Supplementary Information for Microscopic Theory of Light-Induced Ultrafast Skyrmion Excitation in Transition Metal Films

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SUPPLEMENTARY NOTE 1: DERIVATION OF THE EFFECTIVE SPIN HAMILTONIAN

We here provide additional details on the derivation of the effective spin Hamiltonian describing the correlated d-electrons in the large U limit. We start from an electronic Hamiltonian of s- and d-orbitals given by $H(t) = H_s(t) + H_{s-d} + H_d$ with

$$H_s(t) = \sum_{i\sigma} \epsilon_{si\sigma}(t)\hat{n}_{si\sigma} - \mathbf{B} \cdot \sum_i \hat{\mathbf{s}}_i + \sum_{\langle ij\rangle\sigma\sigma'} e^{i\theta_{ij}(t)} c_{i\sigma}^\dagger (-t_s\mathbf{1} + \boldsymbol{\alpha}_{sij}\cdot\boldsymbol{\tau})_{\sigma\sigma'} c_{j\sigma'} \tag{1}$$

$$H_d(t) = U_0 \sum_i \hat{n}_{di\uparrow}\hat{n}_{di\downarrow} + \sum_{\langle ij\rangle}\left(\frac{V_0}{2}\hat{n}_{di}\hat{n}_{dj} - J_0\hat{\mathbf{S}}_i\cdot\hat{\mathbf{S}}_j\right) + \sum_{\langle ij\rangle\sigma\sigma'} e^{i\theta_{ij}(t)} d_{i\sigma}^\dagger(-t_d\mathbf{1} + \boldsymbol{\alpha}_{dij}\cdot\boldsymbol{\tau})_{\sigma\sigma'}d_{j\sigma'} - \mathbf{B}\cdot\sum_i\hat{\mathbf{S}}_i \tag{2}$$

$$H_{s-d} = t_{s-d}\sum_{i\sigma}\left(c_{i\sigma}^\dagger d_{i\sigma} + d_{i\sigma}^\dagger c_{i\sigma}\right) + U_{s-d}\sum_{i\sigma\sigma'}\hat{n}_{si\sigma}\hat{n}_{di\sigma'} - J_{s-d}\sum_{i\sigma}\hat{\mathbf{s}}_i\cdot\hat{\mathbf{S}}_i. \tag{3}$$

In Supplementary Equations (1-3), $c_{i\sigma}^\dagger$ ($d_{i\sigma}^\dagger$) creates an s- (d-) electron at site i with spin projection σ, and $\hat{n}_{ai\sigma}$ is the spin density operator for orbital $a \in \{s, d\}$ at site i. The orbital energy is given by $\epsilon_{ai\sigma}$, t_a is the hopping amplitude between nearest-neighbor sites i and j, and $\boldsymbol{\alpha}_{aij}$ accounts for Rashba spin-orbit interactions. The s- and d-electron spin operators are given by $\hat{\mathbf{s}}_i = c_{i\sigma}^\dagger \boldsymbol{\tau}_{\sigma\sigma'}c_{i\sigma'}$ and $\hat{\mathbf{S}}_i = d_{i\sigma}^\dagger\boldsymbol{\tau}_{\sigma\sigma'}d_{i\sigma'}$, where $\boldsymbol{\tau}$ denotes the vector of Pauli matrices and repeated spin indexes are summed over.

In H_d, both a local interaction U_0 as well as nearest-neighbor direct and exchange interactions V_0 and J_0 are included[1]. In the $s - d$ interaction term [Supplementary Equation (3)], t_{s-d} is the hybridization strength, and U_{s-d} and J_{s-d} the direct and exchange interactions. For $\langle\hat{n}_{di}\rangle = 1$ (as assumed in the rest of this work), the direct term just renormalizes the orbital energy $\epsilon_{i\sigma}$. Finally, both s- and d- electron spins interact with an external static magnetic field $\mathbf{B}$ via Zeeman coupling.

Assuming the d-electron system is at half-filling, doubly occupied sites will be penalized by an energy $\sim U$, and the effective Hilbert space can be defined by projecting out the doubly occupied sites. This is achieved by the projection operator $\mathcal{P} = \prod_i \mathcal{P}_i$, where $\mathcal{P}_i = 1 - \hat{n}_{di\uparrow}\hat{n}_{di\downarrow}$. In the following we decompose the Hamiltonian as $H(t) = H_0(t) + H_1$, where H_1 is the interacting part of H_d defined in the main text. For virtual excitations out of the subspace defined by $\mathcal{P}$, where exactly one doubly occupied site is involved, we can write (up to an irrelevant constant[2])

$$H_1 = U_0\sum_i \hat{n}_{di\uparrow}\hat{n}_{di\downarrow} + \frac{V_0}{2}\sum_{\langle ij\rangle}\hat{n}_{di}\hat{n}_{dj} = U\sum_i\hat{n}_{di\uparrow}\hat{n}_{di\downarrow}, \tag{4}$$

where $U = U_0 - V_0$. We note that H_1 only acts in the high energy subspace defined by $\mathcal{Q} = 1 - \mathcal{P}$.

The time-dependent Schrieffer-Wolff transformation is defined as the unitary transformation that at each time t removes the coupling between the low and high energy subspaces[?]. Given a state $|\Psi(t)\rangle$ that evolves under the original Hamiltonian $H(t)$, the unitary transformation $|\tilde{\Psi}(t)\rangle = e^{iS(t)}|\Psi(t)\rangle$ corresponds to a Hamiltonian $\tilde{H} = e^{iS(t)}[H - i\partial_t]e^{-iS(t)}$. Assuming that $S(t)$ can be written as $S(t) = \gamma S_1(t) + \gamma^2 S_2(t) + \mathcal{O}(\gamma^3)$, with γ a small parameter, we find to second order in γ that

$$\tilde{H} = H_0 + H_1 + \gamma\left(i[S_1, H_0] + i[S_1, H_1] - \partial_t S_1\right) + \gamma^2\left([S_2, H_1] - \frac{1}{2}[S_1, i\partial_t S_1 + [S_1, H_1]] - \partial_t S_2\right). \tag{5}$$

