

Ultrafast spin injection in graphene via dynamical carrier filtering at transition metal dichalcogenide interfaces

Shunsuke Yamada ^{1,*}, Arqum Hashmi ², and Tomohito Otake ¹

¹*Kansai Institute for Photon Science, National Institutes for Quantum Science and Technology (QST), Kyoto 619-0215, Japan*

²*Department of Nuclear Engineering and Management, Graduate School of Engineering, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan*



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We report a real-time first-principles study of ultrafast spin injection in a WSe₂-graphene heterobilayer under circularly polarized laser irradiation, using time-dependent density functional theory. Contrary to conventional expectations, spin transfer into graphene is not a passive process but is actively driven by spin-selective carrier filtering at the interface. Spin-polarized carriers generated in the WSe₂ layer induce a preferential migration of opposite-spin carriers from graphene, which results in net spin magnetization in graphene. This process is governed by interlayer band offsets, density-of-state asymmetry, and Pauli blocking. These findings indicate a microscopic mechanism of spin injection in nonmagnetic systems and identify a guiding principle for the design of ultrafast optospintronic functionalities in van der Waals heterostructures.

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I. INTRODUCTION

Two-dimensional (2D) materials such as transition metal dichalcogenides (TMDs) and graphene have emerged as promising platforms for spintronic applications, where the spin degrees of freedom are utilized as information carriers [1–4]. In particular, graphene exhibits exceptional carrier mobility and long spin diffusion lengths that exceed several micrometers, which makes it ideal for spin transport [5,6]. However, the intrinsically weak spin-orbit coupling (SOC) in graphene limits the efficient generation and manipulation of spin polarization, which poses a challenge for its integration into spintronic devices [7].

To address this limitation, van der Waals (vdW) heterostructures composed of graphene and TMDs have attracted considerable attention [8]. TMD monolayers possess strong intrinsic SOC due to the presence of heavy metal atoms and exhibit valley-dependent spin polarization that arises from spin-valley locking [9–11]. A first-principles computational study predicted ultrafast magnetization via SOC in nonmagnetic 2D materials including TMD monolayers under circularly polarized laser pulses on the femtosecond timescale [12]. Although graphene cannot generate spin magnetization optically due to its negligible SOC, its high mobility makes it an ideal medium for spin transport. Therefore, efficient spin transfer from a TMD layer to a graphene layer in a stacked heterostructure could enable optospintronic devices that combine efficient spin generation and long-range spin transport.

Such spin injection and transfer processes have been experimentally demonstrated in several TMD-graphene heterostructures using circularly polarized light pulses [13–16]. These systems have also exhibited several spintronic functionalities, such as spin-charge conversion via the spin Hall and Rashba-Edelstein effects [17,18], and tunable spin relaxation dynamics [19,20]. In particular, spin injection in TMD-graphene heterostructures has been discussed phenomenologically; however, its microscopic mechanism remains largely unexplored.

Most theoretical studies on TMD-graphene heterostructures have focused on their static band structures [21–29]. In contrast, real-time first-principles calculations based on time-dependent density functional theory (TDDFT) [30] provide a robust framework for the simulation of ultrafast spin and charge dynamics beyond empirical models. Previous TDDFT studies on ultrafast spin dynamics in 2D heterostructures [31–34] have primarily addressed the optical intersite spin transfer effect in magnetic materials [35,36]. A previous first-principles study of electron dynamics in TMD-graphene heterostructures addressed charge transfer, but it did not consider spin degrees of freedom [37]. The ultrafast dynamics of spin injection in TMD-graphene heterostructures via laser pulses remains unexplored to date.

In this paper, we reveal a dynamical mechanism of spin injection in a heterobilayer (HB) of WSe₂ and graphene in a nonequilibrium phase using real-time first-principles simulations based on TDDFT. Spin polarization generated in the WSe₂ layer induces the preferential migration of opposite-spin carriers from graphene, which leads to net spin polarization in graphene.

The remainder of this paper is organized as follows: Section II describes the theoretical methods and numerical conditions. In Sec. III, the calculation results are presented and analyzed in detail. Finally, the conclusions are presented in Sec. IV.

*Contact author: yamada.shunsuke@qst.go.jp

II. COMPUTATIONAL METHODS

A. Structure optimization

The lattice constants of monolayer WSe₂ and graphene, which are 3.32 and 2.46 Å, respectively, were used to construct the bilayer heterostructure. The HB was modeled using a 2×2 supercell for WSe₂ and a 3×3 supercell for graphene, with the graphene layer rotated by 19°. This configuration leads to a lattice mismatch within a tolerable range of 1.65%. Structural relaxation is performed using the OPENMX code [38,39]. During ionic relaxation, the shape and size of the supercell are fixed, while allowing for full relaxation of the atomic positions. The pseudoatomic basis sets used are $s^2p^2d^1$ for carbon and $s^3p^2d^2$ for both tungsten and selenium, with norm-conserving pseudopotentials taken from the OpenMX library [40]. Exchange-correlation effects are treated with the local spin density approximation (LSDA), namely, the LSDA-CA functional [41]. The Brillouin zone is sampled using a $15 \times 15 \times 1$ k -point grid, and an energy cutoff of 320 Ry is employed. A vacuum layer of 20 Å is introduced along the z direction to eliminate spurious interactions between periodic images. The optimization proceeds until the Hellmann-Feynman force on each atom is less than 0.0003 hartree/bohr, with the electronic energy convergence threshold set to 3.7×10^{-6} hartree. The bilayer distance between WSe₂ and graphene is found to be 3.47 Å, which is in good agreement with previous studies [22–24,42]. The interlayer distance reflects a weak vdW interaction, indicating that the interlayer coupling is minimal and the intrinsic electronic properties of graphenelike linear dispersion near the Fermi level are well preserved in the heterostructure. The optimized structure of the HB is parallel to the xy plane and the positions of the tungsten (W) and graphene layers are $z = 0$ and $z = 5.2$ Å, respectively. The selenium (Se) layers are located at $z = \pm 1.7$ Å.

B. TDDFT

TDDFT calculations for the HB under an electric field, $\mathbf{E}(t) = -(1/c)d\mathbf{A}(t)/dt$, were performed using the SALMON code [43–46]. The orbital wave functions are defined in a box that contains the unit cell of the 2D material sandwiched between vacuum regions. Each Bloch orbital $u_{n\mathbf{k}}(\mathbf{r}, t)$ is a two-component spinor, where n and $\mathbf{k}$ denote the band index and 2D crystal momentum, respectively.

