyrmions and point defects $V_{s-h}(r)$ for various temperatures ($T_c = 42.25$ K) as calculated from the radial distribution function, where $V_{s-h}(r)$ has a glassylike behavior until $V_{s-h}(r) = 0$ for $T \geq T_c$ due the total vanishing of skyrmions.

point defects repel each other for high temperatures and cannot easily be close to each other lest they overlap the nearest hedgehogs. As the temperature decreases below T_c , the pairwise potential $W(r)$ becomes slightly attractive, and clusters begin to form. The radial distribution function obtained at these temperatures significantly differs from that obtained at $T \geq 50$ K, implying that the point defects' behavior (as individual particles) is also very different. At this density of point defects, the depletion forces can be described by the Asakura and Oosawa model [30,31], which predicts a monotonically attractive potential. In this temperature range, point defects are in the subpercolation regime, where $n_{\pm} < p_c$ and point defects form separated structured clusters as depicted

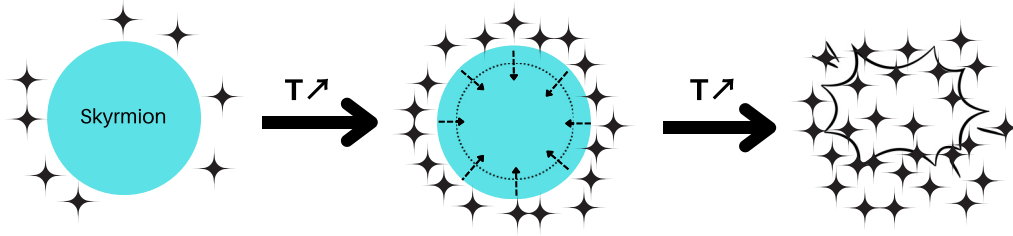


FIG. 4. Schematic representation of the process of skyrmion annihilation by the point defects (black stars) showing the gradual destruction of the skyrmion as a result of the increasing applied pressure of hedgehog-antihedgehog pairs, which is due to the increasing density of point defects in the space between skyrmions as temperatures increase, leading to total destruction of the skyrmion phase as we enter the paramagnetic phase.

in Fig. 3(a). In other words, point defects become trapped in a cage formed by their neighbors, similar to a glassy state of particles. The pairwise potential becomes even more attractive as we reduce temperatures and get closer to the activation temperature T_A .

Interestingly, since the total topological charge is constrained to be zero in systems with periodic boundary conditions, the decrease of n_{\pm} upon cooling can only happen by annihilation among defects of opposite topological charge, i.e., between hedgehogs with charge Q_+ and antihedgehogs with charge Q_- . For the dependence of n_{\pm} on temperature in Fig. 1(b), we find that for $T > T_A$, the density can be very well approximated by

$$n_{\pm} = n_{\pm}^0 + (n_{\pm}^{\infty} - n_{\pm}^0) \exp\left(-\frac{A}{T - T_A}\right). \quad (5)$$

This leads to the Vogel-Fulcher-Tammann-law-like equation for relaxation time τ associated with the annihilation process between hedgehogs and antihedgehogs, $\tau \sim \tau_0 \exp(\frac{A}{T - T_A})$ [32], which is the same equation used to model viscosity near the glass transition as in the glassy state behavior evoked in the previous paragraph.

An important question about the role of topological defects is whether the observed proliferation of point defects, as T increases, is accompanied by an unbinding of defect pairs. For that, we calculated the number of total bonds as a function of temperature and the number of bonds at the minimal distance as a function of temperature. It is thus essential to develop criteria by which a given system is in a bound or unbound hedgehog state. The ratio of the bonds at the minimum distance to the total bonds will give an idea about the binding-unbinding mechanism of hedgehog-antihedgehog pairs occurring in Cu_2OSeO_3 . In our case, we observe a decreasing ratio with increasing temperatures, which means that the pairs tend to unbind, rendering the gas of defects into a plasmalike state of hedgehogs and antihedgehogs. This unbinding mechanism is also shown in Fig. 2 by the pairwise potential $W(r)$ changing in nature from attractive to repulsive as we pass from $T < T_c$ to $T > T_c$. Although the nature of the total pairwise potential stays the same in the absence (at 0 mT) and in the presence (at 40 mT) of the skyrmion lattice phase, being attractive in $T < T_c$ and repulsive in $T > T_c$, the existence of the skyrmions adds an additional component to the pairwise potential, which can be interpreted as the interaction potential between skyrmions and point defects $V_{s-h}(r)$. This potential can be extracted by comparing the pairwise potential

of point defects in the presence and absence of skyrmions.

$$V_{s-h}(r) = W(r)_{\mathbf{B}_{\text{ext}}=40\text{mT}} - W(r)_{\mathbf{B}_{\text{ext}}=0\text{mT}}. \quad (6)$$

Figure 3(b) shows the evolution of the interaction potential between skyrmions and point defects (hedgehogs and antihedgehogs) and exhibits the behavior of the periodic structure (solidlike) melting into a liquidlike potential as the temperature increases, until we have no interaction above T_c due to total annihilation of skyrmions. To understand the effect of point defect clusters on the skyrmion lattice, we analyzed how the point defects are scattered on the (111) plane in the presence of skyrmions. As seen previously, in a magnetic field of 40 mT applied along the (111) direction, a skyrmion lattice phase emerges near T_c . In the presence of skyrmions, the point defects are arranged in clusters in the space between skyrmions, which can be related to V_{s-h} being attractive in these distances (between skyrmions) as shown in Fig. 3(a), leading to applied pressure by point defects on skyrmions. As the temperature increases, the point defects' density grows. The point defects exert increasing pressure as the temperature rises, leading to annihilation of the skyrmion lattice phase. This process of skyrmion annihilation by the point defects is schematically depicted in Fig. 4. We observe that the applied pressure gradually reduces the total skyrmion charge until the complete annihilation of the skyrmion lattice phase. The potential V_{s-h} in Fig. 3(b) shows the behavior of a gradual melting of the skyrmion lattice phase as a result of the pressure of point defects; hence there is a gradual decrease in the skyrmion number Q_{sk} as temperatures increase toward T_c as previously seen in Fig. 1(a). The impact of point defects on the annihilation of skyrmions during the helical-to-skyrmion-lattice or conical-to-skyrmion-lattice transition was previously seen in a study by Birch *et al.* [33], where they demonstrated that Bloch points, a specific type of three-dimensional point defect, facilitated the creation and annihilation of skyrmions under a magnetic field in $(\text{Cu}_{1-x}\text{Zn}_x)_2\text{OSeO}_3$.

In contrast, our study investigates the role of point defects in eliminating the skyrmion lattice phase as the temperature rises and Cu_2OSeO_3 transitions to the paramagnetic phase. In essence, our findings reveal the influence of the proliferation of three-dimensional point defects on the annihilation of ordered phases, particularly the melting and annihilation of the skyrmion lattice phase as a result of the pressure applied by point defects.

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