To eliminate the leading order off-diagonal term (in $\mathcal{P}$ and $\mathcal{Q}$), we take $S_2 = 0$ and require that $i\gamma[S_1, H_1] - \gamma\partial_t S_1 = -\mathcal{P}H_0\mathcal{Q} - \mathcal{Q}H_0\mathcal{P}$. Since the projection operators $\mathcal{P}$ and $\mathcal{Q}$ act in the subspace of d-electrons, only the d-electron

kinetic term T_d and the coupling Hamiltonian H_{s-d} contribute to S_1. Projecting the expression for $\tilde{H}$ onto the low energy subspace, we have

$$\tilde{H} = \mathcal{P}H_0\mathcal{P} + \frac{i\gamma}{2}\mathcal{P}S_1\mathcal{Q}\mathcal{Q}H_0\mathcal{P} - \frac{i\gamma}{2}\mathcal{P}H_0\mathcal{Q}\mathcal{Q}S_1\mathcal{P}. \tag{6}$$

It now remains to solve the differential equation for S_1. This can be achieved by introducing the retarded and advanced Green's functions

$$G^R(t,t') = -ie^{-i(H_1-i\eta)(t-t')}\theta(t-t') \tag{7a}$$

$$G^A(t,t') = ie^{i(H_1-i\eta)(t'-t)}\theta(t'-t), \tag{7b}$$

in terms of which the projections of the operator S_1 are given by

$$\mathcal{Q}S_1(t)\mathcal{P} = i\int dt'\, G^R(t,t')\mathcal{Q}H_0(t')\mathcal{P} \tag{8a}$$

$$\mathcal{P}S_1(t)\mathcal{Q} = -i\int dt'\, \mathcal{P}H_0(t')\mathcal{Q}G^A(t',t). \tag{8b}$$

Since we work at half-filling the operator H_1 appearing in the exponential of the Green's function always acts after a single excitation has been created, and can therefore by replaced by U in the exponential. The effective Hamiltonian is then

$$\tilde{H} \approx \frac{i}{2}\int dt'\,\theta(t-t')\Big[e^{i(U+i\eta)(t-t')}\mathcal{P}H_0(t')\mathcal{Q}\mathcal{Q}H_0(t)\mathcal{P} - e^{-i(U-i\eta)(t-t')}\mathcal{P}H_0(t)\mathcal{Q}\mathcal{Q}H_0(t')\mathcal{P}\Big] + H_0. \tag{9}$$

where η is a real infinitesimal introduced to regularize the integral as $t' \to -\infty$. In this and following equations containing η, it should be understood the the evaluation of the integral is followed by the limit $\eta \to 0$ from the positive side. The operator products can be evaluated following the procedure detailed in Section below, and leads to a time-dependent spin Hamiltonian

$$\tilde{H}(t) = H_0 + \sum_{\langle ij \rangle}\Big[J_{ij}(t)\hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j + \mathbf{D}_{ij}(t) \cdot (\hat{\mathbf{S}}_i \times \hat{\mathbf{S}}_j) + \hat{\mathbf{S}}_{i\mu}\Gamma_{i\mu,j\nu}(t)\hat{\mathbf{S}}_{j\nu}\Big] + H_{s-d}, \tag{10}$$

with the parameters $J_{ij}(t) = 4t_d^2 I_{ij}(t) - J_x$, $\mathbf{D}_{ij}(t) = 8it_d\boldsymbol{\alpha}_{dij}I_{ij}(t)$ and $\Gamma_{i\mu,j\nu}(t) = (8\alpha_{dij}^\mu\alpha_{dij}^\nu + 4|\boldsymbol{\alpha}_{dij}|^2\delta_{\mu\nu})I_{ij}(t)$. Here we use Greek letters to denote the components of the spin-orbit vector $\boldsymbol{\alpha}_d$. The time-dependent function $I_{ij}(t)$ is given by

$$I(t) = \mathrm{Im}\int dt'\, e^{i(U+i\eta)(t-t')}\cos(\theta_{ij}(t) - \theta_{ij}(t')). \tag{11}$$

Similarly the $s-d$ exchange interaction (obtained from the time-independent part of Supplementary Equation 9) is found to be of the form

$$H_{s-d} = -g\sum_i \hat{\mathbf{s}}_i \cdot \hat{\mathbf{S}}_i, \tag{12}$$

where $g = J_{s-d} - 4t_{s-d}^2/U$.

SUPPLEMENTARY NOTE 2: EVALUATION OF THE OPERATOR PRODUCTS OF THE EFFECTIVE HAMILTONIAN

Given the formal expression for $\tilde{H}$ in Supplementary Equation 9, we can evaluate the operator products following the procedure in Ref.[3]. At half-filling only virtual transitions that involve two sites contribute to the Hamiltonian, and thus the projection operators $\mathcal{Q}$ always give unity and can be removed. The products arising from the kinetic energy of the d-electrons are then of the form

$$T_d(t')T_d(t) = \sum_{\langle ij \rangle} M_{\sigma_1\sigma_2}^{ij}(t')(\delta_{\sigma_2\sigma_3} - d_{j\sigma_3}^\dagger d_{j\sigma_2})M_{\sigma_3\sigma_4}^{ji}(t)d_{i\sigma_1}^\dagger d_{i\sigma_4}, \tag{13}$$

where summation over repeated spin indexes is implied and the projection operators $\mathcal{P}$ have been left out for notational simplicity. We now use that the electronic bilinears can be represented in terms of Pauli matrices as $d_{i\sigma}^\dagger d_{i\sigma'} = (1/2 + \hat{\mathbf{S}}_i \cdot \boldsymbol{\tau})_{\sigma'\sigma}$. Similarly, the products arising from the $s-d$ exchange term are of the form

$$H_{s-d}H_{s-d} = t_{s-d}^2 \sum_i (\delta_{\sigma_1\sigma_2} - c_{i\sigma_2}^\dagger c_{i\sigma_1})d_{i\sigma_1}^\dagger d_{i\sigma_2}, \tag{14}$$

where we can represent the s-electron bilinear by $c_{i\sigma}^\dagger c_{i\sigma'} = (1/2 + \hat{\mathbf{s}}_i \cdot \boldsymbol{\tau})_{\sigma'\sigma}$.