The time-dependent Kohn-Sham equation is described as follows:

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} u_{n\mathbf{k}}(\mathbf{r}, t) &= \left[\frac{1}{2m} \left(-i\hbar \nabla + \hbar \mathbf{k} + \frac{e}{c} \mathbf{A}(t) \right)^2 \right. \\ &\quad \left. - e\varphi(\mathbf{r}, t) + \hat{v}_{\text{NL}}^{\mathbf{k} + \frac{e}{\hbar c} \mathbf{A}(t)} + v_{\text{xc}}(\mathbf{r}, t) \right] u_{n\mathbf{k}}(\mathbf{r}, t), \end{aligned} \quad (1)$$

where the scalar potential $\varphi(\mathbf{r}, t)$ includes the Hartree potential from the electrons and the local component of the ionic pseudopotentials and we have defined $\hat{v}_{\text{NL}}^{\mathbf{k}} \equiv e^{-i\mathbf{k} \cdot \mathbf{r}} \hat{v}_{\text{NL}} e^{i\mathbf{k} \cdot \mathbf{r}}$. Here, $\hat{v}_{\text{NL}}$ and $v_{\text{xc}}(\mathbf{r}, t)$ represent the nonlocal component of

the ionic pseudopotentials and exchange-correlation potential, respectively.

The dynamics of the valence electrons are treated with the norm-conserving pseudopotential [47], and the adiabatic local spin density approximation (ALSDA) is assumed for the exchange-correlation energy functional [48]. SOC is incorporated through the j -dependent nonlocal potential [49]. Although the ALSDA exchange-correlation term is unable to describe spin torque effects, the SOC term in the nonlocal potential converts the angular momentum of excited carriers into spin-flipping torque [12]. ALSDA remains a standard choice in state-of-the-art TDDFT studies for femtosecond spin dynamics [12,36,50]. Because the exchange-correlation spin-torque contribution is very small on tens-of-femtosecond timescales even in magnetic systems [51], it can be safely neglected in our nonmagnetic case.

The α component ($\alpha = x, y, z$) of the spin magnetization per unit area is defined as

$$m_\alpha(t) = \frac{1}{N_k} \sum_{\mathbf{k}, n} f_{n\mathbf{k}} \int d\mathbf{z} \int_\Omega \frac{dxdy}{\Omega} u_{n\mathbf{k}}^\dagger(\mathbf{r}, t) \frac{\sigma_\alpha}{2} u_{n\mathbf{k}}(\mathbf{r}, t), \quad (2)$$

where σ_α is the Pauli matrix and Ω is the area of the 2D unit cell. N_k is the number of k -points and $f_{n\mathbf{k}}$ is the occupation rate. Based on the first-order perturbative expansion of the wave function $u_{n\mathbf{k}}(\mathbf{r}, t) \approx u_{n\mathbf{k},t}(\mathbf{r}) + \delta u_{n\mathbf{k}}(\mathbf{r}, t)$, where $u_{n\mathbf{k},t}(\mathbf{r})$ is the adiabatic eigenstate and $\delta u_{n\mathbf{k}}(\mathbf{r}, t)$ represents the first-order correction, the first-order contribution to Eq. (2) is given by

$$\text{Re}[u_{n\mathbf{k},t}^\dagger(\mathbf{r}) \sigma_\alpha \delta u_{n\mathbf{k}}(\mathbf{r}, t)] \propto \mathbf{A}, \quad (3)$$

while the second-order contribution is

$$\delta u_{n\mathbf{k}}^\dagger(\mathbf{r}, t) \frac{\sigma_\alpha}{2} \delta u_{n\mathbf{k}}(\mathbf{r}, t) \propto \mathbf{A}^2. \quad (4)$$

The $\alpha\beta$ component of the spin-current density averaged over the unit cell is defined as

$$\begin{aligned} J_{\alpha\beta}^{\text{spin}}(t) &= \frac{1}{m} \int_\Omega \frac{d^3r}{\Omega} \frac{1}{N_k} \text{Re} \sum_{\mathbf{k}, n} f_{n\mathbf{k}} u_{n\mathbf{k}}^\dagger(\mathbf{r}, t) \sigma_\alpha \\ &\quad \times \left[-i\hbar \nabla_\beta + \hbar k_\beta + \frac{e}{c} A_\beta(t) \right] u_{n\mathbf{k}}(\mathbf{r}, t) + J_{\text{NL},\alpha\beta}^{\text{spin}}(t), \end{aligned} \quad (5)$$

$$\begin{aligned} J_{\text{NL},\alpha\beta}^{\text{spin}}(t) &= \int_\Omega \frac{d^3r}{\Omega} \frac{1}{N_k} \text{Re} \sum_{\mathbf{k}, n} f_{n\mathbf{k}} u_{n\mathbf{k}}^\dagger(\mathbf{r}, t) \sigma_\alpha \\ &\quad \times \frac{1}{i\hbar} \left[r_\beta, \hat{v}_{\text{NL}}^{\mathbf{k} + \frac{e}{\hbar c} \mathbf{A}(t)} \right] u_{n\mathbf{k}}(\mathbf{r}, t), \end{aligned} \quad (6)$$

where $J_{\text{NL},\alpha\beta}^{\text{spin}}(t)$ is a correction term that stems from the nonlocal potential. Without the Pauli matrix σ_α , the above definition is equivalent to the β component of the electron current density averaged over the unit cell, $J_\beta(t)$.

The occupancy of the time-dependent wave functions at time t is estimated by projecting $\{u_{n\mathbf{k}}(t)\}$ onto the ground-state orbitals $\{u_{n\mathbf{k}}^{\text{GS}}\}$ as

$$F_{n\mathbf{k}}(t) = \sum_m f_{m\mathbf{k}} | \langle u_{n\mathbf{k}}^{\text{GS}} | u_{m\mathbf{k}}(t) \rangle |^2. \quad (7)$$

At $t = 0$, $F_{n\mathbf{k}}(t)$ reduces to the ground-state occupancy $f_{n\mathbf{k}}$. To visualize changes in occupancy, we define the weighted local density of states as

$$w(\varepsilon, \mathbf{r}, t) = \frac{1}{N_k} \sum_{k,n} \Delta F_{n\mathbf{k}}(t) |u_{n\mathbf{k}}^{\text{GS}}(\mathbf{r})|^2 \delta(\varepsilon - \varepsilon_{n\mathbf{k}}^{\text{GS}}), \quad (8)$$

where $\Delta F_{n\mathbf{k}}(t) = F_{n\mathbf{k}}(t) - f_{n\mathbf{k}}$. This quantity represents the density distribution of excited carriers at time t . In the absence of $\Delta F_{n\mathbf{k}}(t)$, the above expression is equivalent to the conventional local density of states (LDoS), $D(\varepsilon, \mathbf{r})$. By sorting each component with respect to $(n, \mathbf{k})$, the spin-up and spin-down contributions can be separated as $w_{\uparrow, \downarrow}(\varepsilon, \mathbf{r}, t)$.

Using the atomic structure of the WSe₂-graphene HB obtained in Sec. II A, first-principles TDDFT calculations are performed via SALMON code [45,46]. The norm-conserving pseudopotentials are the same as those used in the structure optimization via the OPENMX code. The spatial grid sizes and k -points are optimized according to the converging results. For both the conventional WSe₂ monolayer and WSe₂-graphene HB, the determined parameter of the real-space grid spacing is 0.21 Å. The optimized k -mesh in the 2D Brillouin zone is 16×16 for the conventional WSe₂ monolayer and 8×8 for the WSe₂-graphene HB. The time step is set to $dt = 5 \times 10^{-4}$ fs. The calculation conditions for the conventional WSe₂ monolayer are almost the same as those employed in Refs. [52–55].

