

Ultrafast laser-induced spin-manipulation scenarios on a long zigzag silicon chain system

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(Received 17 July 2025; revised 6 May 2026; accepted 26 June 2026; published 17 August 2026)

Long-distance spin transfer is essential for realizing scalable, all-optical magnetic logic units. In this work, we employ high-level quantum chemistry methods to demonstrate ultrafast laser-driven spin dynamics in a zigzag silicon chain functionalized with two Ni atoms. The first result of this study is the realization of coherent spin transfer in both cis and trans configurations, achieving transfer distances of 6.547 and 6.021 nm, respectively, within the subpicosecond regime, demonstrating the potential of silicon-based spin channels for ultrafast logic operations. The second outcome reveals that even minor variations in laser polarization angles can cause a significant reduction in spin-transfer fidelity. Third, we find that changes in the strength of an applied inhomogeneous magnetic field B strongly affect the fidelity of spin transfer. The fourth key finding is that, for a double-pulse sequence, the fidelity of the spin-flip process is highly sensitive to the time delay between two laser pulses. Fifth, a combination of a single laser pulse and a magnetic field B enables the selective addressing of individual spin centers with high spatial resolution. In particular, the cis configuration exhibits a resolution limit for Ni-Ni separation of just 3.0 Å (well below the projected CMOS scaling limit of 20 Å), highlighting its potential for miniaturization.

DOI: [10.1103/kwq8-7tvl](https://doi.org/10.1103/kwq8-7tvl)

I. INTRODUCTION

The pursuit of enhanced computing performance drives the development of quantum-based systems [1–4], enabling advanced calculations and materials simulations. A critical step toward realizing such systems is the demonstration of quantum integrated circuits (ICs) [5–8]. Although the IC industry has advanced rapidly [9,10], with complementary metal-oxide semiconductor (CMOS) technology [11–13] remaining dominant, CMOS is nearing its physical limits. Quantum ICs, based on spintronics, offer new opportunities by using electron spin for information processing, transmission, and storage. The combination of magnetic materials with conductive elements has led to innovative system designs [14–17]. Recently, Liu *et al.* [18] proposed a microscopic mechanism for ultrafast long-range spin transfer in carbon chains with two nickel (Ni) atoms acting as spin carriers, bridged by conjugated p_z orbitals, using advanced quantum chemistry. The process is driven by virtual excitations between Ni and carbon

orbitals, enabling indirect coupling. Its efficiency depends on orbital phase alignment and degeneracy, while coherent superposition supports long-distance transfer even with asymmetric coupling. They also investigated local spin-flip addressability and ultrafast spin dynamics in carbon cross configurations with Ni atoms [19]. They used a Λ -type process, where the population is transferred from an initial state to a target state, to accomplish the spin dynamics. The specific nature of the process—such as spin-transfer or spin-flip—is dictated by the exact magnetic properties of the two states. In particular, when the spin density of the two states is localized on different atoms, the mechanism corresponds to a spin-transfer process (more details are shown below in Sec. II).

Besides, Li *et al.* [20,21] highlighted endohedral fullerenes as promising molecular spintronics candidates due to their long magnetic relaxation times. Barhoumi *et al.* further examined laser-induced spin manipulation in nonlinear zigzag carbon chains with Ni atoms [22]. These advances support the development of molecular spintronics and all-optical magnetic logic, which require materials with long spin coherence, semiconductor compatibility, and nanoscale spin control. Although carbon-based conjugated chains (e.g., graphene nanoribbons, polyenes) have been widely explored for spin transport, their limited integration with silicon electronics and the remaining challenges in achieving stable, long-distance spin transfer under optical excitation hinder their practical applicability. In parallel, the identification of materials that

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simultaneously provide the required logic functionality and can be seamlessly integrated into scalable spintronic heterostructures remains a central and unresolved challenge for spin-based device engineering. While spin-transfer processes have been successfully demonstrated between directly connected magnetic atoms or nearby atoms bridged by small ligands, extending these processes over long distances remains difficult, in part due to the high computational cost of quantum chemistry methods. In light of these challenges, Liu *et al.* [23] employed high-level *ab initio* calculations to demonstrate ultrafast laser-induced spin manipulation in a 40-atom zigzag carbon chain functionalized with two Ni atoms, achieving subpicosecond spin transfer over a distance of 4.428 nm, comparable to the 5 nm CMOS technology node in 2020. However, as CMOS technology continues to scale well below 4 nm [24], such distances become increasingly incompatible with advanced device architectures.

To address these limitations and to maintain compatibility with ongoing miniaturization while ensuring integration with silicon-based technology, we considerably extend this route to the investigation of the isoelectronic and technologically ubiquitously important element silicon (Si) using a 40-atom-long zigzag Si-chain system (one dimensional) with two magnetic centers [25–30] attached (as a prototypic device building block [31,32]) in the present work. We chose a zigzag Si chain from the silicene sheet, which connects the properties of silicon to those of graphene and represents an additional step in miniaturizing nanoelectronics. The zigzag chain [33] is selected for several reasons, e.g., (i) zigzag chains have been extensively studied in the context of spin transport in carbon-based systems [34–38], providing a direct benchmark for comparing the performance of silicon versus carbon. For example, this choice is strongly supported by the recently established dynamic Goodenough-Kanamori-Anderson (GKA) rules for ultrafast spin transfer [18]. This work systematically demonstrated that for laser-induced spin transfer over conjugated chains, the orbital symmetry and overlap dictated by the bridge geometry are paramount. It explicitly shows that a linear bridge is highly inefficient, while a bent, zigzag geometry like the one we use provides the necessary orbital phase alignment for strong, long-range dynamic magnetic coupling. (ii) They exhibit a nonzero density of states at the Fermi level due to edge states, which facilitates spin transport [39]. Moreover, the zigzag geometry allows for asymmetric attachment of the Ni atoms (as we have implemented), which promotes better spin localization on the magnetic centers—a crucial requirement for the Lambda (Λ) process [40,41] (see Fig. S1 of the Supplemental Material [42] and Refs. [43–45]).

It should be noted that silicene is the equivalent of graphene for silicon with a two-dimensional (2D) [46–52] structure. The atoms are arranged in a hexagonal honeycomb lattice, creating an atomic monolayer of silicon. The three kinds of silicene are: planar (pure sp^2), low-buckled (mixed between sp^2 and sp^3), and high-buckled (pure sp^3). The vertical separation between the two Si atom planes is known as the buckling parameter (Δ_0). Takeda and Shirashi [53] first reported this bending of Si in 1994. Silicene is similar to graphene when $\Delta_0 = 0$ Å. $\Delta_0 = 0.44$ Å means low buckling while it is considered high buckling when $\Delta_0 = 2.15$ Å [54]. The pseudo-Jahn-Teller (PJT) [55] distortion renders

two sublattices in the crystal lattice of every free-standing Xene (graphene, silicene, etc.) vertically shifted. This buckled crystal lattice is projected to appear on the atomic scale. Density functional theory (DFT) computations [56] indicate that buckled silicene is more stable than planar silicene [57,58]. This means Si favors sp^3 hybridization, whereas sp^2 hybridization is dominant in graphene. Thus, in contrast to planar graphene, planar silicene is not stable [58]. Nevertheless, the planar form of silicene can occur and the PJT effect can be efficiently suppressed by the existence of the substrate [59–63]. This has been shown in silicene prepared on Au(111) films produced on Si(111) and mica substrates [64]. Several two-dimensional monolayers of planar silicene were synthesized [65–68], where the structure was created using surface-segregated Au(111) thin films grown on a Si(111) substrate [69]. Hogan *et al.* [70] successfully reproduced the key experimental features and isolated the optical signal of the epitaxial silicene overlayer. Furthermore, Takahashi reported a strategy and design for realizing flat zigzag silicene nanoribbons. Through theoretical analysis of edge-substituted structures, she demonstrated that zigzag silicene nanoribbons incorporating beryllium bridges exhibit high stability in a planar configuration [71]. Using atomic-resolution scanning tunneling microscopy, Lalmi *et al.* [72] experimentally demonstrated the presence of a graphenelike silicene sheet epitaxially grown on the close-packed Ag(111) surface. A quasifreestanding silicene layer was reported, successfully obtained via oxidation of bilayer silicene on the Ag(111) surface [73]. A new 2D crystalline silica polymorph formed on Ru(0001) is presented and characterized using scanning tunneling microscopy (STM), low energy electron diffraction (LEED), infrared reflection absorption spectroscopy (IRAS), and x-ray photoelectron spectroscopy (XPS) measurements [74]. Silicon nanowire ensembles with vertical and zigzag architectures have been achieved via wet chemical etching of bulk silicon wafers [75]. Kopciuszynski and co-workers [76] investigated the electronic structure of an epitaxial planar silicene heterostructure grown on ultrathin gold films deposited on Si(111) substrates and capped with 0.1 monolayer of silver, antimony, and lead. Herein, the impact of submonolayer coverage on planar silicene was investigated through detailed angle-resolved photoemission spectroscopy (ARPES) measurements. Thus, the zigzag silicon chain considered here is not merely a theoretical abstraction, but is rooted in experimentally feasible structures. Also, planar silicene can exhibit a rich diagram of topological phases under an external magnetic field [77]. Silicene operates as an advantageous platform to manipulate also different spin-related effects. Investigating the spin degree of freedom of an electron allows even more functionalities to this material and thus novel spintronic devices [78–84].

In our present theoretical work, two Ni atoms are attached to the zigzag chain of silicon (chain taken from planar silicene) because Xenes (or even graphene) fail to exhibit strong magnetism due to the delocalization of s and p orbitals. Herein, we achieve ultrafast coherent spin transfer over distances of 6.547 nm (cis configuration) and 6.021 nm (trans configuration)—substantially exceeding the carbon-based benchmark. Notably, depending on the relative position of the two Ni atoms, the silicon chain structures can be divided

into two classes, namely *cis*- and *trans* configurations, which differ in the relative position of the two Ni atoms. In *cis* configurations, the Ni atoms are sitting on the same side of the silicon chain, while in *trans* configurations they are attached to opposite sides of the silicon chain. More importantly, we attain a spatial resolution limit for individually addressing spin centers of only 3.0 Å, which lies well below the projected CMOS scaling limit of 20 Å. Beyond these improved metrics, we uncover several previously unobserved effects inherent to the silicon platform, including a pronounced sensitivity of spin-transfer fidelity to laser polarization angles, a strong and nonmonotonic dependence on the strength of an applied inhomogeneous magnetic field, and a critical sensitivity of the fidelity to the interpulse time delay in double-pulse sequences. Collectively, these findings establish silicon-based zigzag chains as a promising and scalable platform for future all-optical magnetic logic circuits.