To this order in the Schrieffer-Wolff expansion no other terms can arise, and in particular, terms of the form $\mathcal{P}T_d H_{s-d}\mathcal{P} = 0$ since they contain an odd number of d operators. Therefore, they necessarily create (or destroy) a doubly occupied state, which takes the system out of the low-energy Hilbert space.

With the above representation of the electronic operators the expression for $\tilde{H}$ takes the form of a trace, explicitly given by

$$\tilde{H} \approx H_0 + \sum_{\langle ij \rangle} \text{Tr}\left[\mathbf{M}^{ij}(t')\mathbf{M}^{ji}(t)\left(\frac{1}{2} + \hat{\mathbf{S}}_i \cdot \boldsymbol{\tau}\right)\right] - \sum_{\langle ij \rangle} \text{Tr}\left[\mathbf{M}^{ij}(t')\left(\frac{1}{2} + \mathbf{S}_j \cdot \boldsymbol{\tau}\right)\mathbf{M}^{ji}(t)\left(\frac{1}{2} + \mathbf{S}_i \cdot \boldsymbol{\tau}\right)\right] \tag{15}$$

$$+ t_{s-d}^2 \sum_i \text{Tr}\left[\frac{1}{2} + \hat{\mathbf{S}}_i \cdot \boldsymbol{\tau}\right] - t_{s-d}^2 \sum_i \text{Tr}\left[\left(\frac{1}{2} + \hat{\mathbf{s}}_i \cdot \boldsymbol{\tau}\right)\left(\frac{1}{2} + \hat{\mathbf{S}}_i \cdot \boldsymbol{\tau}\right)\right]$$

We evaluate these traces using a representation of $\mathbf{M}$ in terms of the Pauli matrices, $\mathbf{M} = -t_d\mathbf{1} + \boldsymbol{\alpha}_d \cdot \boldsymbol{\tau}$, together with the property $(\mathbf{a} \cdot \boldsymbol{\tau})(\mathbf{b} \cdot \boldsymbol{\tau}) = (\mathbf{a} \cdot \mathbf{b})\mathbf{1} + i(\mathbf{a} \times \mathbf{b}) \cdot \boldsymbol{\tau}$ and the trace identities $\text{Tr}\,\mathbf{1} = 2$ and $\text{Tr}\,\boldsymbol{\tau} = 0$. The first term in $\tilde{H}$ (for given i and j) can be shown to be independent of the spin vectors for real hoppings $t_d^* = t_d$ and imaginary spin-orbit couplings $\boldsymbol{\alpha}_d^* = -\boldsymbol{\alpha}_d$, which holds when the spin-orbit interaction is of the Rashba form. This term can therefore be neglected. The same argument holds for the terms proportional to the identity matrix in the second, third and fourth term, which can thus be omitted too. The remaining part of the trace is then

$$\text{Tr}\left[\mathbf{M}^{ij}(t')(\mathbf{S}_j \cdot \boldsymbol{\tau})\mathbf{M}^{ji}(t)(\mathbf{S}_i \cdot \boldsymbol{\tau})\right] = e^{i(\theta_{ij}(t')-\theta_{ij}(t))} \times \left[2t_d^2\mathbf{S}_i \cdot \mathbf{S}_j + 2i(t_d\boldsymbol{\alpha}_d^* - t_d\boldsymbol{\alpha}_d) \cdot (\mathbf{S}_i \times \mathbf{S}_j)\right.$$
$$\left. - 2(\boldsymbol{\alpha}_d \times \mathbf{S}_i) \cdot (\boldsymbol{\alpha}_d^* \times \mathbf{S}_j) + 2(\boldsymbol{\alpha}_d \cdot \mathbf{S}_i)(\boldsymbol{\alpha}_d^* \cdot \mathbf{S}_j)\right].$$

for the kinetic part, and

$$\text{Tr}\left[t_{s-d}^2(\mathbf{s}_i \cdot \boldsymbol{\tau})(\mathbf{S}_i \cdot \boldsymbol{\tau})\right] = 2t_{s-d}^2\mathbf{s}_i \cdot \mathbf{S}_i. \tag{16}$$

for the $s-d$ exchange part. Inserting these results in the expression for $\tilde{H}$, and noting that the term with i and j swapped gives an analogous contribution to the kinetic part but with the phase $e^{-i(\theta_{ij}(t')-\theta_{ij}(t))}$, we find the effective spin Hamiltonian of Supplementary Equation (10). The final expression for $I_{ij}(t)$ comes from combining the terms in Supplementary Equation (9) relating to the retarded and advanced Green's functions. Similarly, the extra factor of two in Supplementary Equation 12 for the $s-d$ exchange comes from the Hermitian conjugate of the term considered above.

SUPPLEMENTARY NOTE 3: INEFFICIENCY OF THE INVERSE FARADAY EFFECT FOR ATOMICALLY THIN SAMPLES

A mechanism proposed to underlie the optical excitation of skyrmions is the direct coupling between the material magnetization and the laser electric field via the inverse Faraday effect (IFE)[4,5]. However, a straightforward estimate of the interaction energies involved in the IFE shows that it can not be responsible for skyrmion excitation in quasi two-dimensional systems. The interaction energy for the IFE in a volume a^3 can be written as[6]

$$U = -\frac{i\theta_F c\sqrt{\epsilon_r}\epsilon_0 a^3}{2\omega}\frac{\mathbf{M}(\mathbf{r})}{M_s} \cdot [\mathbf{E}^*(\mathbf{r}) \times \mathbf{E}(\mathbf{r})], \tag{17}$$