The vector potential of the applied circularly polarized light pulse is given by

$$\begin{aligned} \mathbf{A}(t) = & -\frac{cE_{\text{max}}}{\omega} \sin^4\left(\frac{\pi t}{T}\right) \\ & \times \left[\hat{\mathbf{x}} \cos\left\{\omega\left(t - \frac{T}{2}\right)\right\} + \hat{\mathbf{y}} \sin\left\{\omega\left(t - \frac{T}{2}\right)\right\} \right], \\ & (0 < t < T), \end{aligned} \quad (9)$$

where $\hbar\omega = 1.5$ eV and $T = 20$ fs unless otherwise described. The peak electric field amplitude E_{max} is chosen to yield a peak intensity ranging from $I = 10^8$ to 10^{12} W/cm².

III. RESULTS

Figure 1(a) shows the applied electric field associated with a left-handed circularly polarized pulse as a function of time. Here, we employ a $\sin^4$ envelope of the vector potential with a pulse duration of $T = 20$ fs, a peak intensity of $I = 10^9$ W/cm², and a photon energy of $\hbar\omega = 1.5$ eV. In Fig. 1(b), the polarization along the z axis, $P_z(t) = \int_0^t dt' J_z(t')$, is compared across a range of laser intensities from 10^9 to 10^{12} W/cm². This quantity represents the amount of electron transfer between the two layers. The excited carrier density is expected to be proportional to the square of the electric field; therefore, the lines in panel (b) are scaled by a factor of 0.1^n ($n = 0, 1, 2, 3$ for $I = 10^{9+n}$ W/cm²) for comparison. The negative sign of the curves in this figure indicates that electrons are transferred from the graphene layer to the WSe₂ layer. This observation is consistent with a previous first-principles study for a MoS₂-graphene HB under a linearly polarized laser pulse [37]. Electrons are transferred from the graphene layer to the

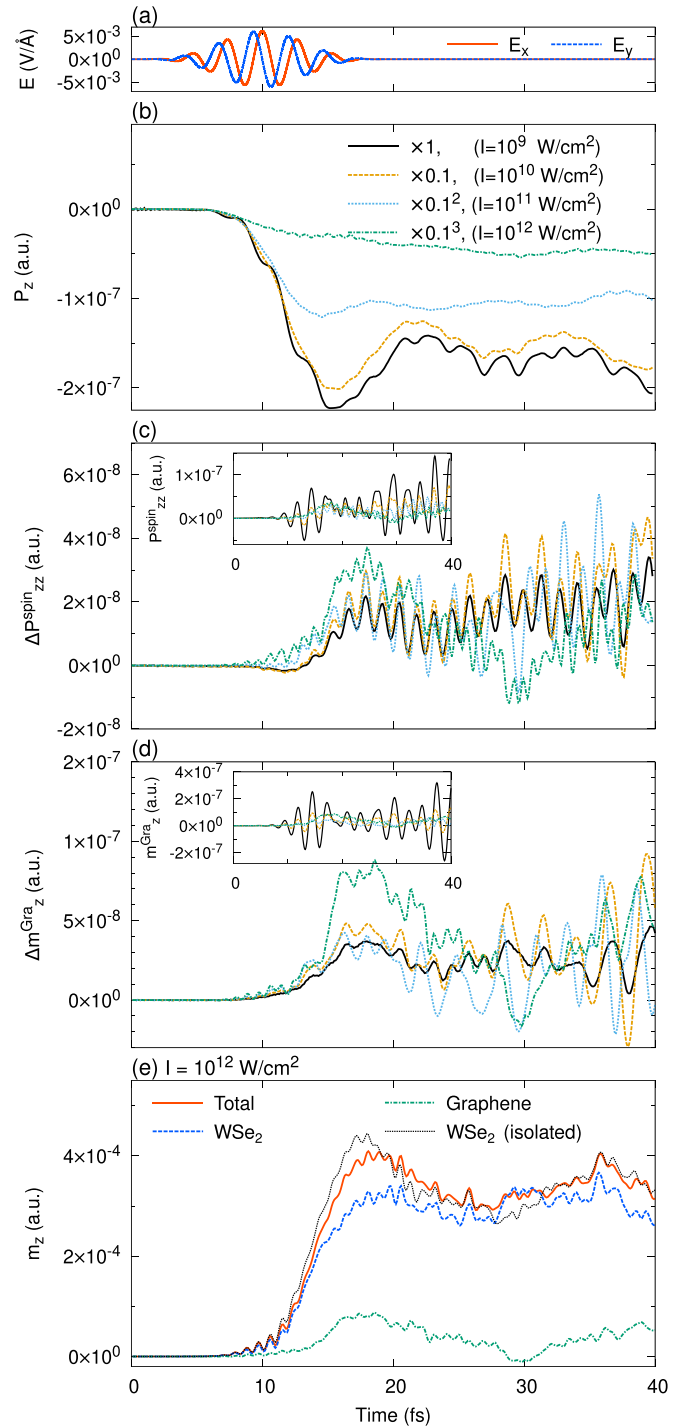


FIG. 1. (a) Applied electric field. (b)–(d) Various quantities for WSe₂-graphene HB under laser intensity I , in the range from 10^9 to 10^{12} W/cm². All curves are scaled by a factor of 0.1^n ($n = 0, 1, 2, 3$ for $I = 10^{9+n}$ W/cm²) for comparison. (b) Electron transfer $P_z(t)$. (c) Nonlinear component of spin transfer $\Delta P_{zz}^{\text{spin}}(t)$. Inset: Spin transfer, $P_{zz}^{\text{spin}}(t)$. (d) Nonlinear component of spin magnetization around graphene, $\Delta m_z^{\text{Gra}}(t)$. Inset: Spin magnetization around graphene, $m_z^{\text{Gra}}(t)$. (e) Total spin magnetization $m_z(t)$, and its decomposition into contributions from each layer under $I = 10^{12}$ W/cm².

TMD layer due to the imbalance in the density of states (DoS) between the two layers. In the low-intensity regime, the polarization is proportional to the square of the electric field, as

discussed in Ref. [37]. At higher intensities, the polarization saturates with increasing intensity, which can be attributed to Pauli blocking by excited and transferred carriers. Here, we use “Pauli blocking” in the general sense of phase-space filling: Photoexcitation and interlayer transfer populate final states and reduce the available phase space for further transitions.

