Spin flips of electron beam in optical near field

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Manipulating the spin polarization of electron beams using light is highly desirable but exceedingly challenging, as the approaches proposed in previous studies using free-space light usually require enormous laser intensities. We here propose to use a transverse electric optical near field extended on nanostructures to efficiently induce spin flips of an adjacent electron beam, exploiting the strong inelastic electron scattering in phase-matched optical near fields. Our calculations show that, using a dramatically reduced laser intensity ($\sim 10^{12}$ W/cm$^2$) and for a short interaction length (16 $\mu$m), an electron spin-flip probability of approximately 12\% can be achieved. More intriguingly, for an unpolarized incident electron beam, its two spin components, parallel and antiparallel to the electric field, are spin-flipped and inelastically scattered to different energy states, providing an analog of the Stern-Galarch experiment in the energy dimension. Our findings are important for optical control of free-electron spins, preparing spin-polarized electron beams, and applications as varied as in material science and high-energy physics.

I. INTRODUCTION

After Stern and Gerlach’s celebrated experiment using inhomogeneous magnetic fields to separate neutral atoms by spin, Bohr and Pauli pointed out that a similar scheme could not be simply applied to polarize charged particles due to the influence of Lorentz force \cite{1}. Consequently, alternative mechanisms are required to prepare spin-polarized electrons \cite{2}. Two main approaches have been implemented experimentally to generate spin-polarized electrons, one using semiconductor photocathodes with negative electron affinities \cite{3,4}, or the Kapitza-Dirac effect (KDE) \cite{29–39}. Importantly, spin-polarized electron beams have served as an essential tool for investigating the magnetic properties of solid-state materials and molecules \cite{9,11,12,14}, and studying fundamental problems in high-energy physics \cite{15,17}.

Despite a few works still questioning the possibility of separating different electron spins by static magnetic fields \cite{18,22}, more concentrated efforts have been devoted to exploring the possibility of polarizing electrons using free-space light. In these studies, the spin dynamics of electrons are manipulated via Compton scattering \cite{23–28}, or the Kapitza-Dirac effect (KDE) \cite{29–39}, where a single laser beam or two counter-propagating lasers (i.e., in the KDE \cite{10,22}) are involved, respectively. However, these free-space phenomena are at least second-order quantum processes, which are significant only for enormous laser intensities (typically, $10^{18} \sim 10^{22}$ W/cm$^2$, depending on the interaction time, see Appendix). The

more efficient first-order quantum processes are simply forbidden by the the energy-momentum conservation laws. More precisely, in free space, it is not possible to switch the spin AM of an electron between $-\hbar/2$ and $\hbar/2$ by directly absorbing or emitting a photon with AM of $\hbar$.

In contrast to these free-space scenarios, the interactions between moving electrons and optical modes in media or localized structures are intrinsically inelastic, mediated by the large photonic momenta in the near field. For example, in Cherenkov radiation (CR) and electron energy-loss spectroscopy (EELS), electron beams spontaneously release photons into vacuum photonic modes. Recently, the photon-induced near-field electron microscopy (PINEM) technique has been intensively investigated \cite{43–61}. Unlike those spontaneous-emission-like phenomena, i.e., CR and EELS, a laser is introduced in PINEM to excite the optical near field of nanostructures. At moderate laser intensity, the probability of an electron being inelastically scattered while moving through the optical near field can reach unity. Meanwhile, multiple events of photon-emission and photon-absorption can occur for a single electron. Surprisingly, by tailoring the sample geometry and illumination to match the electron velocity and near-field phase velocity, recent experiments have achieved scattered electron energy shifted by hundreds of photon energy quanta \cite{59,60}. Provided such strong inelastic scattering of electrons experienced in PINEM, it is thus illuminating to consider whether electron spin-flip transitions can be efficiently achieved by direct AM exchanges with optical near fields.

In this work, we reveal a pathway to achieve significant spin flips of electron beams via interactions with a transverse electric (TE) optical near field, exploiting the efficient inelastic electron scattering in the optical near field as in PINEM. Compared to previous studies involving only free-space light, the intensity of a laser used to in-
duce pronounced electron spin-flip transitions is dramatically reduced (∼10^{12} \text{ W/cm}^2 in our calculation, which can be further decreased). As the interactions between the electron beam and optical near field are intrinsically inelastic, electron spin can be flipped by directly absorbing or emitting a photon of energy $E_\text{p}$, while its spin could be flipped.