where θ_F is the Faraday angle, ϵ_r the relative permittivity of the material, a the lattice parameter, ω the frequency of the laser and M_s the saturation magnetization. The Faraday angle can be written as $\theta_F = \mathcal{V}B$ where $\mathcal{V}$ is the so-called Verdet constant, which is smaller than ~ 100 rad/Tm. Taking a large value $E = 10^9$ V/m, $a = 5$ Å and $\lambda = 800$ nm, giving $\omega \approx 2360$ THz, we find the energy density $U = g\mathbf{S} \cdot \mathbf{e}$ with $g \approx 1.2 \cdot 10^{-7}$ eV, $\mathbf{S} = \mathbf{M}/M_s$ and $\mathbf{e} = (\mathbf{E}^* \times \mathbf{E})/E^2$. Since typical values of spin parameters are on the order of ~ 1 meV, the IFE coupling is at least a factor 10^{-3} smaller than the direct Zeeman term. The reason for this small coupling is that the strength of the IFE is proportional to the propagation length through the system[6], and is therefore strongly suppressed for atomically thin systems. In these systems, the IFE is to a very good approximation negligible.
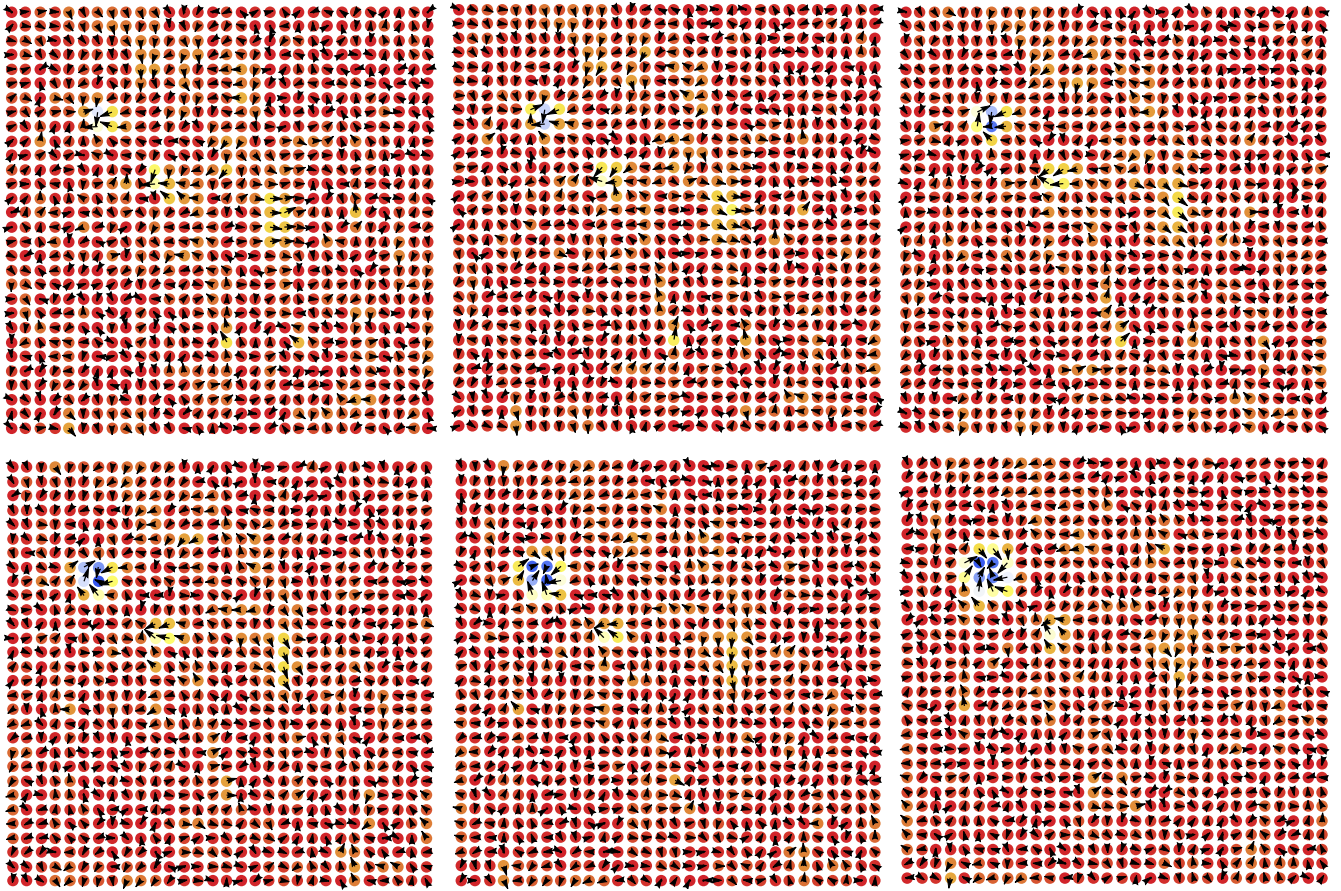
Supplementary Figure 1. Dynamics of skyrmion nucleation. Snapshots of the spin configuration around the time of skyrmion nucleation. The time between the snapshots is 3.3 fs and show the interval between $t = 121.8$ and $t = 138.2$ fs. The skyrmion is created in the upper right panel at approximately $t = 127$ fs. The coloring shows the out-of-plane component of the spin vector ranging from $S_z = -1$ (blue) to $S_z = 1$ (red). The system has 30×30 sites and the spin parameters are $J = 50$ meV, $D = 25$ meV and $B = 10$ meV. The parameters of the s-electrons are $t = 1$ eV and $\alpha = 0.5$ eV, and the $s - d$ exchange coupling is $g = 2.5$ eV. The laser has a field strength $E = 10^9$ V/m, frequency $\hbar\omega = 0.5$ eV, pulse length $\tau = 30$ fs, and spot size of ≈ 14 nm.
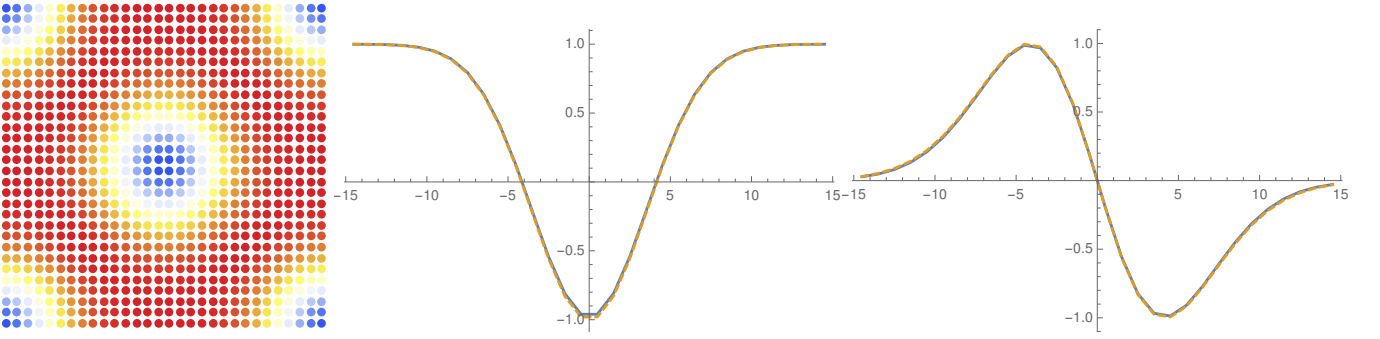
SUPPLEMENTARY NOTE 4: SKYRMION NUCLEATION DYNAMICS