Let us now consider the spin transfer, as represented by the time-integrated zz component of the averaged spin-current density, $P_{zz}^{\text{spin}}(t) = \int_0^t dt' J_{zz}^{\text{spin}}(t')$. This quantity corresponds to the polarization of the z -component spin along the z axis. Unlike the electron transfer $P_z(t)$, the spin transfer $P_{zz}^{\text{spin}}(t)$ includes a prominent first-order term that is proportional to the electric field, as suggested by Eq. (3). Figure 1(c) presents the nonlinear component of the spin transfer, $\Delta P_{zz}^{\text{spin}}(t)$, scaled by the factor 0.1^n . This is defined as $\Delta P_{zz}^{\text{spin}}(t) = P_{zz}^{\text{spin}}(t) - \sqrt{10}^{n+1} P_{zz}^{\text{spin}}(t)|_{I=10^8 \text{ W/cm}^2}$, where the scaled linear component obtained from the 10^8 W/cm^2 (linear response regime) result is subtracted. This procedure reveals the behavior of real excitation while eliminating the first-order contribution. The inset shows the original data for the spin transfer $P_{zz}^{\text{spin}}(t)$ scaled by the factor 0.1^n and demonstrates that the first-order contribution does not affect the nodal structure of the real excitation. The positive sign of the curves in Fig. 1(c) indicates that spin transfer occurs in the opposite direction to electron transfer, i.e., from WSe₂ to graphene.

Next, we consider the transferred spin magnetization around the graphene layer; $m_z^{\text{Gra}}(t)$ is defined in the same manner as Eq. (2) but with the integration region restricted to the vicinity of graphene [right-hand side of the vertical line in Fig. 2(d)]. Figure 1(d) shows the nonlinear component, $\Delta m_z^{\text{Gra}}(t) = m_z^{\text{Gra}}(t) - \sqrt{10}^{n+1} m_z^{\text{Gra}}(t)|_{I=10^8 \text{ W/cm}^2}$, scaled by the factor 0.1^n . The inset shows the original data $m_z^{\text{Gra}}(t)$ scaled by the factor 0.1^n . A comparison of Figs. 1(c) and 1(d) reveals that the behavior of spin magnetization in graphene and spin transfer along the z axis is qualitatively similar.

Figure 1(e) shows the total spin magnetization $m_z(t)$ and its decomposition into contributions from each layer under $I = 10^{12} \text{ W/cm}^2$. The green dash-dotted line, $m_z^{\text{Gra}}(t)$, represents the same data as shown in the inset of Fig. 1(d) but without the scaling. The thin black dotted line indicates the results for the isolated WSe₂ monolayer, which were calculated using the same atomic positions and supercell as in the HB case but with graphene eliminated. The total spin magnetization of the HB closely matches that of the isolated WSe₂ monolayer. From the time average of the data in Fig. 1(e) over the interval $t = 20\text{--}40$ fs, the spin transfer efficiency from WSe₂ to graphene is estimated to be approximately 0.1.

These results indicate that spin transfer occurs via migration of a portion of spin magnetization from the WSe₂ layer to the graphene layer. The amount of spin transfer is approximately proportional to the square of the electric field, which is consistent with Eq. (4).

Figure 2 shows LDos weighted by the change in occupancy from the ground state, as a function of the orbital energy ε and z coordinate, $w_{\uparrow,\downarrow}(\varepsilon, z)$, at the pulse end ($t = 20$ fs) with $I = 10^9 \text{ W/cm}^2$. The positions of the atomic layers are indicated by small arrows. At $z = 0$, the conduction band minimum and valence band maximum for WSe₂ appear around

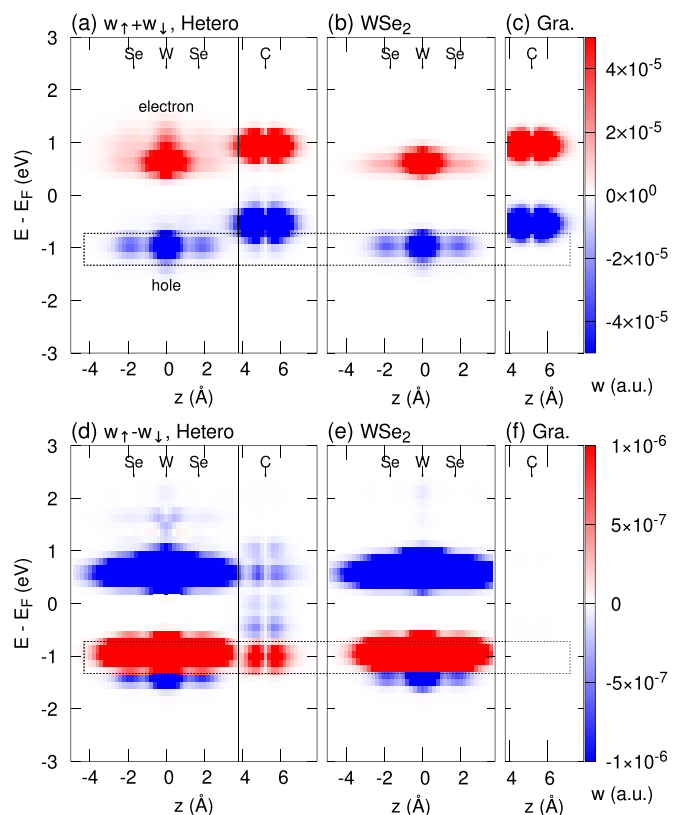


FIG. 2. Local density of states weighted by the change in occupancy from the ground state, $w_{\uparrow,\downarrow}(\varepsilon, z)$, at the pulse end with an intensity of $I = 10^9 \text{ W/cm}^2$. (a)–(c) [(d)–(f)] Sum (difference) of the spin-up and spin-down contributions. Panels (a) and (d) correspond to the HB results, while panels (b) and (e) [(c) and (f)] show the results for the isolated WSe₂ (graphene) monolayer.

0.5 and -1 eV, respectively. Panels (a)–(c) [(d)–(f)] show the sum (difference) of the spin-up and spin-down contributions, $w_{\uparrow} + w_{\downarrow}$ ($w_{\uparrow} - w_{\downarrow}$). In panels (a)–(c) [(d)–(f)], the positive and negative regions represent the spatial distributions of excited electrons and holes (up-spin and down-spin polarizations), respectively. Panels (a) and (d) correspond to the HB results, while panels (b) and (e) [(c) and (f)] show the results for the isolated WSe₂ (graphene) monolayer. At this low laser intensity, the excitation is expected to remain within the linear response regime.

In Figs. 2(a)–2(c), the result for the HB [panel (a)] shows strong similarity to the superposition of the isolated monolayer results [panels (b) and (c)], but the hot spots of excited carriers differ along the energy axis. Around $z = 0$ and $\varepsilon = 1$ eV in Fig. 2(a), excited electrons are observed that are absent in Fig. 2(b). These electrons are transferred from the graphene layer to the WSe₂ layer due to the band offset and the DoS imbalance between the two layers [37]. Regarding the spin imbalance of excited carriers, Fig. 2(d) clearly shows spin transfer from the WSe₂ layer to the graphene layer. As expected, the isolated graphene does not exhibit spin excitation [Fig. 2(f)]. From Fig. 2(d), the spin distribution in the graphene layer corresponds to that in the WSe₂ layer.