II. MODEL SYSTEM

We consider in this work a periodic two-dimensional (2D) nanostructure normally illuminated by a laser, as exemplified by a nanowire array (see Fig. 1). The excited optical near field can induce inelastic and spin-flip scattering of an adjacent electron beam (propagating along $+\hat{z}$). For simplicity, we also assume a 2D electron beam with its y-component wave vector vanishing. In PINEM, the electric field component parallel to the electron beam dominates and induces only spin-preserving transitions. Instead, we consider here a TE configuration (only $\mathbf{E}_y$ is present, see Fig. 1), in which the spin-flip transitions can occur (see below). Regarding the nanowire array under such TE illumination, the total electric field includes a near-field component and can be approximated by $\tilde{\mathbf{E}}_y(r,t) = \tilde{\mathbf{E}}_y^{\text{in}}(x) + 2\Delta \mathbf{E}_y(x) \cos(gz)$, where $g = 2\pi/a$.

We also assume a monochromatic light field for analysis, i.e., $\tilde{\mathbf{E}}_y(r,t) = \tilde{\mathbf{E}}_y^{\text{in}}(x) e^{-i\omega t} + c.c.$, which can be easily extended to a pulsed laser field as in the PINEM theory [59].

Similar to the PINEM experiment [59, 60], to guarantee a strong electron-photon interaction, we consider throughout this work a resonant situation, assuming that the initial electron velocity matches the near-field phase velocity, i.e., $\beta = v_0/c = a/\lambda$, where $\lambda$ is the vacuum photon wavelength. In contrast to the high-energy electron beams (>100 keV) usually adopted in PINEM (see exceptions in [62, 63]), as we shall explain below, the spin-flip transitions of the electrons are only significant for low-energy electrons. We also note that such a low-energy electron beam is not tightly bounded (see red shade in Fig. 1) as in PINEM, and more complicated is that a high laser intensity required by significant spin-flip transitions can also cause reshaping of the electron beam.

Our theory presented below can fully capture both the spin dynamics and diffraction effects of electron beams in the optical near field.

III. THEORY OF SPIN DYNAMICS

As the spin-flip effect is significant only for low electron velocity (see below), we restrict this work to non-relativistic situations. In this limit, the spin dynamics of an electron beam is governed by Pauli’s equation, $\hat{\mathbf{p}}^2/2m_e + \hat{H}_I \Psi(r,t) = i\hbar \partial_t \Psi(r,t)$ ($\mathbf{r} = (x,z)$), where the interaction Hamiltonian reads

$$\hat{H}_I = \frac{e}{m_e c} \mathbf{A}(r,t) \cdot \hat{\mathbf{p}} + \mu_B \mathbf{B}(r,t) \cdot \hat{\sigma} + \frac{e^2}{2m_e c^2} \hat{\mathbf{A}}^2(r,t),$$

$\hat{\mathbf{p}}$ is the momentum operator, $\mu_B = e\hbar/2m_e c$ is the Bohr magneton, and $\hat{\sigma} = [\sigma_x, \sigma_y, \sigma_z]$ is the three-vector Pauli matrix. The wave function in Pauli’s equation is described using the two-component spinors, $\Psi(r,t) = \Psi^+(r,t) \hat{s}^+ + \Psi^-(r,t) \hat{s}^-$, where $\hat{s}^+ = [\cos(\theta/2), \sin(\theta/2)e^{i\varphi}]^T$ and $\hat{s}^- = [\sin(\theta/2), -\cos(\theta/2)e^{i\varphi}]^T$ denote the two possible spins of electrons along the direction defined by two polar coordinates $(\theta, \varphi)$ (see Fig. 1). To derive the interaction Hamiltonian $\hat{H}_I$ above, a radiation gauge is chosen so that the scalar potential vanishes and the vector potential is related to the electric field by $\mathbf{A}(r) = -ie/\omega \mathbf{E}(r)/q$, where $q = \omega/c$. In what follows, we refer to the three terms in $\hat{H}_I$, from left to right, as $\hat{H}^{(1)}_I$, $\hat{H}^{(2)}_I$, and $\hat{H}^{(3)}_I$, respectively.

Considering an electron beam of initial energy $E_0$ interacting with an optical near field, the electron energy should spread only among discrete levels, i.e., $E_n = E_0 + n\hbar \omega$. In the case of a paraxial and slowly varying electron beam, we can expand the electron wave function as $\Psi^\pm(r,t) = \sum_n \psi_n^\pm(r)e^{i(p_n z - E_n t)/\hbar}$, where $p_n = \sqrt{2m_e E_n} \approx p_0 + n \hbar \omega / v_0$. Using this expansion and together with a nonrecoil approximation, Pauli’s equation can be rewritten as a set of coupled equations (see Appendix),

$$\left( v_0 \hat{p}_z + \frac{p_z^2}{2m_e} \right) \psi_n^+(r) = - \sum_{n',s'} M_{ns's'}^{n'n'} \psi_{ns'}^+(r),$$

(1)
where the nonzero transition matrix elements for the TE electromagnetic field are

\[ M_{n,n-1}^{ss'} = e^{-i \frac{\omega}{c} \Delta E_y (x) / q}, \]

\[ M_{n,n}^{ss} = \frac{e^2}{m_e \omega^2} |E_y (r)|^2, \]

and their Hermitian conjugates \( M_{n,n}^{s's} = (M_{n,n}^{ss'})^* \). The above matrix elements reveal that the magnetic field can induce both spin-flip and spin-preserving transitions. In addition, the interaction term \( H_I^{(3)} \), which corresponds to the ponder-motive force, introduces an additional phase to the wave function [64].