The skyrmion nucleation process displayed in Supplementary Figure 1 of the main text shows snapshots of the spin configuration shortly after the skyrmion has nucleated, and at some later times to illustrate the oscillation of the skyrmion radius. Here, we supplement this figure by showing snapshots of the spin configuration across the skyrmion nucleation process. As can be seen from Supplementary Figure 1, the skyrmion nucleates by a single spin-flip and then expands to a size determined by the magnetic parameters. This is in line with previous work on current induced skyrmion nucleation[7].

SUPPLEMENTARY NOTE 5: PARAMETRIZATION OF RELAXED SKYRMIONS

The parameterization of the relaxed skyrmion state employed in the main text is given $\mathbf{n}(\mathbf{r}) = (-xf(u), -yf(u), 1 - 2e^{-u^2})$, where $f(u) = (2/Ru)(e^{-u^2} - e^{-2u^2})^{1/2}$, $u = r/R$ and r is measured from the center of the system. In Supplementary Figure 2 this parameterization is compared to a relaxed skyrmion configuration obtained from the spin Hamiltonian

$$H_d = \sum_{\langle ij \rangle} \left[J_{ij} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j + \mathbf{D}_{ij} \cdot (\hat{\mathbf{S}}_i \times \hat{\mathbf{S}}_j) \right] - \mathbf{B} \cdot \sum_i \hat{\mathbf{S}}_i \tag{18}$$

Supplementary Figure 2. Parameterization of a relaxed skyrmion. The left panel shows a relaxed skyrmion configuration containing two skyrmions obtained from the ground state of Supplementary Equation 18. The central and right panels show cross-sections of the z- and x-components of the spins along the line $y = 0$ through the skyrmion center. The blue lines give the cross-section of the skyrmion texture in the left panel, while the dashed orange lines show the parameterization $\mathbf{n}(\mathbf{r})$ used in the main text.

by simulated annealing with the Metropolis Monte Carlo algorithm and subsequently relaxed by time-propagation with a small Gilbert damping. The parametrization is seen to give an excellent description of the relaxed skyrmion configuration.

SUPPLEMENTARY NOTE 6: SPIN EQUATION OF MOTION IN THE LARGE $s - d$ EXCHANGE LIMIT

To better understand the influence of itinerant electrons on the dynamics of the spin system, we derive an effective equation of motion for the spins in the limit of large $s - d$ exchange[8–12]. To simplify the algebraic manipulations we adopt a continuum description $\psi_\sigma(\mathbf{r}_i) = c_{i\sigma}/a$ and $\mathbf{n}(\mathbf{r}_i) = \mathbf{n}_i$, where the Lagrangian for the itinerant electrons is given by

$$\mathcal{L} = \int d^2\mathbf{r} \left(\psi_\sigma^\dagger \left[i\hbar\partial_t + \frac{\hbar^2}{2m}\nabla^2 - \frac{i}{2}(\boldsymbol{\alpha}_i \cdot \boldsymbol{\tau})\overset{\leftrightarrow}{\nabla}_i \right]\psi_\sigma + g\psi_\sigma^\dagger(\boldsymbol{\tau} \cdot \mathbf{n})\psi_{\sigma'} \right). \tag{19}$$

Here $\psi_\sigma^\dagger\overset{\leftrightarrow}{\nabla}\psi_\sigma = \psi_\sigma^\dagger(\nabla\psi_\sigma) - (\nabla\psi_\sigma^\dagger)\psi_\sigma$, and the spin-orbit interaction has been written in a general form in order to facilitate the algebraic manipulations. We note that here α_i^a is a matrix with i denoting the spatial and a the spin component, which reduces to the standard Rashba spin-orbit coupling at an interface, $H_{so} = \alpha\hat{\mathbf{e}}_z \cdot (\nabla \times \boldsymbol{\tau})$, by taking $\boldsymbol{\alpha}_x = \alpha\hat{\mathbf{e}}_y$, $\boldsymbol{\alpha}_y = -\alpha\hat{\mathbf{e}}_x$ and $\boldsymbol{\alpha}_z = 0$.

We assume the magnetization is described by a normalized spin texture $\mathbf{n}(\mathbf{r}, t) = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$ where the angles $\theta = \theta(\mathbf{r}, t)$ and $\phi = \phi(\mathbf{r}, t)$ are functions of space and time. Exploiting the gauge invariance of the theory, we perform a local $SU(2)$ transformation $\psi_\sigma(\mathbf{r}, t) \rightarrow U_{\sigma\sigma'}(\mathbf{r}, t)\psi_{\sigma'}(\mathbf{r}, t)$ of the electronic field operators to align the electronic spins with the underlying magnetic texture. The transformation corresponds to a local rotation and is implemented by the operator $U = \mathbf{m} \cdot \boldsymbol{\tau}$ for $SU(2)$ vectors and by the matrix $R_{ab} = 2m_a m_b - \delta_{ab}$ for $SO(3)$ vectors, where $\mathbf{m}(\mathbf{r}, t) = (\sin\frac{\theta}{2}\cos\phi, \sin\frac{\theta}{2}\sin\phi, \cos\frac{\theta}{2})$.