Figure 3 presents the same analysis as Fig. 2, but under a higher laser intensity of $I = 10^{12} \text{ W/cm}^2$. At this intensity,

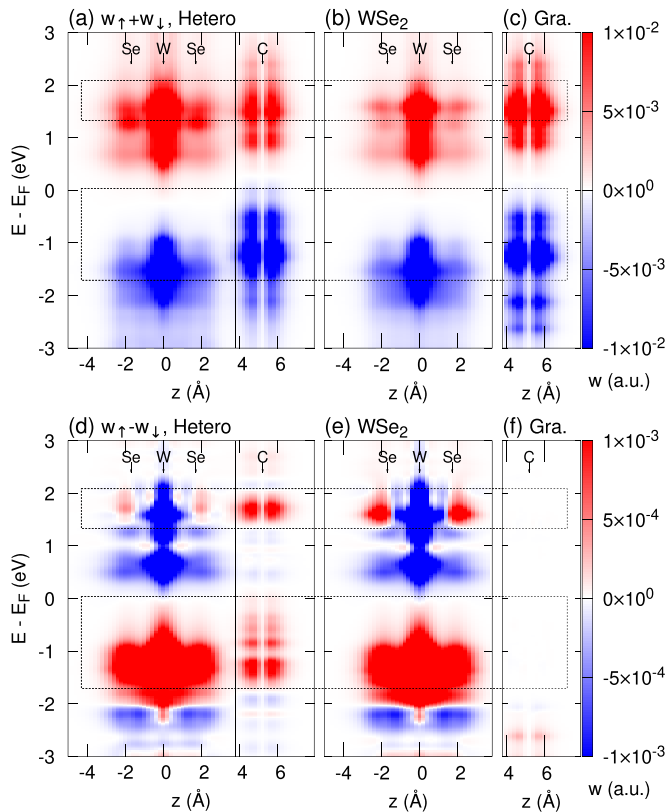


FIG. 3. Local density of states weighted by the change in occupancy from the ground state, $w_{\uparrow,\downarrow}(\epsilon, z)$, at the pulse end with an intensity of $I = 10^{12} \text{ W/cm}^2$. (a)–(c) [(d)–(f)] Sum (difference) of the spin-up and spin-down contributions. Panels (a) and (d) correspond to the HB results, while panels (b) and (e) [(c) and (f)] show the results for the isolated WSe_2 (graphene) monolayer.

excited carriers are distributed over a broader energy range due to nonlinear multiphoton excitation. The nonlinear response can arise from both multiphoton excitation and strong-field modifications of the electronic structure such as transient band renormalization or the dynamical Franz-Keldysh effect [55]. While the overall trend remains similar to that in Fig. 2, the spin distribution shown in Figs. 3(d) and 3(e) becomes more complex. The spin distribution around the Se layer at $z = 1.7 \text{ \AA}$ appears to be transferred to the graphene layer, although it differs from the distribution observed near the W layer. In Figs. 2(d) and 3(d), the spin distribution in the graphene layer corresponds to that in the Se layer at $z = 1.7 \text{ \AA}$. In Figs. 2(d) and 3(d), the prominent hole contribution in graphene reflects spin-valley locking in the WSe_2 valence band and favorable interfacial overlap of valence states between the adjacent Se layer and graphene layer, leading to efficient coupling to graphene states just below the Fermi energy.

We focus on the migration of up-spin electrons because it is the main contribution to the spin transfer. The boxed areas outlined by the black dotted lines in Figs. 2 and 3 highlight the regions where the spin-up polarization emerges in the graphene layer of the HB. The horizontal (z -axis) transport of carriers within these regions gives rise to a net positive spin current from WSe_2 to graphene, as indicated by the

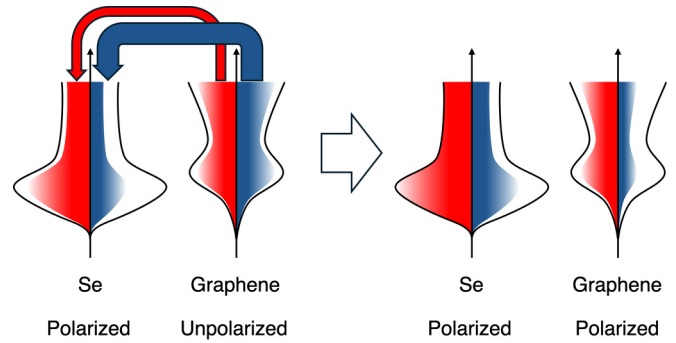


FIG. 4. Schematic of spin-selective electron migration between the Se layer at $z = 1.7 \text{ \AA}$ and the graphene layer.

positive spin transfer in Figs. 1(c) and 1(d). A comparison of the vicinity of graphene in Figs. 2(a) and 2(c), as well as in Figs. 3(a) and 3(c), within the boxed areas, reveals that the corresponding carrier densities exhibit a decrease in electron density and an increase in hole density in the graphene layer of the HB. This observation suggests that electrons are transferred from graphene to WSe_2 , which is consistent with the negative electron transfer shown in Fig. 1(b).

Spin-polarized carriers are generated in WSe_2 , and the electron current from graphene to WSe_2 is subject to Pauli blocking, which selectively inhibits one spin channel. As a result, migration preferentially occurs in the opposite-spin channel, which leads to an accumulation of blocked spins in the graphene layer. This spin-selective migration induces a correlated spin distribution between the Se layer at $z = 1.7 \text{ \AA}$ and the graphene layer (Fig. 4). The occurrence of Pauli blocking due to excited carriers is supported by the saturation of electron transfer observed in Fig. 1(b). The similarity in the total spin magnetization between the HB and the monolayer WSe_2 is consistent with photoexcitation occurring almost independently in the WSe_2 and graphene layers (Fig. 7). The observed charge and spin transfer can be attributed to vertical (energy-axis) photoexcitation within each layer, followed by horizontal (z -axis) carrier transport across energy levels. Such spin transfer has been reported in an HB of magnetic and nonmagnetic 2D materials under a linearly polarized pulse, but is attributed instead to intrinsic spin splitting of the static band structure [34]. In the present case, the spin-selective carrier filtering at the interface is entirely driven by dynamical effects in the nonmagnetic system.

We note that Pauli blocking and spin accumulation via asymmetric spin-dependent interfacial transport have been extensively discussed in spintronics, including the concept of spin extraction [56]. The present work extends this logic to an ultrafast, optically driven, nonmagnetic vdW heterostructure and identifies its time-domain microscopic implementation from real-time first-principles calculations.

Recent experiments indicate that electron transfer from graphene to TMD driven by DoS asymmetry is universal in heterostructures [57,58]. Hence, the present mechanism, based solely on electron transfer and Pauli blocking, is generic and does not depend on fine details of the band structure. Consistent with this, we find that varying the interlayer distance by $\pm 0.5 \text{ \AA}$ leaves the direction of spin transfer unchanged (Appendix C). Clarifying the impact at small twist angles,

where moiré minibands may form, is an important direction for future work.