As seen from Eq. (2), for the infinitely extended nanostructure shown in Fig. 1 resonantly-enhanced interactions arise when \( \omega / v_0 = g \) [59, 60]. Additionally, in Eq. (2), only the Fourier component of the near field, with momentum along the \( z \)-direction equal to \( g \), contributes to the electron dynamics, e.g., the electric field component \( \Delta E_y (x) e^{i \omega x} \) (see Fig. 1). Such Fourier component is an evanescent wave captured by a factor \( e^{-i q x} \), where \( q_x = \pm \sqrt{g^2 - q^2} \) (we chose + sign hereafter), so the corresponding magnetic field component is found to be \( \Delta B (r) = q \times \Delta E_y (x) e^{i \omega x} / q \), according to Maxwell’s equations, where \( q = (i q_x, 0, g) \). Using the magnetic field component \( \Delta B (r) \), we can further simplify the off-diagonal matrix elements in Eq. (2) to

\[ M_{n,n+1}^{s's} = \mu_B \langle s | (q \times \hat{y}) \cdot \vec{d} | s' \rangle \Delta E_y (x) / q. \]  

Importantly, we note that \( \Delta B (r) \) is circularly polarized when \( g \gg q \), which is associated with the spin AM in the evanescent near field [65, 66]. Meanwhile, when \( g \gg q \), the above matrix element is approximated by \( M_{n,n+1}^{s's} \approx 2 \mu_B \Delta E_y \beta^{-1} \) for spin flips along the \( y \)-direction.

In the weak-interaction regime, the spin-flip transition probabilities over a propagation distance of \( L \) can be evaluated, according to Eqs. (1) and (3), by \( |2 \mu_B \Delta E_y L / c h|^2 \beta^{-4} \), which is greatly increased when electrons slow down. However, an excessively small electron velocity requires a more localized near field (\( \sim e^{-g x} \)) to fulfill the phase-match condition, while widening the electron beam waist and thus reducing the chance of electron-photon interaction. Consequently, we assume a moderate electron velocity \( \beta = 1/10 \) in the detailed calculations below.

### IV. DIFFRACTIONLESS APPROXIMATION

To concisely reveal the spin-flip physics of an electron beam in optical near fields, we first proceed with calculations based on the diffractionless approximation (\( \Delta g^2 = 0 \)), assuming that the electron dynamics on different straight trajectories (\( x = \text{const.} \)) are independent. With this approximation, the electron spin dynamics is simply governed by a one-dimensional (1D) equation reduced from Eq. (1). The numerical results calculated

- by the reduced 1D equation are shown in Fig. 2, where we adopt the simplified matrix elements in Eq. (3) and assume spin-polarized incident electron beams.

Since the magnetic near-field component \( \Delta B \) is circularly polarized, the electron spin dynamics in the \( x-z \) plane is isotropic. Consequently, we chose the incident electron spin along \( +\hat{z} \) as an example to illustrate the electron spin dynamics in the \( x-z \) plane, as shown in Fig. 2(a) and (b). During interaction with the near field, the

![FIG. 2. (a)-(f) Distributions of (a),(c),(e) spin-preserving (|\psi_n^+|^2) and (b),(d),(f) spin-flipped electrons (|\psi_n^-|^2) among discrete energy levels (plotted in color scale as ribbons) for different propagation distances, calculated within the diffractionless approximation (x=const.). The input electron beams are spin-up polarized (see black arrows, i.e., \( \psi_0^+ \)) along (a) +\( \hat{z} \), (c) +\( \hat{y} \), and (e) -\( \hat{y} \), respectively. (g) Spin-flip probabilities for incident electron spins along \( \hat{z} \) and \( \hat{y} \) show the same period of \( L_p \). (h) The period \( L_p \) for different \( \Delta E_y \) (see definition in Fig. 1). We assume a electric field component \( \Delta E_y = 1 \times 10^7 \) V/cm in (a)-(g), and use parameters, \( \lambda = 1000 \) nm and \( a = 100 \) nm, for all calculations.