Based on the above information and motivated by the spin-dynamics of the zigzag carbon chain, intriguing magnetic properties of graphene [85,86], and the spin-dynamic processes on a zigzag carbon cross structure, we provide a theoretical concept for an integrated logic processor made up of an array of single molecular spin processing nodes connected by bridges.

The structure of this paper is as follows: Section II presents the computational methods and details. The laser-induced ultrafast spin dynamics, encompassing both spin-flip and spin-transfer processes, are analyzed and discussed in Sec. III. The main conclusions are summarized in Sec. IV.

II. COMPUTATIONAL DETAILS AND METHODS

In this paper, as a model of building blocks, we investigate the ultrafast laser-induced spin-manipulation processes such as spin-flip and spin-transfer on a zigzag silicon chain (a zigzag planar Si chain, one dimensional), using high-level quantum chemistry together with the Λ process (more details are shown in the Supplemental Material [42]). Consequently, we extend the successful and promising investigations of long-distance ultrafast spin-transfer over a zigzag C chain to this Si system for a conceivable integration strategy. To start with, we compute the molecular orbitals (MOs) of this system and the single-body Slater determinant, employing the Hartree-Fock (HF) method [87–90]. Coupled-cluster (CC), widely used in molecular quantum chemistry, is among the most successful post-Hartree-Fock methods. It provides an accurate treatment of electron correlation and generally outperforms approaches such as configuration interaction (CI) [91], showing excellent agreement with experiment [92–94]. Importantly, CC captures both static correlation effects, relevant for magnetism, and dynamic correlations, essential for optical properties [95]. The coupled-cluster singles and doubles (CCSD) [96–98] method is used to obtain the many-body wave function of the ground state. Subsequently, the equation-of-motion coupled-cluster method with single and double excitations (EOM-CCSD) [99,100] is employed to compute correlated excited states (singlets and triplets). These excited states are constructed on top of the CCSD reference state, enabling the description of strongly multiconfigurational wave functions within a single-reference framework and allowing

the evaluation of transition matrix elements using Slater-Condon rules [101].

We emphasize that while HF provides the initial orbitals, CCSD and EOM-CCSD systematically account for electron correlation effects through the exponential excitation operator, capturing both dynamic and nondynamic correlations. CCSD typically recovers most of the correlation energy for molecular systems of the size considered here, while EOM-CCSD provides a balanced treatment of ground and excited states, including open-shell and multiconfigurational character where present. The reliability of this approach is further supported by its successful application in previous ultrafast spin-dynamics investigations on carbon-based systems [19,22,23], as well as its reasonable agreement with experimental findings [102–104]. Thus, CCSD and EOM-CCSD provide a reliable and balanced description of the electronic correlations essential for the spin-flip and spin-transfer processes investigated in this work. Note that the CCSD and EOM-CCSD methods together with the 6-31G basis set [105], which is used for the silicon, hydrogen, and nickel atoms, are implemented in the GAMESS code [106].

These approaches have been successfully applied in the earlier investigation, and they are in reasonable agreement with a wide range of experimental findings [102–104].

We investigate the spin-dynamics (spin-flip and spin-transfer) processes on a zigzag silicon chain employing the Λ process, where an external femtosecond laser pulse [107–115] drives population transfer from an initial state to a target state via intermediate states (see Supplemental Material [42]). As previously mentioned in Sec. I, the nature of the process is determined by the spin-density localization: Spin-transfer occurs when the initial and target states are localized on different Ni atoms, while spin-flip occurs when they are on the same atom but with opposite spin orientations. Although the Λ process is a two-photon process, it is often much faster than direct excitation, which is typically optically restricted and therefore slow. Within the electric-dipole approximation, direct transitions between opposite spin states are forbidden. Therefore, spin-orbit coupling (SOC) is essential to induce spin-mixed intermediate states that couple the initial and final states with opposite spin directions. These spin-mixed states serve as bridges that enable the population transfer. More details about the Λ process are provided in the Supplemental Material [42].

In our case, SOC leads to spin mixing in the intermediate state. Furthermore, the presence of magnetic Ni atoms in the system induces spin polarization. The d orbitals of these metal centers contribute significantly to SOC, providing an additional justification for its inclusion in our analysis. For an accurate description of the initial and target states involved in a Λ process driven by an external laser pulse, a finite energy separation between these states is required. However, these states are typically degenerate or nearly degenerate (often corresponding to two substates originating from the same triplet state). Therefore, an external magnetic field B is applied to lift this degeneracy via Zeeman splitting. The direction of the magnetic field is set along the z axis (out of plane). Additionally, the magnetic field serves to define the direction of magnetization and produces the appropriate spin-mixed states that serve as intermediate states in the Λ process. SOC and

B, which are needed to achieve this process, are added to the Hamiltonian perturbation:

$$\hat{H}^{\text{SOC+B}} = \sum_{i=1}^N \frac{Z_a^{\text{eff}}}{2c^2 R_i^3} \hat{\mathbf{L}} \cdot \hat{\mathbf{S}} + \mu_L \hat{\mathbf{L}} \cdot \mathbf{B}_{\text{stat}} + \mu_S \hat{\mathbf{S}} \cdot \mathbf{B}_{\text{stat}}. \quad (1)$$

The spin angular momentum operators are represented by $\hat{\mathbf{L}}$ and $\hat{\mathbf{S}}$. The gyromagnetic ratios are μ_L and μ_S . The total spin angular momentum operator is the vector sum of the spin vectors of each single electron: $\hat{\mathbf{S}} = \sum_{i=1}^N \hat{\mathbf{s}}_i$. The total orbital angular momentum operator is $\hat{\mathbf{L}} = \sum_{i=1}^N \sum_a \hat{\mathbf{l}}_{i,a}$ (the angular momentum of the electron with respect to the nucleus of the atom). The sum “a” is over all nuclei in the system at positions R_i with effective charge Z_a^{eff} . n is the number of electrons, c is the speed of light, and R_i is the absolute value of the position of the i -th electron. A noticeable point is that the Breit-Pauli [116] Hamiltonian’s one-electron effects are typically the main ones. Since approximate effective nuclear charges Z_a^{eff} correctly compensate for the two-electron contributions up to the third row of the periodic table [117], we simply take into account the one-electron effects in this case. The difference between dominant and minority spins is arbitrary because, in the absence of a Zeeman splitting, the spin-up and spin-down states are degenerate, making any linear combination of them an eigenstate of the total Hamiltonian. Even after the incorporation of spin-orbit coupling, this issue still exists. The individual spin states are -1 ($|\downarrow\downarrow\rangle$), 0 ($\frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$), and 1 ($|\uparrow\uparrow\rangle$), which are responsible for the magnetic properties of a system. We identify easily distinguishable many-body states with various spin orientations or spin-density localization after the perturbative inclusion of SOC and a static B, which could be separately treated with a laser pulse.

$$\hat{H}'(t) = \hat{\mathbf{D}} \cdot \mathbf{E}_{\text{laser}}(t). \quad (2)$$

$\hat{\mathbf{D}}$ denotes the vector electric dipole-transition operator, while the laser electric field is written as $\mathbf{E}_{\text{laser}}(t) = E_{\text{laser}}(t) \mathbf{e}$, where $\mathbf{e}$ is the complex polarization vector. The time-dependent electric field of the laser pulse $E_{\text{laser}}(t)$ is taken to be sech^2 -shaped: $E_{\text{laser}}(t) = E_0 \text{sech}^2(\frac{t-b}{a}) \sin(\omega t)$, where E_0 is the amplitude, a and b are parameters representing the width and the center of the envelope, respectively, and ω is the carrier frequency. Note that, in a coordinate system where the pulse propagates along a direction specified by the spherical angles θ (polar angle) and ϕ (azimuthal angle), the polarization vector $\mathbf{e}$ can be written as $\mathbf{e} = \frac{1}{\sqrt{1+\beta^2}}(\hat{\mathbf{x}} + i\beta\hat{\mathbf{y}})$. Here, $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ are the unit vectors in the x and y directions, respectively, which define the plane perpendicular to the propagation direction. The parameter β is the ellipticity: $\beta = 0$ corresponds to linear polarization, $\beta = \pm 1$ to circular polarization, and intermediate values to elliptical polarization. The angle γ defines the orientation of the polarization ellipse relative to the optical plane.

In the interaction picture [118,119], a laser pulse generates an additional time-dependent perturbation that propels the population to the desired final many-body state. The dynamic perturbation can be expressed mathematically.

$$\frac{\partial c_n(t)}{\partial t} = \frac{-i}{\hbar} \sum_k \langle \Phi_n | \hat{H}'(t) | \Phi_k \rangle c_k(t) \exp(-i(E_k - E_n)t/\hbar). \quad (3)$$

The initial and final states are designated by n and k . Φ , received from CCSD and EOM-CCSD computations and changed with the impact of SOC and B, is the wave function of the many-body states. E and c represent the energy and population of the many-body states, respectively. We employ the fifth-order Runge-Kutta process with Cash-Karp adaptive step size [120] to solve these equations, and we employ a genetic algorithm for optimizing the laser pulse and successfully arrive at the ideal spin process. Notice that the laser parameters are optimized using a specially developed genetic algorithm [121]. Similar to numerical optimal control approaches [122], this method does not require any experimental input. The algorithm has been successfully applied to a variety of systems and is highly parallelized, incorporating analytical filters that eliminate inefficient laser-pulse configurations. The optimization involves eight parameters: the angles of incidence in spherical coordinates, θ (0° – 180°) and ϕ (0° – 360°); the angle γ between the light polarization and the optical plane; the ellipticity β ; the full width at half maximum (FWHM) of the laser pulse (0–500 fs); the laser energy E_{laser} ; the pulse amplitude; and the chirp, which varies between -5% and $+5\%$. In our genetic algorithm optimization, all eight parameters are treated as independent optimization variables, allowing the laser pulse to adapt its polarization state to maximize the fidelity of the desired spin process.

Within the genetic algorithm framework, each laser pulse is treated as an individual, with these eight parameters representing its genes. Individuals yielding higher fidelity are selected and propagated to subsequent generations through genetic operations such as crossover and mutation. This evolutionary process enables a global improvement of the parameters, allowing the most effective gene combinations to emerge. The fidelity is defined as the population of the target final state following interaction with the laser pulse. We regard a process with fidelity higher than 50% as successful.

Each generation consists of 200 individuals, and the optimization procedure typically converges within 200 generations. For the first generation, the genes are generated randomly with a random-number generator based on the nanosecond clock of the computer’s CPU. After inserting the parameters into Eq. (3), we obtain the fidelity of the Λ process induced by each individual.