Under the above gauge rotation the derivatives transform like $\partial_\mu\psi_\sigma = U(\partial_\mu + iA_\mu)\psi_\sigma$, where $A_\mu = -iU^\dagger\partial_\mu U$ acts like an emergent electromagnetic field. The Lagrangian for the itinerant electrons then becomes

$$\mathcal{L} = \int d^2\mathbf{r} \left(i\hbar\psi_\sigma^\dagger(\partial_t + iA_0)\psi_\sigma + g\psi_\sigma^\dagger\tau^z\psi_{\sigma'} \right. \tag{20}$$
$$\left. + \sum_i \left[\frac{\hbar^2}{2m}\psi_\sigma^\dagger(\nabla_i^2 + iA_i\overset{\leftrightarrow}{\nabla}_i - A_i^2)\psi_{\sigma'} - \frac{i}{2}\psi_\sigma^\dagger(\boldsymbol{\alpha}_i' \cdot \boldsymbol{\tau})\overset{\leftrightarrow}{\nabla}_i\psi_{\sigma'} + \frac{1}{2}\psi_\sigma^\dagger(\boldsymbol{\alpha}_i' \cdot \boldsymbol{\tau})A_i\psi_\sigma \right] \right),$$

and we note that the exchange interaction is now diagonal. Instead the coupling between the spins and electrons is mediated via the gauge fields and the rotated spin-orbit interaction $\alpha_i'^a = \sum_a R_{ab}\alpha_i^b$.

We can decompose the Lagrangian into a part independent of the local magnetization and a part which depends on A_μ and $\boldsymbol{\alpha}_i$. The first part is

$$\mathcal{L}_0 = \int d^2\mathbf{r} \left[\psi_\sigma^\dagger \left(i\hbar\partial_t + \frac{\hbar^2}{2m}\nabla^2 \right)\psi_\sigma + g\psi_\sigma^\dagger\tau^z\psi_{\sigma'} \right], \tag{21}$$

which describes free electrons in presence of a magnetic field $\mathbf{B} = g\hat{\mathbf{e}}_z$. The second part of the Lagrangian is

$$\mathcal{L}_A = -\int d^2\mathbf{r} \left[\hbar a_0 \rho_\sigma + \frac{i}{2}\sum_i \left(\psi_\sigma^\dagger (\boldsymbol{\alpha}_i' \cdot \boldsymbol{\tau})\overset{\leftrightarrow}{\nabla}_i \psi_{\sigma'} - \frac{i\hbar^2}{2m}\psi_\sigma^\dagger(\mathbf{a}_i \cdot \boldsymbol{\tau})\overset{\leftrightarrow}{\nabla}_i\psi_{\sigma'} - \frac{1}{2}(\boldsymbol{\alpha}_i' \cdot \mathbf{a}_i)\rho_\sigma + \frac{\hbar^2}{2m}\mathbf{a}_i^2 \rho_\sigma\right)\right], \qquad (22)$$

where $\rho_\sigma = \psi_\sigma^\dagger \psi_\sigma$ is the electronic spin density operator and we have written $A_0 = a_0\mathbf{1}$ and $A_i = \mathbf{a}_i \cdot \boldsymbol{\tau}$.

We note that the last three terms of $\mathcal{L}_A$ can be combined into a coupling between the electronic spin current $\mathbf{j}_i$ and the emergent electromagnetic field $\mathbf{a}_i$, written on the form

$$\mathcal{L}_c = -\int d^2\mathbf{r} \sum_i \mathbf{j}_i \cdot \mathbf{a}_i \qquad (23)$$

$$\mathbf{j}_i = -\frac{i\hbar^2}{2m}\psi_\sigma^\dagger \boldsymbol{\tau}\overset{\leftrightarrow}{\nabla}_i\psi_{\sigma'} + \left(\frac{\hbar^2}{2m}\mathbf{a}_i - \frac{1}{2}\boldsymbol{\alpha}_i'\right)\rho_\sigma \qquad (24)$$

Due to the gauge transformation the spin components of the current are given in the local frame specified by the operator U. Rotating $\mathcal{L}_c$ back to the laboratory frame and using the fact that $R_{ab}a_i^b = (\nabla_i\mathbf{n} \times \mathbf{n})^a + n^a a_i^{z\,[10]}$, we find the coupling

$$\mathcal{L}_c = -\int d^2\mathbf{r} \sum_i \left(\mathbf{D}_i \cdot (\nabla_i\mathbf{n} \times \mathbf{n}) + j_i^\| a_i^z\right). \qquad (25)$$

The first term describes a Dzyaloshinskii-Moriya (DM) coupling between the localized spins, with a DM vector given by $\mathbf{D}_i = \mathbf{j}_{\perp,i} = \mathbf{j}_i - j_{\|,i}$. Here $\mathbf{j}_i$ is the current in the laboratory frame and $j_{\|,i} = \mathbf{n} \cdot \mathbf{j}_i$ is the spin component of the current parallel to the magnetic texture. The parallel component can be subtracted from $\mathbf{D}_i$ since $\mathbf{n} \cdot (\nabla_i\mathbf{n} \times \mathbf{n}) = 0$.

So far the algebraic manipulations have been exact and the Lagrangian $\mathcal{L} = \mathcal{L}_0 + \mathcal{L}_A$ gives an exact reformulation of the initial problem. To derive an effective Lagrangian for large values of g we note that for $g \to \infty$ the electronic spin component antiparallel to the local magnetization will be strongly suppressed. In this limit we can write the spin-orbit interaction like $\boldsymbol{\alpha}' \cdot \boldsymbol{\tau} \approx (\boldsymbol{\alpha} \times \mathbf{n})\tau^z$. Noting that $\mathbf{a}_i^2 = \frac{1}{4}(\partial_i\mathbf{n})^2 + (a_i^z)^{2\,[11]}$, and writing $a_i^z = \mathbf{a}$, the effective spin Lagrangian can be written

$$\mathcal{L}_s = -\int d^2\mathbf{r}\left[\frac{2S\hbar}{a^2}a_0 + H_s + \hbar a_0\rho + \frac{\hbar^2\rho}{8m}(\partial_i\mathbf{n})^2 + \sum_i \mathbf{D}_i \cdot (\nabla_i\mathbf{n} \times \mathbf{n}) + \mathbf{j}_\| \cdot \mathbf{a} + \frac{i}{2}(\boldsymbol{\alpha} \times \mathbf{n})\cdot\psi^\dagger\tau_z\overset{\leftrightarrow}{\nabla}\psi\right]. \qquad (26)$$

Here the first two terms give the Lagrangian of the isolated spin system, and ψ and ρ are the field operator and density operator for the spin component parallel to the local magnetization. We note that in the strong coupling limit the emergent vector potential is given by $a_\mu = (a_0, \mathbf{a}) = \frac{1}{2}(1 - \cos\theta)\partial_\mu\phi$.