![Diagram](https://via.placeholder.com/150)
focused at $z = 0$ and $x = 0.3a$ with a waist of 3 nm. In (c), the spin-flipped electrons are scattered to the state of energy $E_{\theta}$. The unperturbed beam width is shown by white dashed curves in (b). The electron beam is slightly deflected due to a reshaping effect in the optical field, and the nanowire array is adaptively rearranged to avoid crossing (the top of the nanowires are shown by white solid curve). The incident electric field is assumed to be $E_{\text{in}} = 4 \times 10^{7}$ V/cm.

occupancy of the initial electron state, $|\psi_{0}^{+}(z = 0)|^{2} = 1$, gradually decreases [Fig. 2(a)], while the adjacent energy levels $\psi_{\pm 1}^{\pm}$ of both spin directions along $\pm \hat{z}$ [see Fig. 2(a) and (b)] are equally populated via inelastic scattering. As the electron beam propagates further, the populations show an oscillatory behavior, with still only these above-mentioned states, $\psi_{0}^{+}$ and $\psi_{R}^{+}$, populated. This is in contrast with the PINEM process, as in PINEM, the electron populations constantly spread out and randomly walk [61] among energy levels.

The incident electron spin along $+\hat{z}$ gives rise to a symmetric distribution of electrons among energy states, due to the lack of a symmetry breaking mechanism. In contrast, for an incident electron spin along $\pm \hat{y}$, the electron spin is parallel to the spin AM of the near field, as revealed by the circular polarization of $\mathbf{B}$. In this configuration, the electron chirally couples to the field, leading to asymmetric patterns of the energy state populations [Fig. 2(c)-(f)]. For the incident electron spin along $+\hat{y}$, the possible transition from the initial state is that, the electron simultaneously releases energy quanta of $\hbar \omega$ and AM of $h$ into the optical near field, namely $\psi_{0}^{+} \rightarrow \psi_{-1}^{-}$ [Fig. 2(c) and (d)]. Likewise, the incident electron spin along $-\hat{y}$ only gives rise to the transition $\psi_{0}^{-} \rightarrow \psi_{-1}^{+}$ [Fig. 2(c) and (f)]. Intriguingly, such asymmetric inelastic scattering offers a pathway to polarize electron beams in a mixed state. To be more precise, for an unpolarized incident electron beam, the electrons scattered to states of energy $E_{n+1}$ and $E_{n-1}$ are spin polarized along $+\hat{y}$ or $-\hat{y}$, respectively, which can be further separated by an energy filter.

The energy level occupancy [Fig. 2(a)-(f)] and the spin-flip probability [Fig. 2(g)], defined by $P_{\text{flip}} = |\Psi^{+}|^{2}/|\Psi|^{2}$, both exhibit oscillatory behaviors with the same period $L_{p}$. At the distance of $L_{p}/2$, where the initial state $\psi_{0}^{+}$ is fully depleted, for the incident electron spin along $+\hat{x}$ and $+\hat{y}$, the total electron population is equally allocated to states $\psi_{0}^{+}$ or totally transferred to the state $\psi_{1}^{-}$, leading to maximum $P_{\text{flip}}$ of 0.5 and 1, respectively [see Fig. 2(g)].

The repetition of population among states observed here is in fact a magnetic Rabi oscillation, and the period can be estimated by $L_{p} = 2\pi v_{0}/\Omega_{e}$, where the magnetic field induces a Rabi frequency of $\Omega_{e} = 4\mu_{B}\Delta E_{g}/e v_{0}h$ [Fig. 2(h)].

V. SPIN-FLIP OF DIFFRACTIVE ELECTRON BEAM

To include the diffraction effects, we perform 2D calculations of Eq. (1), taking into account the transverse momentum (the $p_{x}^{2}$ term) and the ponderomotive force. In the calculation, we adopt a numerically solved electromagnetic field [see Fig. 3(a)], where $\mathbf{B}$ is enhanced near the nanowire. We consider here a Gaussian electron beam, spin-polarized along $+\hat{y}$ and propagating though the near field calculated above. The electron beam is focused at $z = 0$ with a distribution of $\Psi(x, z = 0) = e^{-\left(x - 30 \text{ [nm]}\right)^{2}/w_{0}^{2}}$, where $w_{0} = 3$ nm is the beam waist. The unperturbed Gaussian beam width, $w(z) = w_{0}\sqrt{1 + \left(z/z_{R}\right)^{2}}$, is indicated by dashed curves in Fig. 3(b), where $z_{R} = \pi w_{0}^{2}/\lambda_{e}$ and $\lambda_{e} = 2\pi \hbar/p_{0}$ is the electron wavelength.

When interaction with the electromagnetic field is considered, the Gaussian electron beam is reshaped due to an additional phase imprinting by the ponder-motive force (see, e.g., [62]). According to Eq. (1), for the TE field considered in this work, the ponder-motive force introduces a phase, captured by $d\phi = eq^{2}\int|E_{y}|^{2}dz/2m_{e}v_{0}$. In this regard, the electron wave segment propagating in a region of higher intensity accumulates a larger
phase, so that the electron beam is deflected toward the nanowires surrounded by lower field intensity [see \( E_{\perp} \) in Fig. 3(a)]. To avoid crossing of the electron beam with the nanowires, as shown by solid curve of \( \gamma \) in Fig. 3(b), we adaptively displace each nanowire slightly in the x-direction, maintaining the distance between the center of each nanowire, \( (x_j, z_j) \) (jth nanowire), and the electron-density peak in the plane \( z = z_j \). With this adaptive design, the electron beam experiences an acceleration along the x-direction, following a parabolic-like trajectory. Additionally, the reshaping effect also leads to a better confinement and even re-focusing of the electron beam. We also note that the paraxial approximation assumed to derive Eq. (1) is valid, since the electron beam deflection angle, \( \delta \approx 1.8a/120a \approx 15\,\text{mrad} \) [see Fig. 3(b)], is still small.