III. RESULTS AND DISCUSSION

We show the top and side views of the geometric structure of planar silicene ($\Delta_0 = 0.0 \text{ \AA}$) in Fig. 1(a). As a building block model, we investigate the spin dynamics on a zigzag silicon chain, which contains 40 silicon, 40 hydrogen, and 2 nickel atoms, using the EOM-CCSD method together with the Λ processes. This structure [zigzag chain (1D)] allows the building of integrated circuits in which the chains wire the individual magnetic centers.

Next, we study the ultrafast-laser-induced spin-manipulation scenarios on two different geometries: the cis arrangement, where the Ni atoms are located on the same side of the chain [see Figs. 1(b) and 1(c)], and the trans arrangement, where they are positioned on opposite sides [see Figs. 1(d) and 1(e)]. For all systems, the total number of atoms is 82 (656 electrons), comprising three atomic species.

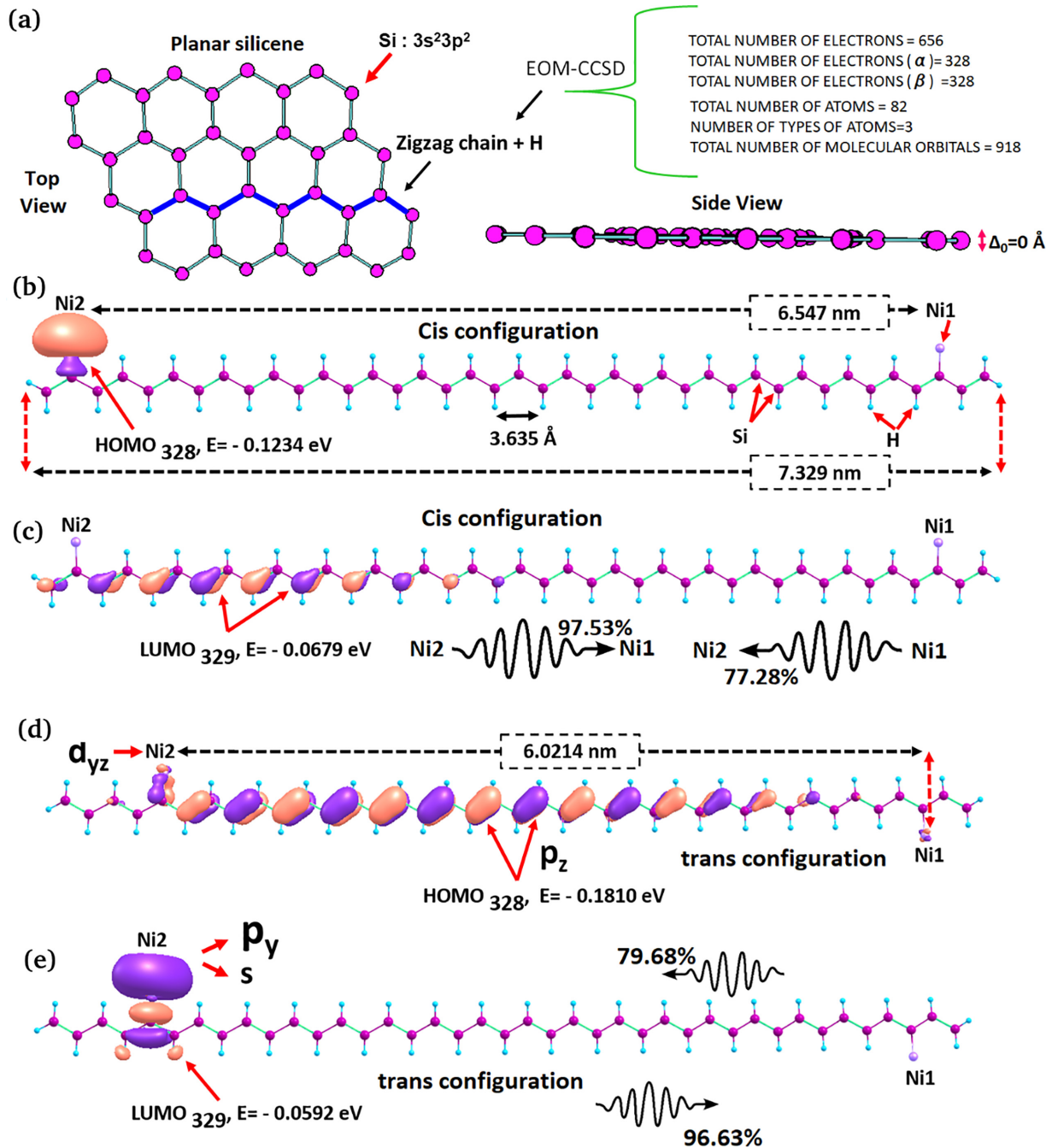


FIG. 1. (a) Top and side views of planar silicene, Δ_0 is the buckling parameter (the vertical separation between the two Si atoms planes). (b) HOMO, which is located on the Ni2 atom, of the cis configuration: s is the dominant atomic orbital in Ni2. (c) LUMO, which is located on some silicon atoms, of cis configuration: p_z is the dominant atomic orbital in Si atoms. (d) Trans configuration: HOMO (MO_{328}) is located on Si atoms: p_z and d_{yz} are the main atomic orbitals. (e) LUMO (MO_{329}) is located on Ni2 atoms: p_z are the main atomic orbitals. E is the energy obtained with EOM-CCSD of each MO. The main atomic orbitals in HOMO and LUMO are shown by the red and violet colors. The percentage numbers indicate the successful spin-transfer processes.

The Ni atoms are asymmetrically attached to the Si chain to enhance spin localization. Both configurations belong to the C_s symmetry point group. The Si-Si bond length in the planar zigzag silicene chain is $2.22 \text{ \AA}$, consistent with typical Si double bonds [123]. Although this bond length slightly decreases upon the addition of H and Ni atoms, it remains within the same range. In the cis configuration, the

distance from Ni1 to Ni2 is 6.547 nm , whereas in the trans configuration it is 6.021 nm ; in both cases, the total length of the Si chain is 7.329 nm . This difference, despite the identical overall chain length, arises directly from the distinct structural layouts. In the cis case, placing both Ni atoms on the same side of the zigzag backbone breaks the spatial symmetry between the two magnetic centers—a crucial requirement

because symmetric Ni coordination would delocalize the spin densities and prevent spin-transfer processes. The cis configuration thus provides a longer transfer distance (6.547 nm) while maintaining the necessary asymmetry for efficient spin dynamics. In the trans configuration, attaching the Ni atoms on opposite sides also breaks the symmetry, yielding a slightly shorter transfer distance of 6.021 nm. Nevertheless, this distance remains sufficiently large for coherent spin transfer, and the total length of the silicon chain stays unchanged at 7.329 nm. Both configurations successfully avoid symmetric Ni coordination and enable coherent spin transfer over technologically relevant distances.

It should be also noted that we consider alternative structural configurations beyond those presented here. In particular, we examine different binding sites for the Ni atoms on the silicon chain; the positions reported in this work correspond to the energetically most favorable adsorption geometries. Symmetric Ni coordination, where both Ni atoms are attached to the same two silicon atoms (i.e., sharing identical binding sites) or positioned symmetrically with respect to the chain axis, was tested but discarded because it results in delocalized spin densities that hinder the selective optical addressability of individual spin centers—a key requirement for the Lambda process. Different chain lengths were considered; however, increasing the chain length beyond 40 silicon atoms leads to a prohibitive increase in computational cost due to the $O(N^6)$ scaling of EOM-CCSD with system size (656 electrons already represent the practical limit for our computational resources). Conversely, shorter chains (e.g., 20 silicon atoms) yield Ni–Ni distances below 3 nm, which are no longer competitive with the projected CMOS scaling limits. The present 40-atom chain thus strikes an optimal balance between computational feasibility and technological relevance.

In Figs. 1(b) and 1(c), we also present the highest occupied molecular orbital (HOMO), and the lowest unoccupied molecular orbital (LUMO), which are necessary to accomplish the spin dynamics (in particular the spin-transfer processes) of the cis configuration. In this framework, HOMO (MO₃₂₈) is located at the Ni2 atom and the main atomic orbitals are s , d_{yy} , d_{zz} , and p_y . However, LUMO (MO₃₂₉) is located on some silicon atoms with energy -0.1234 eV and the main atomic orbital here is p_z [see Fig. 1(b)]. For the trans configuration, HOMO (MO₃₂₈) (p_z and d_{yz} are the main atomic orbitals on Si and Ni2 atoms, respectively) and LUMO (MO₃₂₉) (p_y and s are the dominant atomic orbitals on Ni2) are shown in Figs. 1(d) and 1(e). Although after including spin-orbit coupling (SOC) the spin is no longer mathematically a good quantum number, its expectation value $\langle S^2 \rangle$ in our system still allows us to loosely differentiate between “singlets” and “triplets”, as well as between spin-up (α) and spin-down (β) states. Several ultrafast spin dynamics processes, including spin-flip and spin-transfer scenarios, are carried out after the application of SOC and a static magnetic field and under different spin-density distribution requirements [23]. Mulliken population analysis on the reduced one-electron density matrix [124,125] determines the spin density on each many-body state. The spin density matrix of the n -th state can be given by

$$\mathbf{P}_n^{(\text{AO})} = \mathbf{C}_n^{(\text{AO})} \mathbf{S}^{(\text{AO})}. \quad (4)$$

$[\mathbf{S}^{(\text{AO})}]_{\nu\mu} = \langle \chi_\nu | \hat{S}_I | \chi_\mu \rangle$, ($I = x, y, z$) is the spin matrix of the atomic orbitals (AO) $|\chi_i\rangle$. The quantity $\sum_\mu [\mathbf{P}_n^{(\text{AO})}]_{\nu\mu}$ corresponds to the gross spin population associated with the ν -th atomic orbital in the n -th state. By summing over all orbitals belonging to a given atom, one obtains the spin density localized on that atom. A positive value indicates spin-up electrons (majority spin), while a negative value indicates spin-down electrons (minority spin). The matrix $\mathbf{C}_n^{(\text{AO})}$ represents the one-electron reduced density matrix of the n -th state in the atomic-orbital basis.