The equations of motion for the spin system are obtained by replacing the electronic operators by their averages and varying the Lagrangian. Using the relation $\delta a_\mu/\delta\mathbf{n} = \frac{1}{2}(\partial_\mu\mathbf{n}) \times \mathbf{n}^{[11]}$ we find

$$\left(\frac{\hbar S}{a^2} + \frac{\hbar\rho}{2}\right)(\partial_t\mathbf{n}) \times \mathbf{n} + \frac{\delta\mathcal{H}}{\delta\mathbf{n}} + ([\mathbf{j}_s \cdot \nabla]\mathbf{n}) \times \mathbf{n} = 0, \qquad (27)$$

where the contribution from the effective spin Hamiltonian is

$$\frac{\delta\mathcal{H}}{\delta\mathbf{n}} = \frac{\delta}{\delta\mathbf{n}}\left(H_s + \frac{\hbar^2\rho}{8m}(\partial_\mu\mathbf{n})^2 + \mathbf{D}^i \cdot (\partial_i\mathbf{n} \times \mathbf{n})\right) + \frac{1}{2}\boldsymbol{\alpha} \times \mathbf{j}_e. \qquad (28)$$

Here $\mathbf{j}_e = \psi^\dagger\overset{\leftrightarrow}{\nabla}\psi$ and the second and third terms in the first line renormalize the exchange and DM interactions to $J \to J + \hbar^2\rho/(4m)$ and $\mathbf{D}_i \to \mathbf{D}_i + \mathbf{j}_{\perp,i}$. The current $\mathbf{j}_s$ is given by

$$\mathbf{j}_s = -\frac{i\hbar^2}{2m}\psi^\dagger\overset{\leftrightarrow}{\nabla}\psi + \left(\frac{\hbar^2}{m}\mathbf{a} + \frac{1}{2}\boldsymbol{\alpha} \times \mathbf{n}\right)\rho. \qquad (29)$$

Multiplying from the right by $\times\mathbf{n}$ we arrive at the modified Landau-Lifshitz equation

$$\partial_t\mathbf{n} = -\left(\frac{\hbar S}{a^2} + \frac{\hbar\rho}{2}\right)^{-1}\left(\mathbf{n} \times \frac{\delta\mathcal{H}}{\delta\mathbf{n}} + \hbar(\mathbf{j}_s \cdot \nabla)\mathbf{n}\right). \qquad (30)$$

We see that apart from the renormalization of the exchange and DM interactions discussed above, the electrons affect the spin dynamics in the following ways: (i) The prefactor describes the total magnetization density (instead

of the density of local moments), with $\rho = \langle \psi^\dagger \psi \rangle$ the density of electrons with spins parallel to the local moments. This can be understood by observing that in the $g \to \infty$ limit the electronic spins and localized moments get locked in a parallel configuration, forming a magnetic moment of magnitude $M = S/a^2 + \rho/2$. (ii) The spin-orbit coupling contributes an effective magnetic field $\mathbf{B}_{so} = (1/2)\boldsymbol{\alpha} \times \mathbf{j}_e$ to the spin Hamiltonian, which for a Rashba type interaction is parallel to the plane of the spins. (iii) The localized moments couple to the parallel component of the spin current via the last term in Supplementary Equation 30.

If we start from a ferromagnetic state $\mathbf{n} = \hat{\mathbf{e}}_z$ the last term of Supplementary Equation 30 is zero. Unless the system spontaneously reorders due to the renormalization of the spin parameters (which happens on long time scales), it is therefore necessary with a non-zero spin-orbit coupling in order to tilt the spins away from their ferromagnetic alignment.

We end this section by calculating the equilibrium renormalization of the DM interaction. Assuming that the first two terms of the spin current $\mathbf{j}_i$ vanish in the ground state, we have $\mathbf{D}_i = -(\rho/2)\boldsymbol{\alpha}_i$ (remembering that the spin current is given in the laboratory frame). We thus find $\mathbf{D}_x = -D\hat{\mathbf{e}}_y$, $\mathbf{D}_y = D\hat{\mathbf{e}}_x$ and $\mathbf{D}_z = 0$ with $D = \rho\alpha/2$, and the DM term in the effective Hamiltonian can be written as

$$\mathbf{D}^i \cdot (\partial_i \mathbf{n} \times \mathbf{n}) = D(\mathbf{n} \cdot \boldsymbol{\partial})n_z - Dn_z(\boldsymbol{\partial} \cdot \mathbf{n}), \tag{31}$$

where $\boldsymbol{\partial} = (\partial_x, \partial_y, 0)$. This is the continuum version of a DM Hamiltonian of the Néel type. The renormalized DM interaction in equilibrium is therefore obtained by the replacement $D \to D + \rho\alpha/2$.

SUPPLEMENTARY NOTE 7: MULTIPLE SKYRMION CONFIGURATIONS

Supplementary Figure 3 in the main text involves spin configurations with multiple skyrmion excitations. Some of these multiple skyrmion configurations are shown in Supplementary Figure 3.

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[1] The Heisenberg term comes from writing the Coulomb exchange operator as $d_{i\sigma}^\dagger d_{i\sigma'} d_{j\sigma'}^\dagger d_{j\sigma} = (n_i n_j)/2 + 2\hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j$, and absorbing the first term by a renormalization of V_0.

[2] Schüler, M., Rösner, M., Wehling, T. O., Lichtenstein, A. I. & Katsnelson, M. I. Optimal hubbard models for materials with nonlocal coulomb interactions: Graphene, silicene, and benzene. *Phys. Rev. Lett.* **111**, 036601 (2013). https://link.aps.org/doi/10.1103/PhysRevLett.111.036601.

[3] Yildirim, T., Harris, A. B., Aharony, A. & Entin-Wohlman, O. Anisotropic spin hamiltonians due to spin-orbit and coulomb exchange interactions. *Phys. Rev. B* **52**, 10239–10267 (1995). https://link.aps.org/doi/10.1103/PhysRevB.52.10239.