Despite the reshaping effect, our adaptive design of the nanowire array guarantees a strong interaction between the electron beam and the magnetic field localized around the nanowire [see B in Fig. 3(a)], which gives rise to efficient spin-flip transitions. As shown in Fig. 3(c) and (d), the population of the spin-flipped electrons of energy \( E_0 - E_p \) [see Fig. 2(d)] continuously increases along the propagation, and the spin-flip probability \( P_{\text{flip}} \) reaches \( \sim 12\% \) at a propagation distance of 16\,\mu m. Similar spin-flip processes can also be observed for an incident electron spin in the x-z plane, but with the spin-flip probability \( P_{\text{flip}} \) nearly halved, as can be anticipated from Fig. 2(g). According to our discussions on Fig. 2(c)-(f), for an unpolarized incident electron beam, when all other parameters same as in Fig. 3 at the propagation distance of 16\,\mu m, 12\% electrons in the beam are scattered to the two states of energies \( E_0 + E_p \) and \( E_0 - E_p \) (see Appendix) (6\% in each state), which have opposite spins in the y-direction and can be further separated by an electron spectrometer.

VI. CONCLUDING REMARKS

In this work, we propose a spin-flip effect of electrons in optical near field, which is exceptionally efficient compared to previous proposals in free space (see Appendix). Apart from the requirement of ultra-strong laser intensity, previous schemes using free-space light to spin flip or polarize electron beams also face other experimental difficulties, such as measurement stability and electron-laser overlap under extreme conditions [83, 89]. The spin-flip effect proposed in this work is feasible, since PINEM is already a well-established technology. The incident laser intensity adopted in Fig. 3 \( I^\text{in} = 8.5 \times 10^{12} \, \text{W/cm}^2 \), is attainable using commercial ultrafast lasers, and the intensity \( I^\text{in} \) required to achieve the same \( P_{\text{flip}} \) as in Fig. 3 can be further reduced, by extending the interaction length. As the typical laser damage threshold of metals is several J/cm² and the plasmon-enhanced absorption is absent for the TE illumination, the nanostructures should sustain a laser illumination with a pulse duration of \( \sim 100\,\text{fs} \), provided the peak intensity of \( I^\text{in} = 8.5 \times 10^{12} \, \text{W/cm}^2 \). In addition, by using a higher input electric current, the laser intensity required to induce observable spin-flipped electrons can be further reduced in future experiments to test the spin-flip effect proposed in this study.

Appendix A: Derivation of Eq. (1) in the main text

In the main text, Eq. (1) is used to describe the spin-flip and beam-diffraction effects. We provide here a detailed derivation of the more general form of Eq. (1), which is capable of describing paraxial electron dynamics in arbitrary electromagnetic field. As explained in the main text, in the nonrelativistic limit, the evolution of an electron wave function involving the spin degrees of freedom can be fully captured by Pauli’s equation,

\[
\left( \frac{\hat{p}^2}{2m_e} + \hat{H}_I \right) \Psi(r, t) = i\hbar \partial_t \Psi(r, t),
\]

where the interaction Hamiltonian is explicitly written as

\[
\hat{H}_I = \frac{e}{2m_e c} \left[ \mathbf{A}(r, t) \cdot \hat{p} + \hat{p} \cdot \mathbf{A}(r, t) \right] + \mu_B \mathbf{B}(r, t) \cdot \mathbf{s} + \frac{e^2}{2m_e c^2} \mathbf{A}^2(r, t).
\]

The Pauli equation can be derived from the Dirac equation in the non-relativistic limit. This approximation can also introduce the spin-orbit interaction term, which can be neglected in our work since we only study the paraxial electron beams here. In fact, in the studies on electron-spin separation, the spin-orbit interaction effect always provides a minor contribution. In Eq. (A1), we also choose the radiation gauge, so that the scalar potential vanishes and the electric field is related to the vector potential by \( \mathbf{E}(r, t) = -\partial_t \mathbf{A}(r, t)/c \). In our work, we also deal only with monochromatic electromagnetic fields, and for all the field quantities, we use the following notation, e.g., \( \mathbf{E}(r, t) = \mathbf{E}(r)e^{-i\omega t} + \text{c.c}. \).