Our calculations show that the states of the cis configuration fulfill the requirement (see Fig. S2 of the Supplemental Material [42]): The spin density is highly localized on Ni1 with an absolute value of 1.113 (state $E_{|36\rangle} = 2.024\,967\,69$ eV with spin direction $\langle S_z \rangle = 0.220\,323\,08$), and the spin density on Ni2 is only slightly smaller with an absolute value of 1.097 (state $|68\rangle$ with energy $E_{|68\rangle} = 3.321\,132\,21$ eV). The rest of the spin density is mainly distributed on silicon atoms. In this configuration, the optimal magnetic field is 1.00×10^{-8} a.u. At this specific field strength, the Zeeman splitting is just sufficient to break the degeneracy between α and β states while still allowing the formation of well-defined spin-mixed states that enable efficient population transfer. If the magnetic field is weaker than this value, the spin states remain nearly degenerate, making it impossible to selectively address individual spin states. If the magnetic field is stronger, the Zeeman splitting becomes too large, which overly purifies the spin states and suppresses the spin mixing that is essential for the Λ process.

In addition, the states of the trans configuration fulfill (see Fig. S3 of the Supplemental Material [42]) the above conditions, and the spin density is highly localized on Ni1 with an absolute value of 0.977 (state $|63 \uparrow\rangle$ with energy $E_{|63 \uparrow\rangle} = 2.741\,897\,73$ eV and $\langle S_z \rangle = 0.965\,739\,89$, see also Table I for more details).

At the EOM-CCSD level, the cis configuration exhibits a singlet ground state ($\langle S_z \rangle = 0$ with zero spin density on both Ni atoms), whereas the trans configuration is characterized by a triplet ground state ($\langle S_z \rangle \neq 0$ with nonzero spin densities of the same sign on both Ni atoms). This difference in ground-state multiplicity can be understood from ligand field theory and the coupling between the two Ni centers. From a ligand field perspective, each Ni center is coordinated to one silicon atom of the zigzag chain. In the cis configuration, both Ni atoms are attached to the same side of the chain, creating a stronger and more symmetric ligand field due to efficient overlap between the Ni $3d$ orbitals and the silicon p_z orbitals. This strong ligand field produces a large crystal field splitting, favoring a low-spin configuration where electrons pair up in the lower d orbitals. Additionally, the strong orbital overlap facilitates antiferromagnetic superexchange coupling through the conjugated silicon backbone, aligning the unpaired electrons on the two Ni centers in opposite directions ($\uparrow\downarrow$). Consequently, the total spin is $S = 0$, yielding a singlet ground state ($2S + 1 = 1$). In contrast, the trans configuration has the Ni atoms attached on opposite sides of the chain, resulting in a weaker and more distorted ligand field. The smaller crystal field splitting cannot overcome the electron-electron repulsion (Hund’s rule), favoring a high-spin configuration with

TABLE I. Spin density, energies, and expectation values of the spin angular momentum elements for the spin-flip and spin-transfer scenarios on the cis configuration for the appropriate many-body states (some states) before and after inclusion of spin-orbit coupling and the magnetic field. The sign of $\langle S_x \rangle$, $\langle S_y \rangle$, and $\langle S_z \rangle$ signifies the spin direction: The positive and negative values are spin-up $\alpha \uparrow$ and spin-down $\beta \downarrow$, respectively. All calculations are performed with EOM-CCSD method.

System	States	Spin density before SOC and B		Energy, spins directions and spin density after SOC and B					
		Ni1	Ni2	E (eV)	$\langle S_x \rangle$	$\langle S_y \rangle$	$\langle S_z \rangle$	Ni1	Ni2
Cis ^c	68)	1.04900000	0.02000000	3.32113221	-0.00000014	-0.00000781	0.00000000	0.000	1.097 ^a
	67)	0.00000000	0.00000000	3.32113107	0.00000018	0.00000987	0.37911312	0.000	0.000
	60)	0.00000000	0.00000000	3.06510595	0.00001850	0.00000128	-0.88315657	-0.001	-0.141
	53)	0.00000000	0.00000000	2.82332165	-0.00000360	-0.00005086	-0.97956628	-1.049	-0.020
	36)	0.00000000	0.00000000	2.02496769	0.00001255	0.00001318	0.22032308	1.113 ^b	0.024
	32)	-0.00100000	-0.14100000	2.01056283	0.00000992	0.00000053	0.14783228	0.000	0.813
	30)	0.00000000	0.00000000	2.01055546	0.00992510	0.00065662	-0.14782925	0.000	-0.813
	29)	-0.00100000	-0.09600000	1.88252351	-0.00006080	-0.00002497	0.91747329	0.000	0.123
	27)	0.00100000	-0.00100000	1.88252232	-0.00008054	-0.00002563	-0.91747330	0.000	-0.123
	23)	0.00100000	0.13200000	1.68224586	-0.00007132	-0.00007090	-0.99422343	-0.001	-0.111
	22)	0.00000000	0.00000000	1.54522634	0.00001842	-0.00036836	0.20537673	1.056	0.054
	20)	0.00000000	0.00000000	1.54522106	0.00000028	-0.00000581	-0.20537757	-1.056	-0.054
	4)	0.00000000	0.00700000	0.01456761	0.00009251	0.00000799	0.56503934	1.099	0.031
	2)	0.00000000	-0.00700000	0.01456568	0.00077694	0.00006550	-0.56503938	-1.099	-0.031
	1) ^d	0.00000000	0.00000000	0.00000000	0.00000000	0.00000000	0.00000000	0.000	0.000
	63)	0.00000000	0.00000000	2.74189773	0.00000036	-0.00000505	0.96573989	0.977 ^e	0.017
	58)	-0.62100000	-0.02900000	2.56176929	-0.00002054	-0.00000392	-0.99963410	-0.285	-0.069
	19)	0.00000000	0.00000000	1.21987177	0.00004968	-0.00000180	-0.99999110	-0.090	-0.029
	16)	0.07700000	0.08500000	1.01925092	0.00002728	-0.00000304	-0.99997528	-0.119	-0.069
Trans ^f	10)	0.11900000	0.06900000	0.42775858	-0.00003454	-0.00000124	0.99718898	0.890	0.018
	8)	-0.11900000	-0.06900000	0.42764904	0.00005344	0.00000473	-0.99718998	-0.890	-0.018
	3)	0.00000000	0.00000000	0.00018865	0.00047754	-0.00003061	0.57369758	0.026	0.127
	1) ^g	0.00000000	0.00000000	0.00000000	-0.04765866	0.00134327	-0.57333007	-0.026	-0.127

^aThe highest spin density value on the Ni2 atom.

^bThe highest spin density value on the Ni1 atom.

^cThe magnetic field strength used here is $B = 1.00 \times 10^{-8}$ a.u.

^dThe ground state is a singlet.

^eThe highest spin density value on the Ni1 atom.

^fThe magnetic field strength used here is $B = 1.00 \times 10^{-6}$ a.u.

^gThe ground state is a triplet.

unpaired electrons. The opposite-side attachment also reduces orbital overlap and weakens the exchange coupling, allowing the unpaired electrons on the two Ni centers to remain aligned in the same direction ($\uparrow\uparrow$) or uncoupled, resulting in a total spin $S = 1$ and a triplet ground state ($2S + 1 = 3$). Thus, the singlet in cis and triplet in trans arise from the combined effects of ligand field strength (crystal field splitting) and exchange coupling mediated by the silicon backbone, both dictated by the attachment geometry.

A noteworthy point is that, for both configurations at their respective optimal magnetic fields ($B = 1.00 \times 10^{-8}$ a.u. for cis and $B = 1.00 \times 10^{-6}$ a.u. for trans), when calculating the expectation values $\langle S_z \rangle$ and $\langle L_z \rangle$, we find that the expectation value $\langle L \rangle$ is generally small (where $\langle L_z \rangle$ is approximately two orders of magnitude smaller than $\langle S_z \rangle$) due to the partial quenching of orbital angular momentum in the Ni atoms coordinated to the silicon chain. This partial quenching arises from the crystal field of the surrounding silicon atoms, which lifts the degeneracy of the Ni d orbitals. It is further enhanced by the large Ni-Ni distance and the extended π -conjugation along the silicon backbone. Nevertheless, the residual orbital

angular momentum, though small, is essential for generating spin-orbit coupling and the subsequent spin-mixed intermediate states required for the Λ process.

Moreover, in our systems, the spin direction is preferentially aligned along the z axis due to the combined effects of SOC and the applied external magnetic field. The anisotropy is predominantly uniaxial, originating from the symmetry of the zigzag silicon chain and the asymmetric attachment of the two Ni atoms. To quantify the energy barrier for spin rotation, we compute the energy difference between the spin aligned along the easy axis (z direction) and the hard axis (perpendicular direction, e.g., x or y). The magnetic anisotropy energy (MAE) for Ni-based systems has been extensively studied in the literature. For single Ni atoms bonded to graphene, MAE values as high as 53 meV have been reported [126]. For Ni clusters and organometallic complexes, MAE values typically range from 8 to 16 meV [127,128]. More generally, theoretical studies indicate that MAE is typically on the order of meV per atom and depends critically on the local atomic environment, orbital hybridization, and spin-orbit coupling strength [129]. In our zigzag silicon chain system with two Ni atoms, the uniaxial

TABLE II. Optimized parameters (ΔE , θ , ϕ , γ , FWHM, E_{laser} , and chirp) of the laser pulse for each spin-transfer and spin-flip process on the cis and trans systems. All calculations are performed at the EOM-CCSD level.