Although our study focuses on the initial ≤ 40 fs regime, spin lifetimes in TMD-graphene heterostructures have been experimentally confirmed to be on the picosecond scale [13,17,19,20,24]. Therefore, the ultrafast spin injection identified here should be observable using time-resolved angle-resolved photoemission spectroscopy, Kerr rotation, or magnetic circular dichroism, all of which are standard techniques for probing femtosecond spin dynamics in 2D materials [59–62].

Our TDDFT framework treats the dynamics as fully coherent and does not explicitly account for dissipative processes (dephasing and scattering) in graphene or at the interface, which may become relevant on ~ 10 – 100 fs timescales. Accordingly, we restrict our focus to spin-current transfer across the interface during the laser pulse (≤ 40 fs), ignoring dissipative processes. The subsequent decay governed by dephasing and scattering, as the spin polarization propagates laterally through graphene toward remote electrodes, lies beyond the scope of the present work. Explicitly incorporating these dissipative processes is an important direction for future research.

IV. CONCLUSION

In conclusion, the microscopic mechanisms behind spin injection in TMD-graphene heterostructures under circularly polarized laser irradiation were analyzed using TDDFT. First-principles calculations reveal that Pauli blocking between WSe₂ and graphene under laser excitation drives spin injection via opposite spin and electron transfer. The carrier transfer occurs on an ultrafast timescale that renders relaxation effects negligible. These nonequilibrium dynamics play a crucial role in the establishment of spin polarization in graphene. These findings clarify the microscopic mechanism that underlies spin injection in nonmagnetic 2D heterostructures and provide a foundation for ultrafast optospintronic applications.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

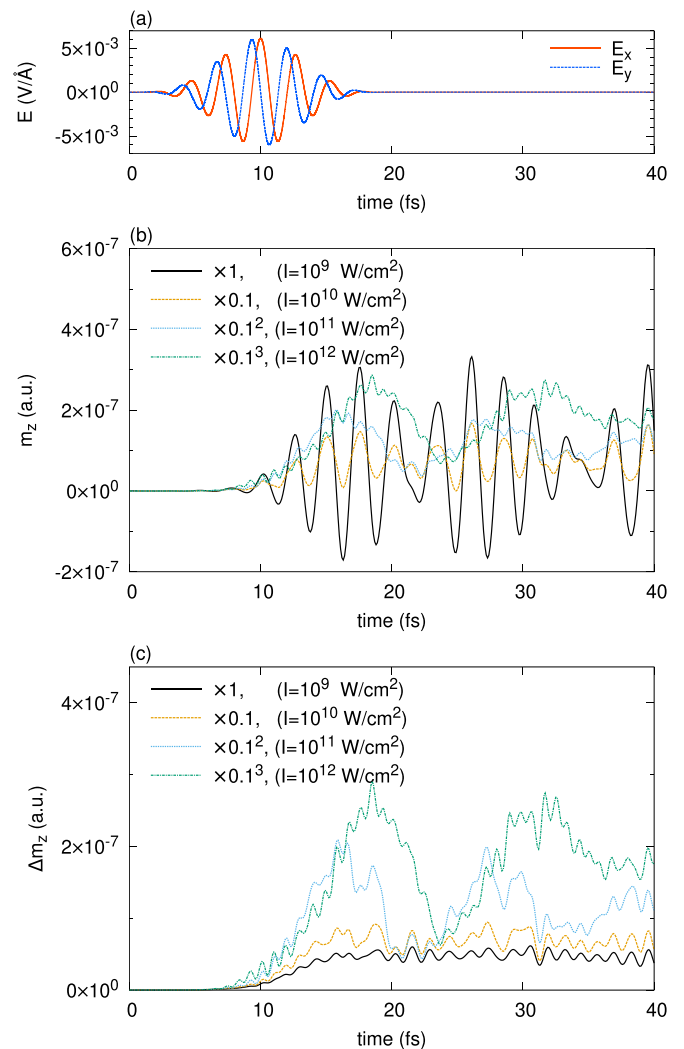


FIG. 5. (a) Applied electric field. (b) Spin magnetization per unit area in the conventional WSe₂ monolayer under the applied electric field. (c) Nonlinear contribution of panel (b) obtained by subtracting the scaled linear response.

APPENDIX A: SPIN MAGNETIZATION IN MONOLAYER WSe₂

Here, we consider ultrafast spin dynamics in the conventional WSe₂ monolayer under circularly polarized laser pulse. Figure 5 shows the calculated spin magnetization per unit area in monolayer WSe₂ under pulsed electric field excitation. Panel (a) is the applied electric field with an intensity of $I = 10^9$ W/cm², while panel (b) shows the resulting spin magnetization $m_z(t)$. Panel (c) isolates the contribution from nonlinear components by subtracting the scaled linear response. In panel (b), the spin magnetization is compared across a range of laser intensities from 10^9 to 10^{12} W/cm². The lines in panel (b) are normalized by a factor of 0.1^n ($n = 0, 1, 2, 3$ for $I = 10^{9+n}$ W/cm²). In the low-intensity regime, the nodal structure for 10^9 W/cm² (black solid line) coincides with that for 10^{10} W/cm² (orange dashed line), but the former exhibits a pronounced oscillation with the same frequency as that of the applied electric field. This indicates that the spin magnetization in this regime is a superposition of