Within the paraxial approximation, the electron dynamics on different straight lines is weakly coupled, so we can expand the electron wave function using the 1D plane wave eigenstates,

\[
\Psi(r, t) = \sum_{s=\pm} \Psi^s(r, t) = \sum_n \psi_n^s(r) e^{i(p_n z - E_n t)/\hbar} |s\rangle,
\]

where \( p_n = \sqrt{2m_e E_n} \) is the canonical momentum of electron in the state of energy \( E_n \), and \( |s = \pm\rangle \) is the spinor denoting the electron spin. Inserting this expansion into the Pauli equation [Eq. (A1)] and eliminating those terms corresponding to the non-perturbative Schrodinger equa-
tion, we find

\[
\sum_{n,s} e^{i(p_nz-E_n t)/\hbar} \left[ \frac{\hat{p}_z^2}{2m_e} + \hat{v}_n \hat{p}_z + \frac{e}{c} v_n A_z(r, t) \right. \\
+ \mu_B B(r, t) \cdot \hat{\sigma} + \frac{e^2}{2m_e \omega^2} A^2(r, t) \left. \right] \psi_n^s(r) \mid s \rangle = 0,
\]

(A2)

where \( v_n = p_n/m_e \) is the canonical velocity of electron in the state of energy \( E_n \), and \( \hat{v}_n = v_n + e A_z(r, t)/m_e c \) is the corresponding kinetic velocity. To derive the above equation, we also neglect the terms containing \( \hat{p}_z^2 \) and \( \hat{p}_z \cdot \hat{A}_z(r, t) \), assuming that the electron wave amplitude \( \psi_n(r) \) and the electromagnetic field amplitude \( A_z(r) \) vary slowly in space with respect to the phase term \( e^{i p_n z} \).

We then multiply the above equation by the eigenfunction \( e^{-i(p_{n'}z-E_{n'}t)/\hbar} \langle s' \mid s \rangle \) and perform a summation over the introduced index \((n', s')\). By retaining only the time-independent terms, Eq. (A2) is rewritten as

\[
0 = \sum_{n,n',s'} \left\{ \sum_{n,s} \delta_{n,n'} \left[ \frac{\hat{p}_z^2}{2m_e} + \hat{v}_n \hat{p}_z + \frac{e^2}{2m_e \omega^2} |\mathbf{A}(r)|^2 \right] \psi_n^s(r) \right. \\
+ \sum_{n,s'} \delta_{n,n'-1} e^{i\pi(p_{n'-1}-p_{n'})z} \left[ \delta_{s,s'} \frac{e}{c} v_n A_z(r) + \langle s' \mid \mu_B B(r) \cdot \hat{\sigma} \rangle \right] \psi_n^{s'}(r) \\
+ \sum_{n,s''} \delta_{n,n'+1} e^{i\pi(p_{n'+1}-p_{n'})z} \left[ \delta_{s,s''} \frac{e}{c} v_n A_z^*(r) + \langle s' \mid \mu_B B^*(r) \cdot \hat{\sigma} \rangle \right] \psi_n^{s''}(r). \]

All other time-oscillating terms are neglected, because their contributions are averaged out along the propagation of electrons. The above equation includes a set of coupled differential equations, which can be rewritten in the matrix form,

\[
(\hat{v}_n \hat{p}_z + \frac{\hat{p}_z^2}{2m_e}) \psi_n^s(r) = -M_{n,n'}^{s,s'} \psi_n^{s'}(r), \quad (A3)
\]

where the nonzero matrix elements of \( M_{n,n'}^{s,s'} \) are

\[
M_{n,n-1}^{s,s} = e^{i\pi(p_{n'-1}-p_{n})z} \left[ -\delta_{s,s'} \frac{ie}{\omega} v_{n-1} \mathcal{E}_z(r) + \mu_B \langle s \mid \mathbf{B}(r) \cdot \hat{\sigma} \rangle \langle s' \rangle \right],
\]

\[
M_{n,n+1}^{s,s} = e^{i\pi(p_{n+1}-p_{n})z} \left[ -\delta_{s,s'} \frac{ie}{\omega} v_{n+1} \mathcal{E}_z^*(r) + \mu_B \langle s \mid \mathbf{B}^*(r) \cdot \hat{\sigma} \rangle \langle s' \rangle \right],
\]

\[
M_{n,n}^{s,s} = \frac{e^2}{m_e \omega^2} |\mathcal{E}(r)|^2.
\]

In the previous PINEM theory, the field contribution in the canonical momentum is regarded as small and thus \( \hat{v}_n \) is approximated by \( v_n \). More safely, for the transverse electric field studied in our work, the component of the vector potential parallel to the electric field is absent, so \( \hat{v}_n = v_n \), rigorously satisfied. In the case of nonrecoil limit, i.e., \( E_0 \gg \hbar \omega \), the above equation can be further simplified using following approximations, \( v_n \approx v_0 \) and \( p_{n+1} - p_n = \hbar \omega/v_0 \), and Eq. (A3) finally reduces to Eq. (1) in the main text. In addition, the matrix \( M \) in the above equation does not seem to be Hermitian, because we neglect the term \( \hat{p}_z \cdot \hat{A}_z(r, t) \) in Eq. (A2). Nevertheless, the Hermiticity is recovered in the nonrecoil approximation.