System	Scenario	Spin localized	Process	Fidelity (%)	ΔE (eV)	θ (deg)	ϕ (deg)	γ (deg)	FWHM (fs)	E_{laser} (eV)	chirp
Cis	spin-flip local	Ni1	$ 4\rangle \rightarrow 2\rangle$	91.08	1.93×10^{-6}	87.62	141.23	256.51	264.61	2.911	1.027
	spin-flip local	Ni1	$ 20\rangle \rightarrow 22\rangle$	88.19	5.28×10^{-6}	84.41	150.53	240.08	116.37	2.079	1.0204
	spin-flip local	Ni1	$ 22\rangle \rightarrow 20\rangle$	91.63	5.28×10^{-6}	87.66	141.23	295.82	264.61	0.022	1.047
	spin-flip local	Ni2	$ 30\rangle \rightarrow 32\rangle$	38.64	8.23×10^{-6}	269.44	298.65	223.03	498.64	1.379	1.036
	spin-flip local	Ni2	$ 32\rangle \rightarrow 30\rangle$	69.95	8.23×10^{-6}	273.70	230.88	19.07	250.81	0.804	1.003
	spin-flip local	Ni2	$ 29\rangle \rightarrow 27\rangle$	94.57	1.19×10^{-6}	78.22	52.95	37.29	295.87	2.452	1.049
	spin-transfer	Ni1 $\rightarrow$ Ni2	$ 53\rangle \rightarrow 60\rangle$	77.28	0.24178430	282.92	181.54	241.70	375.03	0.010	1.043
	spin-transfer	Ni2 $\rightarrow$ Ni1	$ 60\rangle \rightarrow 53\rangle$	97.53	0.24178430	273.93	312.52	335.62	471.72	1.299	0.992
Trans	local spin-flip	Ni1	$ 1\rangle \rightarrow 3\rangle$	69.17	1.8865×10^{-4}	52.93	267.70	220.60	92.54	2.899	0.981
	spin-transfer	Ni2 $\rightarrow$ Ni1	$ 16\rangle \rightarrow 1\rangle$	96.63	1.01925092	88.60	186.20	63.19	330.71	1.439	0.968
	spin-transfer	Ni1 $\rightarrow$ Ni2	$ 1\rangle \rightarrow 16\rangle$	79.68	1.01925092	266.75	320.07	115.48	384.60	1.417	1.034
	spin-transfer	Ni2 $\rightarrow$ Ni1	$ 1\rangle \rightarrow 8\rangle$	71.57	0.42764904	121.36	211.19	29.79	447.47	2.956	0.954
	spin-transfer	Ni2 $\rightarrow$ Ni1	$ 3\rangle \rightarrow 10\rangle$	74.62	0.42756993	45.40	302.78	214.68	405.51	2.625	0.970

MAE for the cis configuration is found to be approximately 15 meV per Ni atom (at vanishingly small external magnetic field), meaning that the energy cost to rotate the spin of a single Ni atom from the z axis to a perpendicular direction is 15 meV in the absence of an applied field. For the entire cis system with two Ni atoms, the total anisotropy energy is approximately 30 meV. For the trans configuration, the zero-field MAE is estimated to be approximately 13 meV per Ni atom (total 26 meV), reflecting the weaker ligand field and reduced orbital overlap due to the opposite-side attachment of the Ni atoms. These values are comparable to the thermal energy at room temperature (25 meV), indicating that the spin configuration is reasonably stable against thermal fluctuations at room temperature.

According to the spin-density distribution of the cis configuration presented in Table I (along with the spin densities, energies, and expectation values of the spin angular momentum components for other many-body states listed in Table S1 of the Supplemental Material [42]), various spin-flip and spin-transfer scenarios, for example, Fig. S4 (spin-flip process $|2\downarrow\rangle \rightarrow |4\uparrow\rangle$), Fig. S5 (spin-transfer process $|53\downarrow\rangle \rightarrow |60\downarrow\rangle$), and Fig. S6 of the Supplemental Material [42]), which are shown in Table II, are performed. In this configuration, the singlet ground state has zero spin density on both Ni atoms, preventing spin-transfer from the ground state. Hence, we use excited states with localized spin density as initial states. The excited states we selected have sufficiently long lifetimes to allow the laser pulse to drive the transfer before relaxation occurs. Moreover, these states are accessible via optical excitation from lower-lying states. For instance, we select the states $|60\downarrow\rangle$ and $|53\downarrow\rangle$ as initial and target states for the spin-down spin-transfer from Ni2 to Ni1 (see Fig. 2). The energy difference between the two states ΔE is 0.241 784 3 eV, which is considered to be large enough to make the two states distinguishable for an external laser pulse and at the same time sufficiently small to slow down and thus suppress the direct relaxation processes [119]. Notice that when the energy difference ΔE between the initial and target states is sufficiently small, the direct relaxation processes (such as phonon-mediated decay or radiative recombination) are slowed down because the density of available phonon states

or electromagnetic modes at that energy is low. Conversely, if ΔE is too large, the states become energetically distinguishable and the laser pulse can no longer efficiently drive the transition. An intermediate ΔE (as in our case, 0.2418 eV) strikes a balance: It is large enough to allow selective optical addressing but small enough to suppress fast relaxation pathways.

In this process $|60\downarrow\rangle \rightarrow |53\uparrow\rangle$, the fidelity reaches 97.53%, which can be considered a successful scenario. Also, this high fidelity is attributed to the favorable alignment of the p_z orbitals along the silicon chain, which facilitates efficient virtual excitations between the Ni atoms and the silicon backbone. Two major intermediate states ($|2\rangle$ and $|23\rangle$) are

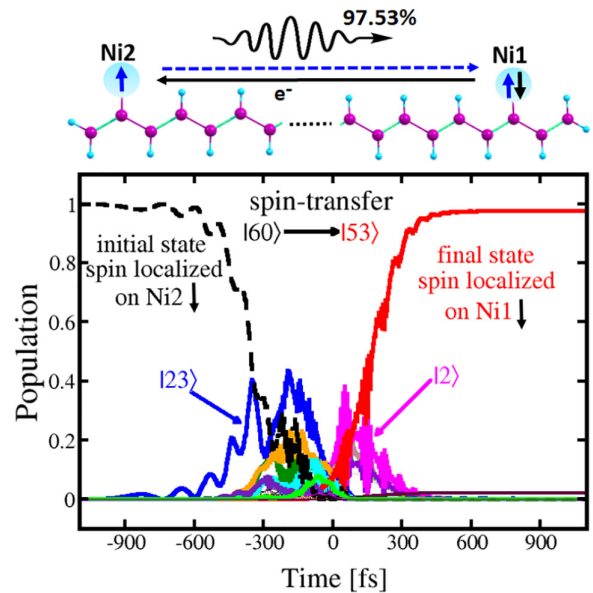


FIG. 2. The cis configuration: Spin-down spin-transfer process from the excited state $|60\downarrow\rangle$ (spin density localized on Ni2) to the target state $|53\uparrow\rangle$ (spin density localized on Ni1). The time-dependent population of the initial (dashed black line), target (solid red line), and intermediate states (solid lines in different colors) is shown. Arrows $\uparrow$ and $\downarrow$ indicate spin-up and spin-down, respectively.

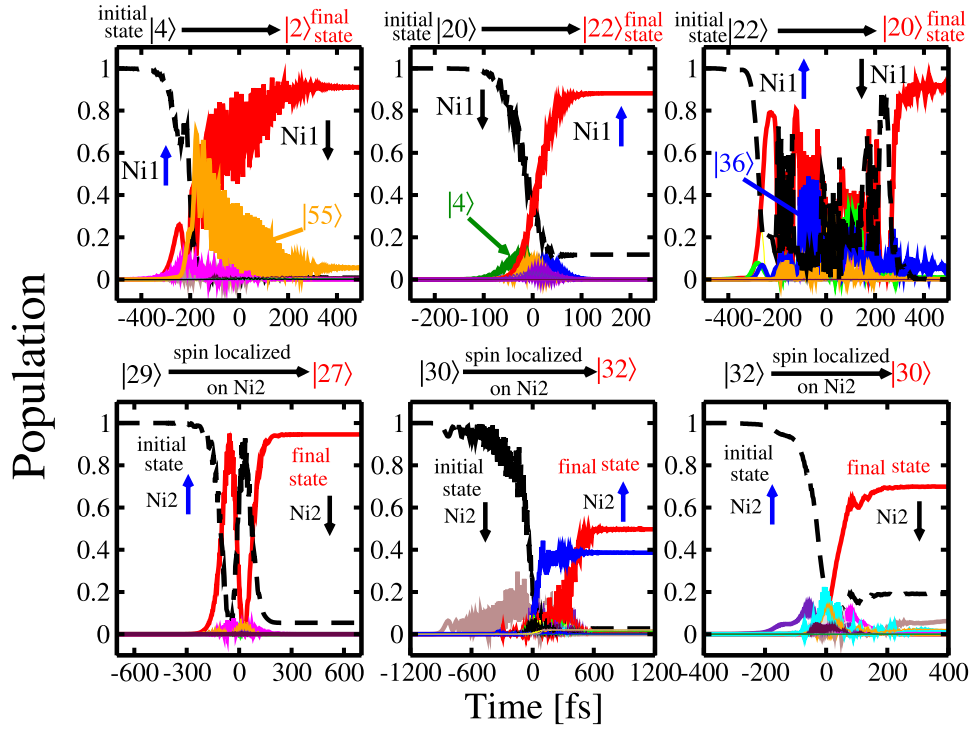


FIG. 3. Cis configuration: Spin-flip scenarios achieved on Ni1 ($|4 \uparrow \rangle \rightarrow |2 \downarrow \rangle$), Ni1 ($|20 \downarrow \rangle \rightarrow |22 \uparrow \rangle$), Ni1 ($|22 \uparrow \rangle \rightarrow |20 \downarrow \rangle$), Ni2 ($|29 \uparrow \rangle \rightarrow |27 \downarrow \rangle$), Ni2 ($|30 \downarrow \rangle \rightarrow |32 \uparrow \rangle$), and Ni2 ($|32 \uparrow \rangle \rightarrow |30 \downarrow \rangle$), respectively. For each process, the time-dependent population of the initial (dashed black line), target (solid red line), and intermediate (solid lines in different colors) states is presented. Notice that the two arrows $\downarrow$ and $\uparrow$ denote the spin direction of the corresponding states ($\alpha \uparrow$ and $\beta \downarrow$).

included and a few clear-cut Rabi oscillations [130,131] are seen in this process. The first major finding of this study is thus that a spin transfer between Ni atoms can be achieved over a distance of approximately 6.547 nm using a silicon-based channel system.

Other spin-flip processes (such as $|4 \uparrow \rangle \rightarrow |2 \downarrow \rangle$, $|20 \downarrow \rangle \rightarrow |22 \uparrow \rangle$, $(|22 \uparrow \rangle \rightarrow |20 \downarrow \rangle)$, $|29 \uparrow \rangle \rightarrow |27 \downarrow \rangle$, $|30 \downarrow \rangle \rightarrow |32 \uparrow \rangle$, and $|32 \uparrow \rangle \rightarrow |30 \downarrow \rangle$) are also achieved on the cis configuration. We show their corresponding time-dependent population of the initial (dashed black line), target (solid red line), and intermediate (solid lines in different colors) states in Fig. 3. Their optimized parameters of the laser pulses are shown in Table II.

Furthermore, we compute the spin-density distribution for the trans configuration, as presented in Table I. Here the optimal magnetic field employed is 1.00×10^{-6} a.u., which is two orders of magnitude higher than that used in the cis configuration. The optimal magnetic field differs between cis and trans configurations because the field must strike a delicate balance: It must be strong enough to lift the degeneracy between spin-up and spin-down states and to produce the necessary spin-mixed intermediate states for the Lambda process, but not so strong that it overly purifies the spin states and destroys the coherent mixing required for the transition.