[4] Ogawa, N., Seki, S. & Tokura, Y. Ultrafast optical excitation of magnetic skyrmions. *Sci. Rep.* **5**, 9552 (2015). https://doi.org/10.1038/srep09552.

[5] Khoshlahni, R., Qaiumzadeh, A., Bergman, A. & Brataas, A. Ultrafast generation and dynamics of isolated skyrmions in antiferromagnetic insulators. *Phys. Rev. B* **99**, 054423 (2019). https://link.aps.org/doi/10.1103/PhysRevB.99.054423.

[6] Kusminskiy, S. V. *Quantum Magnetism, Spin Waves, and Optical Cavities* (Springer International Publishing, 2019). https://doi.org/10.1007/978-3-030-13345-0.

[7] Yin, G. *et al.* Topological charge analysis of ultrafast single skyrmion creation. *Phys. Rev. B* **93**, 174403 (2016). https://link.aps.org/doi/10.1103/PhysRevB.93.174403.

[8] Tan, S., Jalil, M., Fujita, T. & Liu, X. Spin dynamics under local gauge fields in chiral spin–orbit coupling systems. *Annals of Physics* **326**, 207–215 (2011). https://doi.org/10.1016/j.aop.2010.11.014.

[9] Fujita, T., Jalil, M. B. A., Tan, S. G. & Murakami, S. Gauge fields in spintronics. *Journal of Applied Physics* **110**, 121301 (2011). https://doi.org/10.1063/1.3665219.

[10] Kikuchi, T., Koretsune, T., Arita, R. & Tatara, G. Dzyaloshinskii-moriya interaction as a consequence of a doppler shift due to spin-orbit-induced intrinsic spin current. *Phys. Rev. Lett.* **116**, 247201 (2016). https://link.aps.org/doi/10.1103/PhysRevLett.116.247201.

[11] Han, J. H. *Skyrmions in condensed matter*. Springer Tracts in Modern Physics (Springer International Publishing, 2017).

[12] Tatara, G. Effective gauge field theory of spintronics. *Physica E: Low-dimensional Systems and Nanostructures* **106**, 208–238 (2019). https://doi.org/10.1016/j.physe.2018.05.011.
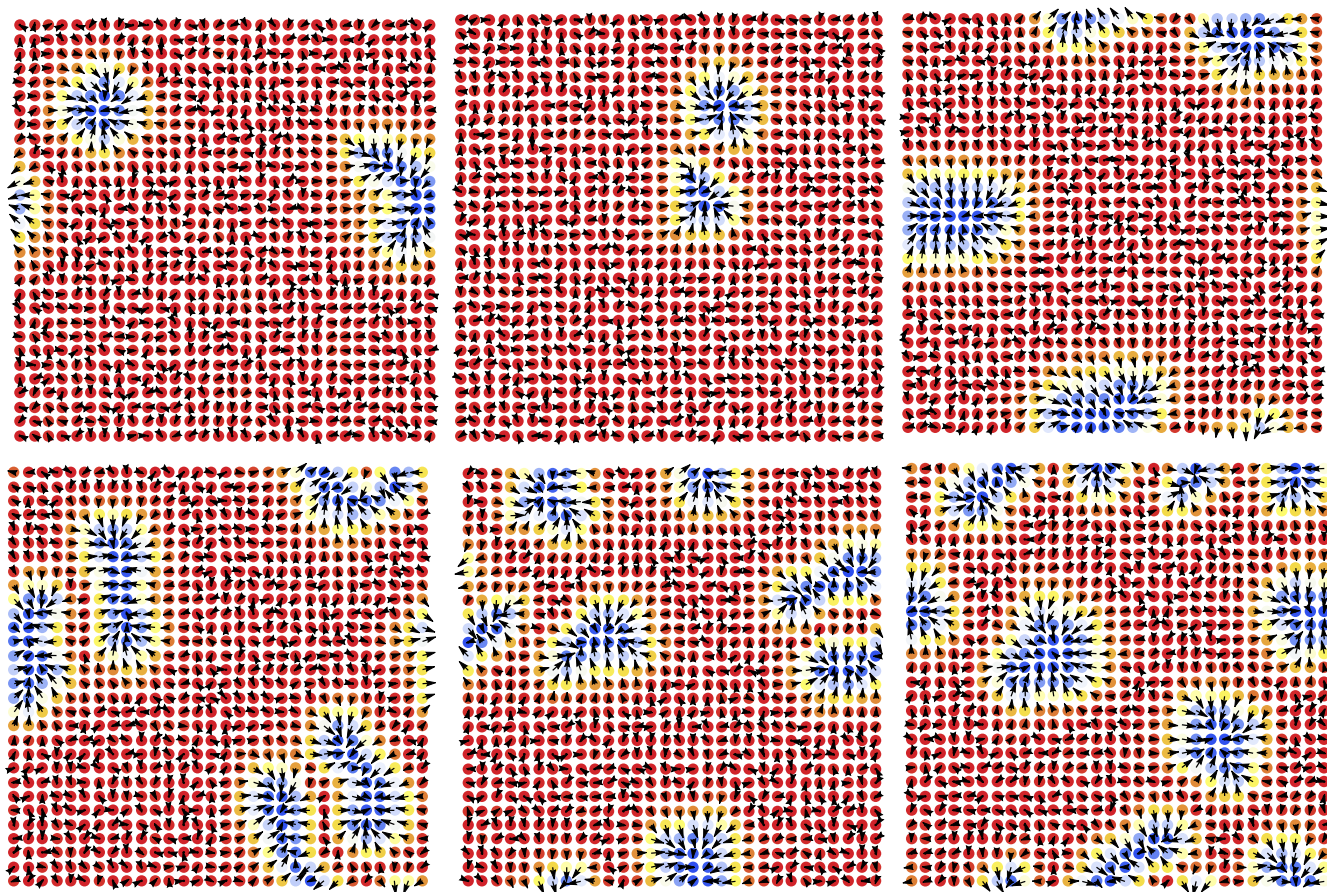
Supplementary Figure 3. Light-induced multiple skyrmion states. Spin configurations at the end of the time-evolution ($t = 330$ fs) corresponding to skyrmion states with a total topological charge $Q = 2$, 2, and 3 for the top row (left to right), and for $Q = 4$, 5 and 7 for the bottom row (left to right).