Appendix B: Comparison of spin-flip efficiency with previous works

In Fig. 3, we chose a laser intensity of \( 8.5 \times 10^{12} \text{ W/cm}^2 \), while in previous studies using free-space laser to induce the electron spin flips, the adopted intensities are mainly within the range of \( 10^{18} \sim 10^{22} \text{ W/cm}^2 \) (see the references in the second paragraph of the main text). Accidentally, a few works even adopted lower laser intensities than our work. The laser intensities in these studies vary by several orders, because the assumed interaction lengths \( L \) and the achieved spin-flip probabilities are different.

In order to make a reasonable comparison of the spin-flip efficiencies in different works, we define the efficiency by averaging the spin-flip probability \( P_h \) over the inter-
action length $L$ and laser intensity $I^{\text{in}}$, i.e.,
\[
\eta = \frac{P_{\text{fl}}}{I^{\text{in}} L^2}, \tag{B1}
\]
where $P_{\text{fl}}$ is the incident laser intensity. In this definition, the spin-flip probability is averaged over $I^{\text{in}} L^2$, because the transition amplitude should be proportional to $|E_{\text{in}} L|$ in the weak interaction regime, according to Eq. (A3). Even though the results in Fig. (3) are beyond the weak interaction regime, we can still try to apply Eq. (B1) to Fig. 3 and find that the corresponding spin-flip efficiency is $\eta \approx 5.5 \times 10^{-9} \text{W}^{-1}$.

As a next step, we chose a work [33] using the lowest incident laser intensity as an example to demonstrate that our results greatly improve the spin-flip efficiency. In Ref [33], regarding the spin-flip effect in the regular Kapitza-Dirac (KD) effect using two lasers of the same frequency, also referred to as the spin-Kapitza-Dirac (SKD) effect, the spin-flip efficiency is found to be very low. More precisely, for an electron velocity of $1 \times 10^7 \text{m/s}$ and an interaction length of 1 mm, an illumination intensity of $\sim 10^{14} \text{W/cm}^2$ finally results in a spin-flip probability of $1.3 \times 10^{-3}$. As a comparison, if their work chose the same interaction length of 16 µm and incident laser intensity of $8.5 \times 10^{12} \text{W/cm}^2$ as in Fig. 3, the spin-flip probability should be further reduced to
\[
P_{\text{fl}} \approx 1.3 \times 10^{-3} \times \left(\frac{16 [\mu m]}{1 [\text{mm}]^2}\right)^2 \times \frac{8.5 \times 10^{12} \text{W/cm}^2}{1.41 \times 10^{14} \text{W/cm}^2} \\
= 2.8 \times 10^{-8} \ll 12\%.
\]
The above comparison can also be made equivalently using the spin-flip probability we defined in Eq. (B1), which leads to a spin-flip efficiency of $\eta \approx 1.3 \times 10^{-13} \text{W}^{-1}$ for the SKD effect, relative to $\eta \approx 5.5 \times 10^{-9} \text{W}^{-1}$ in our study.

Also in Ref [33], a scheme is proposed to realize the electron spin flip based on the two-color Kapitza-Dirac (KD) effect, where the spin-flip efficiency is much higher than the SKD. According to Ref [33], for an electron velocity of $1 \times 10^7 \text{m/s}$ and an interaction length of 1 mm, an illumination intensity of $\sim 10^{13} \text{W/cm}^2$ results in a spin-flip probability of $7.4 \times 10^{-4}$. Compared with our result, the spin-flip efficiency is found to be $\eta \approx 7.4 \times 10^{-13} \text{W}^{-1}$, which is still smaller by several orders than work.

Appendix C: Three dimensional electron beam

In the main text, we consider a 2D Gaussian electron beam, which consists of the Fourier components of different wave vectors (in the $x$-$z$ plane) perpendicular to the transverse electric field $E_y$. For such a 2D electron beam, the spin-preserving PINEM scattering is absent, which can only be caused by the electric field parallel to the electron wave vector [see Eq. (A3)]. However, in a realistic experiment, the incident electron wave function should be three-dimensionally confined, with a Gaussian beam profile also in the $y$-$z$ plane, as illustrated in Fig. 4. Unlike the 2D situation, the 3D electron beam also includes Fourier components of the wave vectors that are not perpendicular to $E_y$. As illustrated in Fig. 4 for a Fourier plane wave of wave vector $k_e$, the electric field $E_y$ includes a component parallel to $k_e$, which can consequently give rise to PINEM scatterings. For simplification, in our discussions below we focus only on the Fourier components of wave vectors in the $y$-$z$ plane, i.e., $k_{e z} = 0$. In what follows, we compare the strengths of the spin-flip and PINEM interactions and reveal the critical beam waist when the spin-flip scattering is dominant.