By employing suitably tailored time-resolved laser pulses, distinct ultrafast spin-dynamics processes—such as spin-flip and spin-transfer—are also achieved in the trans configuration. For example, we show the spin-down spin-transfer

process $|16 \downarrow \rangle \rightarrow |1 \downarrow \rangle$ in Fig. 4. We take the states $|16 \downarrow \rangle$ (spin density is localized on Ni1 with energy $E_{|16 \downarrow \rangle} = 1.019\,250\,92$ eV) and $|1 \downarrow \rangle$ (spin density is localized on Ni2 with energy $E_{|1 \downarrow \rangle} = 0.0000$ eV) as the initial and target states, respectively. The energy difference of 1.019 250 92 eV is sufficient to distinguish the two states for an external laser pulse while suppressing direct relaxation mechanisms, thereby extending the lifetime of the higher-energy state. The fidelity of this scenario reaches 96.63%, which is considered a complete scenario. That means the spin density can be easily moved by 6.0214 nm from Ni2($|16 \downarrow \rangle$) to Ni1($|1 \downarrow \rangle$) in the trans configuration. The reverse spin-down transfer process, $|1 \downarrow \rangle \rightarrow |16 \downarrow \rangle$, is also accomplished. We find that the spin density is moving from Ni2 to Ni1 under a laser pulse and the fidelity reaches 79.68%. Note that the optimized parameters of the laser pulse are: $\theta = 266.75^\circ$, $\phi = 320.07^\circ$, $\gamma = 115.48^\circ$, $\Delta E = 1.019\,250\,92$ eV, $E_{\text{laser}} = 1.417$ eV, and chirp = 1.034 (see Table II for further details).

Besides, we show some of the time-dependent population of the states in Fig. 5. Their optimized parameters of the laser pulses are shown in Table II. For example, in the ground state, we take the states $|1 \downarrow \rangle$ and $|3 \uparrow \rangle$ as the initial and final states, respectively. The spin density is localized on the Ni2 atom for both states, which stem from the same triplet state. The fidelity of this process is 79.17% and the optimized parameters of the laser pulse are $\theta = 52.93^\circ$, $\phi = 267.70^\circ$, and the laser energy = 2.899 eV. In parallel, we show other three spin-transfer processes in Fig. 5. In the spin-down spin-transfer process $|1 \downarrow \rangle \rightarrow |8 \downarrow \rangle$, we take $|1 \downarrow \rangle$ (spin density is localized on

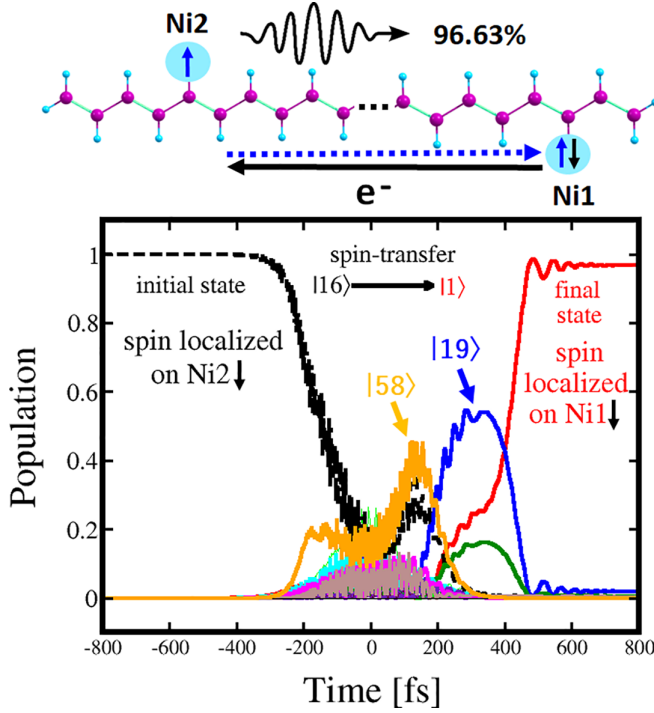


FIG. 4. The trans configuration: Spin-down spin-transfer process $|16\downarrow\rangle \rightarrow |1\downarrow\rangle$. The $\text{Ni2} \rightarrow \text{Ni1}$ process fidelity reaches 96.63%. The time-dependent population of the initial (dashed black line), final or target (solid red line), and intermediate (solid lines in different colors) states. Arrows $\downarrow$ indicate the spin-down spin transfer scenario ($|\beta\downarrow\rangle$). $|19\rangle$ and $|58\rangle$ are the major intermediate states.

Ni2 with energy $E_{|1\downarrow\rangle} = 0.00 \text{ eV}$ and $|8\downarrow\rangle$ (spin density is localized on Ni1 with energy $E_{|8\downarrow\rangle} = 0.427\,649\,04 \text{ eV}$) as the

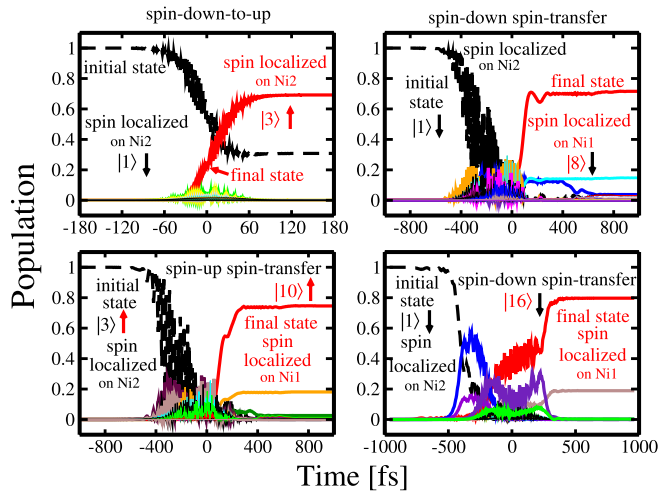


FIG. 5. Trans configuration: Spin-flip and spin-transfer scenarios are achieved on: $\text{Ni2}(|1\downarrow\rangle \rightarrow |3\uparrow\rangle)$ spin-flip), $\text{Ni2} \rightarrow \text{Ni1}(|1\downarrow\rangle \rightarrow |8\downarrow\rangle)$ spin-down spin transfer), $\text{Ni2} \rightarrow \text{Ni1}(|3\uparrow\rangle \rightarrow |10\uparrow\rangle)$ spin-up spin-transfer), $\text{Ni2} \rightarrow \text{Ni1}(|1\downarrow\rangle \rightarrow |16\downarrow\rangle)$ spin-down spin-transfer), respectively. For each process, the time-dependent population of the initial (dashed black line), target (solid red line), and intermediate (solid lines in different colors) states is presented. Arrows $\uparrow$ and $\downarrow$ indicate the spin-up ($|\alpha\uparrow\rangle$) and spin-down ($|\beta\downarrow\rangle$), respectively.

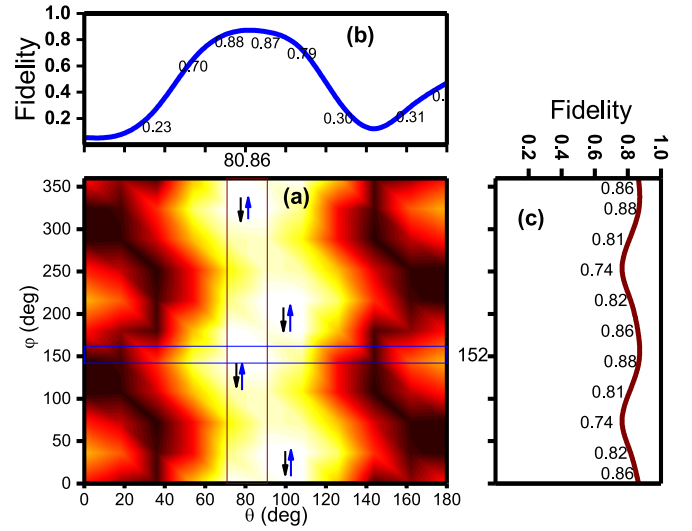


FIG. 6. (a) Influence of the polar angle θ and azimuthal angle ϕ of the laser pulse on the ultrafast spin-flip transition $|20\downarrow\rangle \rightarrow |22\uparrow\rangle$ in the cis configuration, evaluated over the full angular range $[0^\circ, 360^\circ]$. The framed region in (a) is analyzed in more detail in (b) and (c): in (b), θ is varied while ϕ is fixed at 152° ; in (c), ϕ is varied from 0° to 360° while θ is fixed at 80.86° . The maximum fidelity of approximately 90% is achieved at $\theta = 80.86^\circ$ and $\phi = 152^\circ$. Black $\downarrow$ and blue $\uparrow$ arrows represent spin-down and spin-up states, respectively.

initial and target states, respectively. We obtain the fidelity of this scenario is $\sim 71.57\%$ and the time-dependent population of the states (initial, final, and intermediate) is depicted in Fig. 5. In the spin-up channel, we can transfer the spin density from Ni2 to Ni1 using the states $|3\uparrow\rangle$ and $|10\uparrow\rangle$ as initial and final states, respectively. The energy difference between the states is $0.427\,569\,93 \text{ eV}$ and the fidelity of this process is 74.62% .

Given that the configurations exhibit anisotropic magnetic properties, the application of laser pulses along different directions is, in principle, expected to produce significant variations in the spin-dynamic processes. Thus, we investigate the influence of the laser pulse on the spin-flip processes and present the effect of the polar angle θ and the azimuthal angle ϕ of the laser pulse on the spin-flip process $|20\downarrow\rangle \rightarrow |22\uparrow\rangle$ in the cis configuration by varying them within the full angle range $[0^\circ, 360^\circ]$ in Fig. 6. Note that the laser pulse exhibits elliptical polarization in cis and trans configurations. It can be seen that the part of the map for $\phi \in [180^\circ, 360^\circ]$ essentially reproduces the structure of the domain $\phi \in [0^\circ, 180^\circ]$ by a simple translation. Thus, it is sufficient to focus on the stripe $0^\circ < \phi < 180^\circ$. Within this stripe, the pattern exhibits an approximate inversion symmetry with respect to the point $(\theta, \phi) = (90^\circ, 90^\circ)$, i.e., $F(\theta, \phi) \approx F(180^\circ - \theta, 180^\circ - \phi)$. This inversion-type symmetry captures the dominant behavior of the fidelity landscape in the primary angular domain.