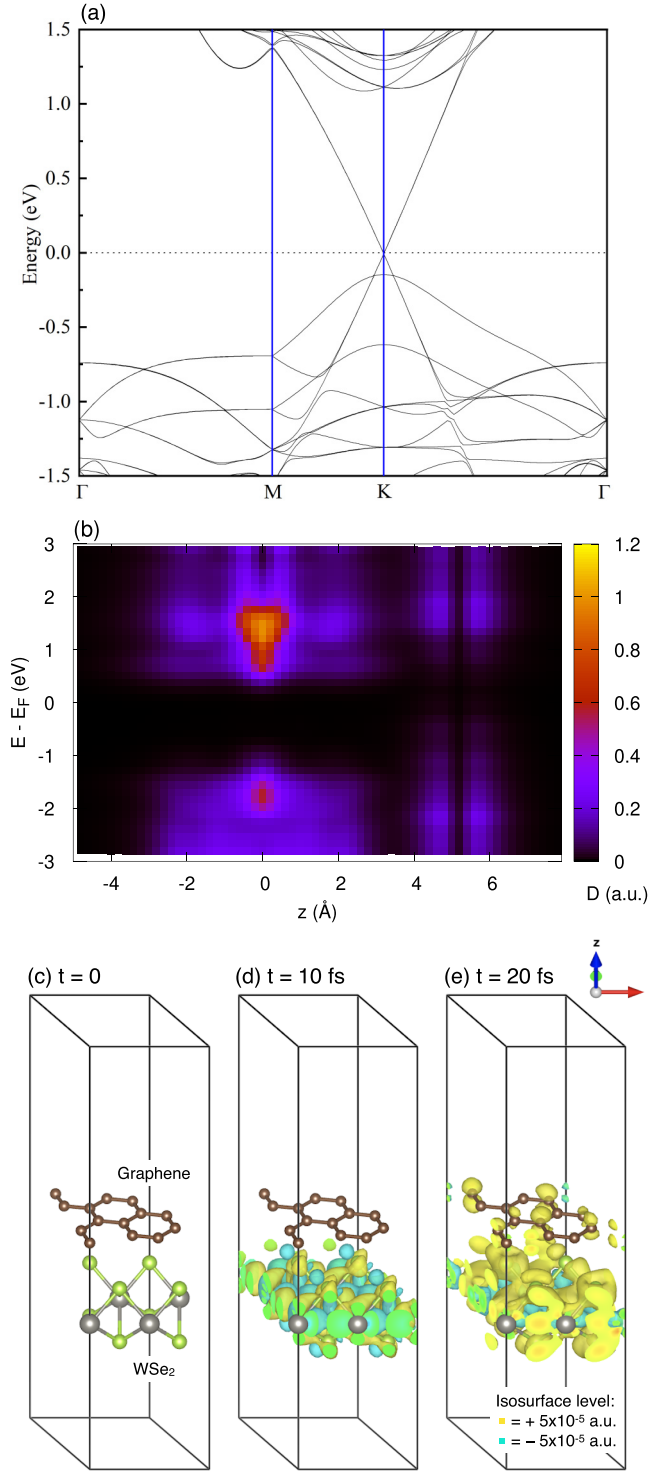


FIG. 6. (a) Band structure of the WSe₂-graphene HB. (b) Local density of states. (c)–(e) Snapshots of the spin magnetization density distribution from $t = 0$ to $t = 20$ fs under laser irradiation with $I = 10^{12}$ W/cm² (visualized by VESTA [63]).

components proportional to the first- and second-order terms of the electric field [see Eqs. (3) and (4)]. The second-order component corresponds to the real excitation of the spin magnetization.

TABLE I. List of the spin transfer efficiency with the pulse duration T (fs) and laser intensity I (W/cm²).

	$T = 10$	$T = 20$	$T = 30$
$I = 10^9$	−1.2	0.42	0.24
$I = 10^{10}$	−1.4	0.40	0.19
$I = 10^{11}$	0.66	0.15	0.11
$I = 10^{12}$	0.023	0.10	0.10

As the intensity increases, the contribution from the linear (first-order) term diminishes its importance. To isolate the nonlinear response, panel (c) subtracts the scaled linear component obtained from the 10^8 W/cm² result and multiplied by $\sqrt{10}^{n+1}$ from each spin magnetization, $\Delta m_z(t) = m_z(t) - \sqrt{10}^{n+1} m_z(t)|_{I=10^8 \text{ W/cm}^2}$. This procedure reveals the behavior of the higher-order terms of the spin magnetization. As expected, the curves for 10^9 and 10^{10} W/cm² closely overlap. At higher intensities, nonlinear excitation involving higher-order terms becomes significant. The behavior indicates that the spin magnetization scales approximately with the square of the electric field.

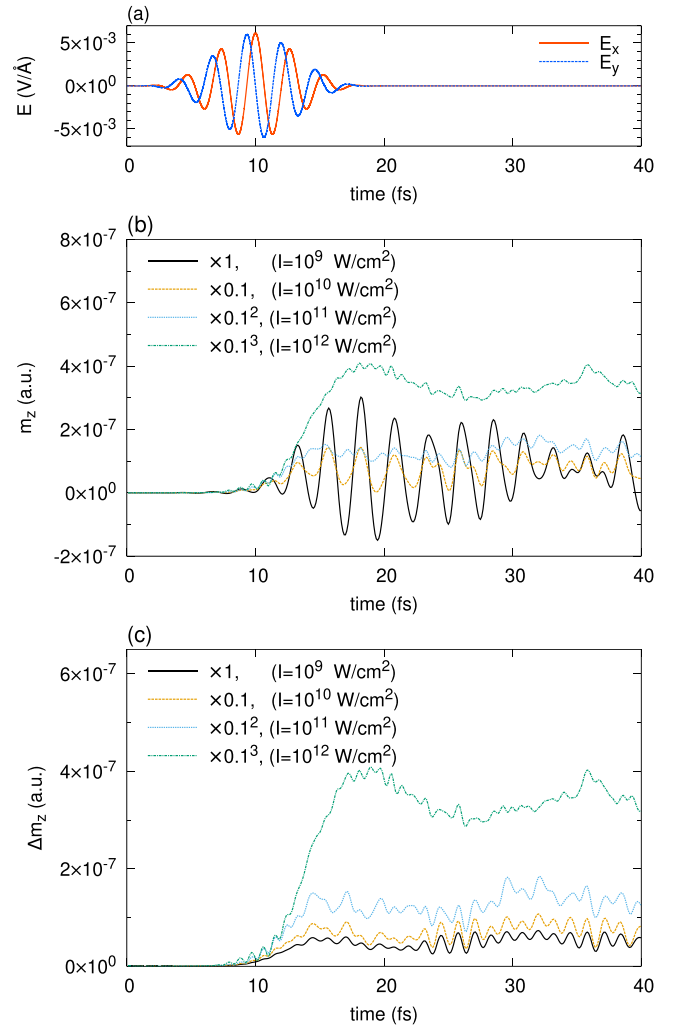


FIG. 7. The same as Fig. 5 but for the WSe₂-graphene HB.