1. Strength comparison: spin-flip vs PINEM interactions

To compare the interaction strengths of the spin-flip and PINEM scattering, we first consider a 2D electron beam along $+\hat{z}$ through an optical near field including electric field components of $E_y$ and $E_z$, as in the main text. We also focus on the spin flip along the $y$-direction, so the term $\mu_B \mathbf{B} \cdot \vec{\sigma}$ only results in the spin-flip scattering. In this configuration, according the corresponding matrix elements in Eq. (A3), the strength of the PINEM interaction induced by $E_z$ is characterized by $\chi_{\text{PINEM}} = |\epsilon_{\text{evu}} E_z/\omega|$. Likewise, for a large wave vector in the near field ($g \gg q$), the $x$- and $z$-components of magnetic field contribute equally, so the interaction strength of the spin-flip transition along the $y$-direction can be evaluated by $\chi_{\text{flip}} = |2 \mu_B g E_y|$, according to Eq. (3) in the main text.

Similarly, for the scenario of interaction between a near field including only $E_y$ and a Fourier component of a wave vector $k_e = (k_{e x}^0, k_{e z}^0)$ (see Fig. 1), the interaction strengths can be found equivalently. In this situation, the spin-flip and PINEM interaction strengths can be found by simply substituting $E_y$ and $E_z$ in $\chi_{\text{flip}}$ and $\chi_{\text{PINEM}}$, using $\chi_{E \parallel}$ and $\chi_{E \perp}$, i.e., the components perpendicular and parallel to $k_e$, respectively. Eventually, we can find the ratio
\[
\frac{\chi_{\text{flip}}}{\chi_{\text{PINEM}}} = \frac{g \epsilon_{\text{evu}}}{k_0 \epsilon_{\text{evu}}} = \frac{\hbar \omega}{m_e c^2} \left(\frac{v_0}{c}\right)^2 \cos(\theta_e) \sin(\theta_e).
\]
This equation again demonstrate that a slow electron is preferable to observe the proposed spin-flip effect, as explained in the main text. Intuitively, the factor $g/k_0$ in above equation reveals that the spin-flip transition becomes more prominent as the photonic scale approaches the that of the electron.

2. Critical Gaussian beam waist along the $y$-direction

For a transverse electric field $E_y$ ($E_z = 0$), we can find a certain critical angle $\theta_{er}$, at which the interaction strengths of the spin-flip and PINEM scattering
FIG. 4. Illustration of a three dimensional Gaussian electron beam propagating through the optical near field of a nanowire array, as in Fig. 1 in the main text.

are equal, i.e., $\chi_{\text{flip}}/\chi_{\text{PINEM}} = 1$. For the parameters adopted in Fig. 3, the corresponding critical angle is easily found to be 

$$\vartheta_{\text{cr}} \approx 0.24 \text{ [mrad]}.$$ 

Alternatively, for a 3D incident Gaussian electron beam, when measured at a far field angle $< \vartheta_{\text{cr}}$, one should observe stronger spin-flip effect than the spin-preserved PINEM effect.

We can also define a critical waist of the Gaussian beam in the $y$-direction, at which the electron beam is dominated by the spin-flip scattering. As shown in Fig. 4, the lateral diffraction angle of the beam in the $y$-z plane is $\vartheta = \lambda/\pi w_0^y$, where $w_0^y$ is the beam waist along the $y$-direction, $\lambda_e = 0.24 [\text{Å}]$ is the electron wave length corresponding to the velocity $v_0 = c/10$. When the diffraction angle of the electron beam coincides with the critical angle, $\vartheta = \vartheta_{\text{cr}}$, the spin-flip scattering significantly dominates over the PINEM interactions in the electron beam. For the parameters in Fig. 3, the corresponding critical waist is found to be $w_{0,\text{cr}}^y = \lambda_e/\pi \vartheta_{\text{cr}} = 32 [\text{nm}]$. By further increasing the waist $w_0^y$, the electron beam is more similar to a plane wave, and the corresponding spin dynamics should be more consistent with our results in the main text.

Appendix D: Supplemented calculations

FIG. 5. Same as Fig. 2(a) and (b) for the incident electron spin along $+\hat{x}$.

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FIG. 6. Same as Fig. 3(b)-(d) for the incident electron spin along $\hat{z}$. Different from Fig. 3(b), the spin-preserving electrons (a) are distributed on the states of energy $E_0$ and $E_0 \pm E_p$ [see Fig. 3(a)]. Likewise, the spin-preserving electrons (b) are distributed on the energy levels, $E_0 \pm E_p$ [see Fig. 2(b)].
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