Also, the magnitude of the magnetic field is maintained at $1.00 \times 10^{-8} \text{ a.u.}$, and optimized parameters such as γ° , β° , FWHM, laser energy, chirp, and amplitude of the laser pulses are fixed. In the range $70^\circ < \theta < 115^\circ$ and $\phi \in [0^\circ, 360^\circ]$, we see a wide tolerance area in which the fidelity value

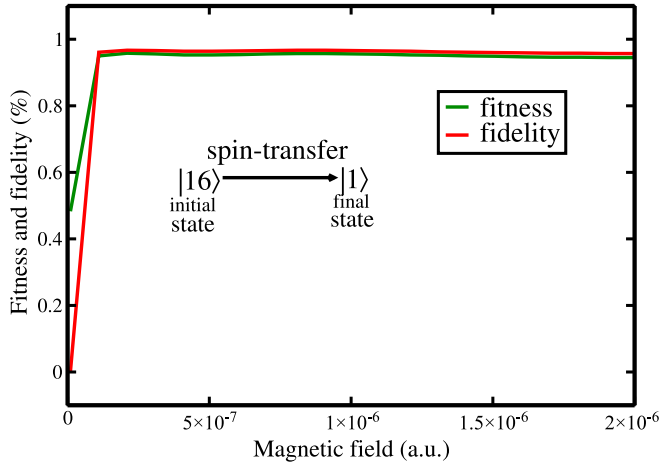


FIG. 7. Fidelity and fitness of the spin β spin-transfer process $|16\downarrow\rangle \rightarrow |1\downarrow\rangle$ on the trans configuration as a function of the magnetic field. The 0.0% fidelity (or fitness) at around $B = 0$ a.u. is simply due to the fact that α and β states are indistinguishable at this magnetic field.

is more than 80%. In the small frame part [see Fig. 6(a)], which extends from $\theta = 70.78^\circ$ to $\theta = 90.76^\circ$ and from $\phi = 141.26^\circ$ to $\phi = 191.90^\circ$, we obtain a fidelity of more than 88%, as shown in the linescans of Figs. 6(b) and 6(c). In addition to these plateaus, we see that even a slight modification in θ or ϕ can sometimes lead to a significant decrease in fidelity (see Figs. S7–S11 in the Supplemental Material [42]). This serves as the second key outcome of our investigation.

Additionally, we study the impact of an inhomogeneous magnetic field B on the spin-flip and spin-transfer scenarios. As the third result, an important outcome is that changing the magnetic field influences the fidelity of the laser-driven ultrafast spin manipulations. For instance, we see that the fidelity of the spin-down spin-transfer process $|16\downarrow\rangle \rightarrow |1\downarrow\rangle$ on the trans configuration quickly increases with B varying from 0 to 1.00×10^{-7} a.u., and reaches its peak at this magnetic field strength. When the magnetic field B becomes stronger than 1.00×10^{-7} a.u., the fidelity and the fitness of this spin-transfer process are preserved (see Fig. 7). This is because the majority of the intermediate states are pure spin states, which are hardly sensitive to changes in the external magnetic field. Notice that a parameter fitness is used to evaluate the quality of the corresponding Λ process (see Fig. S12 of the Supplemental Material [42]) where the fitness $f = p^2 - 0.7p_{\max}(1 - p)$. p is the fidelity that characterizes the population of the final state under the laser pulse, $p_{\max}$ is the maximum population of the final state.

Moreover, we study the double laser pulse-induced spin-down-to-up spin-flip and spin-down spin-transfer processes on the Ni atoms attached to the zigzag silicon chain. For a double pulse, we use two identical laser pulses with a specified phase relation at different times [23,44,45]. Equation (3) becomes in this case:

$$\frac{\partial c_n(t)}{\partial t} = \frac{-i}{\hbar} \sum_k \langle \Phi_n | [\hat{H}_{\text{int1}}(t) + \hat{H}_{\text{int2}}(t + \delta t)] | \Phi_k \rangle c_k(t) \times \exp(-i(E_k - E_n)t/\hbar). \quad (5)$$

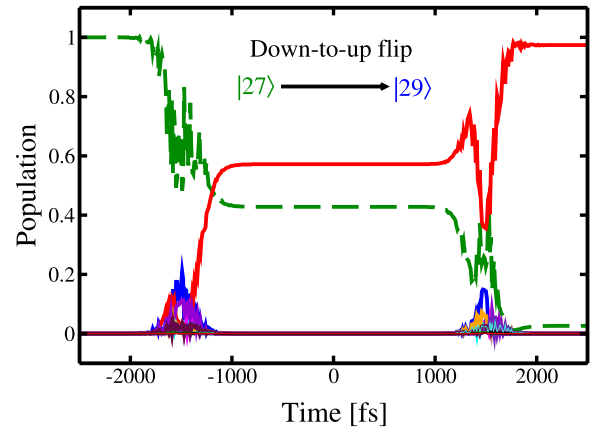


FIG. 8. The double laser-pulse-induced spin-flip process ($|27\rangle$ to $|29\rangle$), spin is localized on Ni2) on the cis configuration. Time evolution of the population of the initial (green dashed line), intermediate (different colors), and final (red solid line) states with $2 \times \text{FWHM}$ as a delay between the two pulses.

The Hamiltonians of the first and second pulses are represented by $\hat{H}_{\text{int1}}(t)$ and $\hat{H}_{\text{int2}}(t)$, respectively. The time delay between the laser pulses is symbolized by δt . The energies of states $|\Phi_n\rangle$ and $|\Phi_k\rangle$ are defined by E_n and E_k , respectively. The complex scalar coefficient of the state $|\Phi_n\rangle$ is expressed by $c_n(t)$.

We find that the double laser pulses with a time delay of 971 femtoseconds successfully induce the spin-flip process $|27\rangle \rightarrow |29\rangle$ on the cis configuration (Fig. 8, another process is shown in Fig. S13 of the Supplemental Material [42]). We take states $|27\downarrow\rangle$ and $|29\uparrow\rangle$ as initial and target states, respectively. The spin density is localized on Ni2 in both states. The fidelity of this process improves slightly from $\sim 96\%$ (with a single pulse) to 97% (with a double pulse). However, the improvement by the application of double pulses might be much more significant when used for otherwise less efficient spin dynamics processes. Since time delay δt is crucial in producing high fidelity, we study its effect on this process (Fig. S14 of the Supplemental Material [42]). We see that the fidelity changes when the time delay is varied, e.g., it is 97% (with $2 \times \text{FWHM}$) while it is 58.90% (with $4 \times \text{FWHM}$). Accordingly, the fourth key finding of this study is that the fidelity of the spin-flip process is highly sensitive to the time delay between the two laser pulses.

Next, we study the spatial resolution limit of individually addressing single magnetic Ni atoms in a circuit. By employing the spin-dynamic processes of zigzag silicon configurations, we show a perfect model of an optically incorporated processing block (Fig. 9).

The most basic building blocks for logic operation are Ni atoms (spin centers). In the context of integrated logic processors, a high degree of addressability at the level of individual spin centers is essential [132]. However, the minimum focal spot size achievable with laser pulses is on the order of several tens of nanometers, which remains approximately one order of magnitude larger than the characteristic dimensions of our system. As a result, laser-based addressing may lack the required spatial selectivity, since a single on-chip

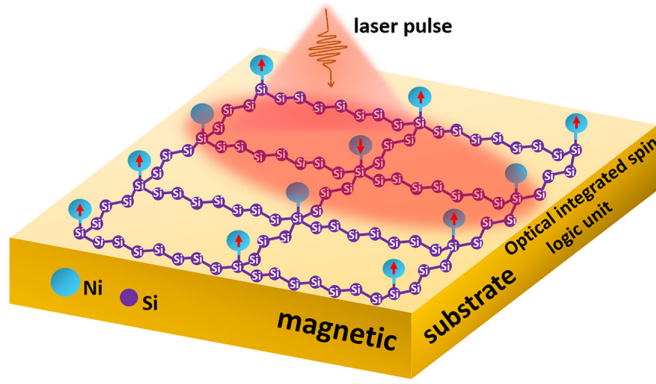


FIG. 9. Representation of an optical incorporated spin processing unit constructed from several zigzag silicon chain configurations. The cyan and violet spheres indicate the nickel and silicon atoms, respectively. The red arrows indicate the spin direction. The magnetic atoms can be independently addressed with the assistance of an inhomogeneous magnetic field.

pulse can simultaneously interact with multiple spin centers in densely packed configurations. Furthermore, given that the Λ process can only be realized with high fidelity under a well-defined magnetic field strength, we propose the use of an inhomogeneous magnetic field as an alternative strategy to selectively address individual spin centers (Ni atoms). Therefore, we establish that a single laser pulse effectively enables the addressability of an individual local spin-flip and hence improves the spatial resolution of the spin logical devices. Based on the effect of the magnetic field on the local spin-flip process $|2\downarrow\rangle \rightarrow |4\uparrow\rangle$ on Ni1, as shown in Fig. 10, we can identify the distance between two identical Ni1 (or Ni2) atoms by using the following formula:

$$d = \frac{|B_{\text{Before/After}} - B_{\text{peak}}|}{10} \mu\text{m}, \quad (6)$$

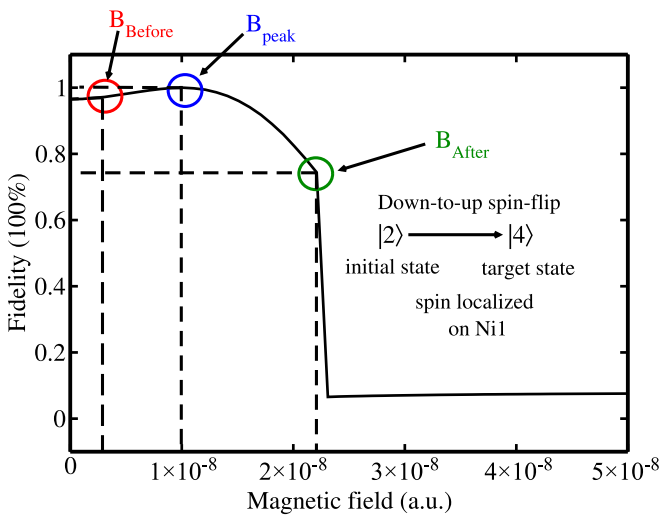


FIG. 10. Fidelity of the spin-down-to-up spin-flip process on Ni1 $|2\downarrow\rangle \rightarrow |4\uparrow\rangle$ in the cis configuration under the effect of various magnetic field values. The 0.0% fidelity at around $B = 0$ a.u., is simply due to the fact α and β states are indistinguishable at this magnetic field.

where d is the distance between two identical Ni atoms. $B_{\text{Before/After}}$ (in tesla) is the magnetic field chosen before or after B_{peak} , which is the magnetic field in which the fidelity reaches its maximum. For the process $|2\downarrow\rangle \rightarrow |4\uparrow\rangle$, we suppose that there are two neighboring cis configurations and the gradient of the magnetic field strength is $10 \text{ T}/\mu\text{m}$ [133–137]. In this scenario, with a weak magnetic field range from 0 to 1.00×10^{-8} a.u. (2.35 mT) the fidelity gradually increases. At this point, the maximum of the fidelity appears. Also, we relate this rise to the Zeeman splitting (the Zeeman effect gets enhanced) as the magnetic field increases, making each state more addressable. When $B > B_{\text{peak}} = 2.35 \text{ mT}$, the fidelity starts to gradually decrease (as illustrated in Fig. 10). This means SOC becomes relatively weak compared to the Zeeman interaction (Paschen-Back effect), and the dominant spin-mixed intermediate states tend to become spin-pure. This also suppresses the population-transfer channels and consequently inhibits the spin-flip process, leading to a drop in fidelity. Herein, to compute the shortest distance between two identical Ni1, we take $0.361_{|2\downarrow\rangle \rightarrow |4\uparrow\rangle}$ (96.80%) and $0.509_{|2\downarrow\rangle \rightarrow |4\uparrow\rangle}$ mT (97.40%) as two magnetic field values before B_{peak} . The distance d between two identical Ni1 atoms on the cis configurations is $1.9 \text{ \AA}$ (with 0.361 mT) while it is $1.8 \text{ \AA}$ when 0.509 mT is used. For $B > B_{\text{peak}} = 2.35 \text{ mT}$, the Ni1-Ni1 distance with $5.204_{|2\downarrow\rangle \rightarrow |4\uparrow\rangle}$ (74.4%) and $5.423_{|2\downarrow\rangle \rightarrow |4\uparrow\rangle}$ mT (6.5%) is 2.8 and $3.0 \text{ \AA}$, respectively. The same procedure is carried out for the spin-flip process $|27\downarrow\rangle \rightarrow |29\uparrow\rangle$ on Ni2. We find that the Ni2-Ni2 distance is less than $2.8 \text{ \AA}$ (see Table III for more detail).

Overall, the distances (reported in Table III) obtained both below and above the optimal magnetic field demonstrate the exceptional spatial resolution of the addressing scheme. Notably, the resolvable distances after the peak ($B > B_{\text{peak}}$) are larger ($2.8\text{--}3.0 \text{ \AA}$ for Ni1 and $0.24\text{--}0.47 \text{ \AA}$ for Ni2) compared to those before the peak ($1.8\text{--}1.9 \text{ \AA}$ for Ni1 and $0.094\text{--}0.14 \text{ \AA}$ for Ni2), indicating that the fidelity decreases more gradually on the low-field side and more abruptly on the high-field side. Nevertheless, all these distances are well below the projected CMOS scaling limit of $20 \text{ \AA}$, highlighting the potential of the cis configuration for extreme miniaturization. Furthermore, the existence of a range of magnetic field values around B_{peak} where high fidelity is maintained provides an important operational advantage: Small fluctuations in the applied magnetic field do not critically compromise the spin-addressing fidelity, ensuring robust device performance under realistic experimental conditions. This is identified as the fifth significant outcome of the current investigation.

Experimental feasibility and outlook. Several experimental techniques have already been successfully applied to probe spin states and ultrafast spin dynamics in systems closely related to ours. For instance, spin-polarized scanning tunneling microscopy (SP-STM) combined with differential conductance spectroscopy has been used to measure spin excitation energies in single metallocene molecules, such as nickelocene (NiCp_2), on surfaces, and to manipulate their spin states by controlling the atomic-scale environment [138,139]. Notably, a nickelocene-terminated STM tip has been demonstrated to act as a sensitive spin detector, enabling the simultaneous determination of the exchange field and spin polarization of iron atoms and cobalt films with atomic-scale resolu-

TABLE III. Effect of the magnetic field (B) on the fidelity of spin-flip process: $|2\downarrow\rangle \rightarrow |4\uparrow\rangle$ and $|27\downarrow\rangle \rightarrow |29\uparrow\rangle$. B_{peak} is the magnetic field corresponding to the highest fidelity. B_{Before} and B_{After} are the magnetic fields before and after the major peak, respectively. Notice that the gradient of the magnetic field strength is 10 T/ μm [133–137].

Spin-flip process ^a	B_{Before} (mT)		B_{peak} (mT)	B_{After} (mT)	
	0.361 $_{ 2\downarrow\rangle\rightarrow 4\uparrow\rangle}$ 1.637 $_{ 27\downarrow\rangle\rightarrow 29\uparrow\rangle}$	0.509 $_{ 2\downarrow\rangle\rightarrow 4\uparrow\rangle}$ 2.115 $_{ 27\downarrow\rangle\rightarrow 29\uparrow\rangle}$	2.35 $_{ 2\downarrow\rangle\rightarrow 4\uparrow\rangle}$ 3.055 $_{ 27\downarrow\rangle\rightarrow 29\uparrow\rangle}$	5.204 $_{ 2\downarrow\rangle\rightarrow 4\uparrow\rangle}$ 3.525 $_{ 27\downarrow\rangle\rightarrow 29\uparrow\rangle}$	5.423 $_{ 2\downarrow\rangle\rightarrow 4\uparrow\rangle}$ 3.774 $_{ 27\downarrow\rangle\rightarrow 29\uparrow\rangle}$
fidelity ^b of: $ 2\downarrow\rangle \rightarrow 4\uparrow\rangle$ ^c	96.80%	97.40%	100%	74.4%	6.5%
fidelity of: $ 27\downarrow\rangle \rightarrow 29\uparrow\rangle$ ^d	80.02%	92.28%	99.80%	94.85%	5.94%
$d_{ 2\downarrow\rangle\rightarrow 4\uparrow\rangle}$ (nm) ^e	0.19	0.18	—	0.28	0.30
$d_{ 27\downarrow\rangle\rightarrow 29\uparrow\rangle}$ (nm) ^f	0.14	0.094	—	0.047	0.024

^aTwo spin-flip processes on the cis configuration.

^bFidelity of each process.

^cSpin density is localized on Ni1.

^dSpin density is localized on Ni2.

^eDistance between identical Ni1 atoms.

^fDistance between identical Ni2 atoms.

tion. Besides, the x-ray magnetic circular dichroism (XMCD) technique provided the distinct advantage of unambiguously probing local magnetic properties through element-specific core-level excitations, thereby offering detailed insight into the mechanisms underlying the observed magnetic behavior. This technique has been successfully applied to study single-ion magnets deposited on metallic surfaces, such as Er(trensal) molecules on Au(111) and Ni/Cu(100) substrates, revealing the molecular structure preservation after sublimation and the magnetic coupling to the substrate [140].

Regarding also spin dynamics in silicon-based systems, significant progress has been made. All-electrical spin injection, transport, and detection have been demonstrated in heavily n -type-doped silicon nanowires using ferromagnetic Co/Al₂O₃ tunnel barrier contacts, achieving spin injection efficiencies of up to 30% and spin diffusion lengths of up to 6 μm at cryogenic temperatures [141]. More recently, ferromagnetic resonance (FMR) and magnetization measurements were performed on a set of samples with the structure [Si(10 Å)/Ni(t_{Ni} Å)]₂₀, where t_{Ni} represented the Ni layer thickness and 20 denoted the number of bilayer repetitions. The samples were fabricated using dc magnetron sputtering, with the Si layer thickness fixed at 10 Å while the Ni layer thickness was varied from 10 to 100 Å, to explore magnetic anisotropy in Si/Ni multilayers [142]. Most importantly for our predictions, ultrafast spin dynamics at Ni/Si interfaces have been experimentally probed using free electron laser-based element-sensitive time-resolved Kerr spectroscopy, revealing photoinduced spin currents propagating at a speed of approximately 0.2 nm/fs across the Ni/Si interface [143]. This directly demonstrates the feasibility of probing subpicosecond spin-transfer processes in Ni-Si hybrid systems. Moreover, the complex interactions at metal-semiconductor interfaces were examined, with particular emphasis on the magnetic proximity effect at Ni/Si interfaces, as revealed by systematic XMCD measurements at the Ni and Si absorption edges [144]. For the material platform itself (as aforementioned in Sec. I), planar silicene has been successfully synthesized on Au(111) films produced on Si(111) and mica substrates, with several two-dimensional

monolayers created using surface-segregated Au(111) thin films grown on Si(111) substrates [64,65,69]. Building on these established experimental achievements, our predictions can be accomplished using similar approaches. The spin state of Ni atoms on the zigzag silicon chain can be detected via SP-STM or XMCD. The subpicosecond spin-transfer dynamics predicted in this work can be tested using time-resolved magneto-optical Kerr effect (TR-MOKE) or time-resolved XMCD at free electron laser facilities (e.g., FERMI or BESSY II), directly leveraging the demonstrated capability to probe spin currents across Ni/Si interfaces. Thus, the proposed system is experimentally accessible, and the ultrafast spin-manipulation scenarios presented here are amenable to direct experimental investigation.

IV. CONCLUSION

In summary, we investigate the ultrafast laser-driven spin dynamics in a zigzag silicon chain functionalized with two Ni atoms using high-level quantum chemistry methods (CCSD and EOM-CCSD). The first major finding of this study is that spin transfer between Ni atoms can be achieved in a silicon-based channel system over distances of approximately 6.547 nm (cis configuration) and 6.021 nm (trans), within the subpicosecond regime, allowing coherent spin-based logic operations. Second, we find that the fidelity of spin transfer is highly sensitive to the orientation of the laser polarization: Even slight variations in the polar and azimuthal angles can cause a significant drop in fidelity, highlighting the need for precise control over excitation conditions. Third, varying the strength of the inhomogeneous magnetic field B is shown to strongly influence the fidelity of laser-driven ultrafast spin manipulations. Fourth, the fidelity of the spin-flip process is found to depend critically on the time delay between two sequential laser pulses, offering a means to fine-tune control protocols. Finally, a combination of tailored laser pulses and magnetic field gradients allows for the selective addressing of individual spin centers with high spatial resolution. Herein, we identify that the Ni2-Ni2 distance in the cis configuration can reach below 2.8 Å, and the minimum Ni-Ni separation resolvable by laser excitation is just 3.0 Å (well below the

projected CMOS scaling limit of 20 Å) emphasizing the platform's potential for extreme miniaturization. We systematically investigate the double-pulse-induced spin-dynamic processes, which may considerably improve the fidelities of otherwise less efficient processes.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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