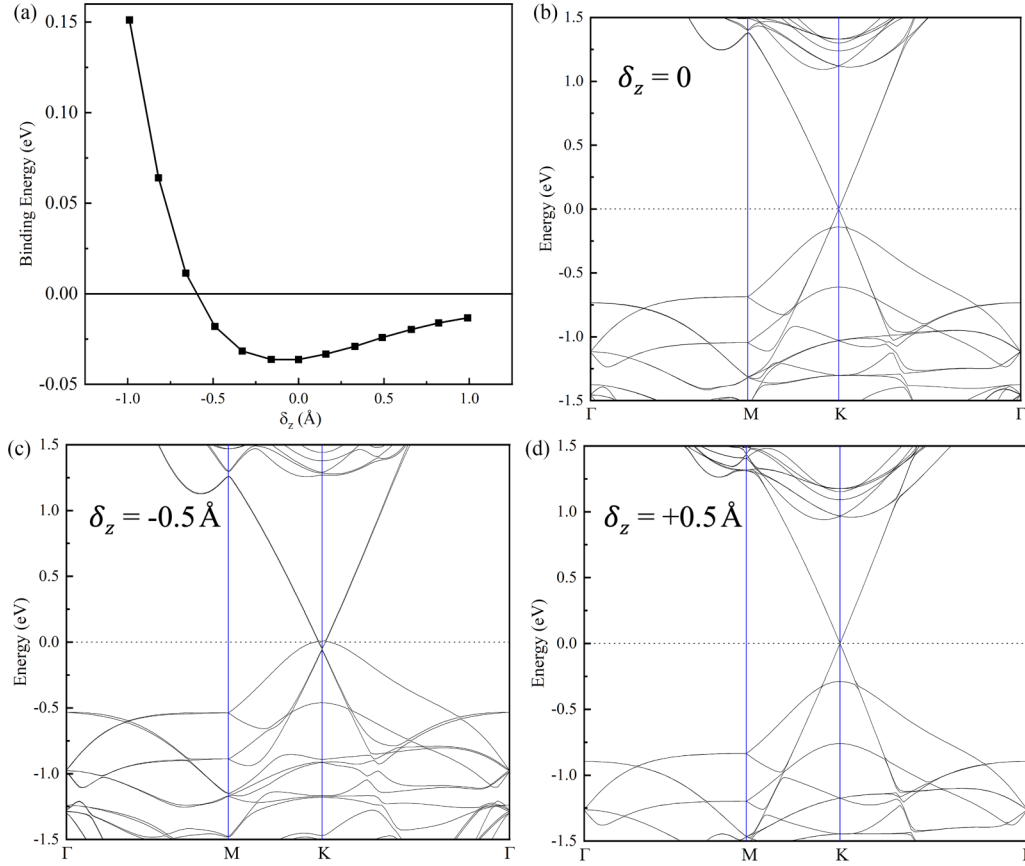


FIG. 8. Interlayer distance dependence of WSe₂-graphene HB. (a) Binding energy as a function of the interlayer distance δ_z . (b)–(d) Band structures for different δ_z .

APPENDIX B: BAND STRUCTURE AND TOTAL SPIN MAGNETIZATION IN WSe₂-graphene HB

In this Appendix, we consider the WSe₂-graphene HB. Figures 6(a) and 6(b) show the band structure and LDoS, respectively, for the atomic structure obtained through structural optimization [Fig. 6(c)]. In panel (b), the origin of the z axis (horizontal axis) is set at the position of the tungsten (W) layer. The selenium (Se) layers are located at $z = \pm 1.7$ Å. At $z = 0$, the conduction band minimum and valence band maximum of WSe₂ appear around 0.5 and -1 eV, respectively. The graphene layer is positioned at $z = 5.2$ Å, and its Dirac cone is centered around the Fermi energy ($= 0$). Panels (c), (d), and (e) present snapshots of the density distribution of the spin magnetization [integrand of Eq. (2)] at time $t = 0, 10$, and 20 fs, respectively, under laser irradiation with $I = 10^{12}$ W/cm².

Figure 7 presents the same analysis as Fig. 5, but for the WSe₂-graphene HB. Panels (b) and (c) show the total spin magnetization per unit area of the HB. The behavior of the total spin magnetization is qualitatively similar to that of the WSe₂ monolayer (Fig. 5), but the results at higher intensities exhibit a slight increase due to changes in the density of states of the WSe₂ layer induced by the heterojunction. This DoS change originates mainly from the cell compression of the WSe₂ layer in the xy plane.

In the main text, the spin transfer efficiency from WSe₂ to graphene (ratio of the spin magnetization around the graphene layer to that around the WSe₂ layer) is estimated to be about

0.1 for the case of $T = 20$ fs and $I = 10^{12}$ W/cm², using the pulse shape defined in Eq. (9). Table I lists the spin transfer efficiencies for various combinations of pulse duration T and laser intensity I , where the time average is taken over the interval $[T, T + 20$ fs]. This provides a rough estimate based on the short-time average. In the low-intensity cases with $T = 10$ fs, negative values are observed because the pulse duration is too short to induce sufficient real excitation of the total spin magnetization. For $T > 10$ fs, the spin transfer efficiency remains stably positive and decreases monotonically with increasing intensity. This trend further supports the interpretation that spin transfer arises from Pauli blocking by excited carriers.

APPENDIX C: INTERLAYER DISTANCE DEPENDENCE

Finally, we present first-principles calculations to investigate the dependence of spin transfer on interlayer spacing.

Here, we perform band-structure calculations by varying the interlayer distance. Figure 8(a) shows the binding energy as a function of δ_z , the deviation of the interlayer spacing from the optimal interlayer spacing. We calculated the binding energy per carbon atom between the graphene sheet and the WSe₂ layer using

$$E_b = \frac{E_{\text{WSe}_2+\text{Gra}} - (E_{\text{WSe}_2} + E_{\text{Gra}})}{N_c}, \quad (\text{C1})$$

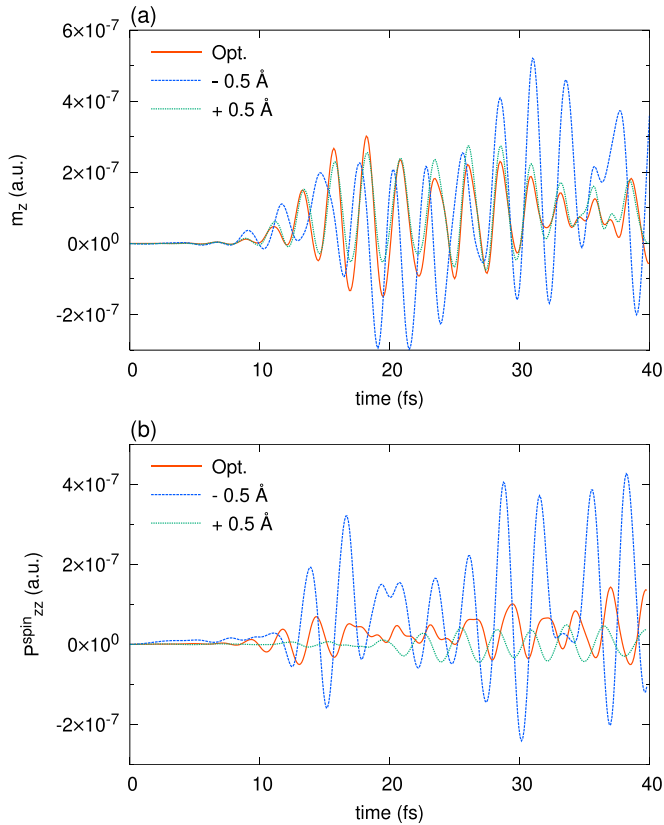


FIG. 9. (a) Total spin magnetization and (b) spin transfer of the HB under laser intensity of $I = 10^9$ W/cm². The optimized, -0.5 Å reduced, and $+0.5$ Å increased interlayer spacing results are plotted as the red, blue, and green lines, respectively.

where $E_{\text{WSe}_2+\text{Gra}}$ is the total energy of the WSe₂-graphene HB, E_{WSe_2} (E_{Gra}) is the total energy of the isolated WSe₂ (graphene) layer, and N_c is the number of carbon atoms in the unit cell. A negative binding energy indicates that the heterostructure is stable.

By changing the interlayer distance (δ_z), we observe significant modifications in the electronic structure. We plot the band structure for the equilibrium interlayer distance, as well as for ± 0.5 Å compression and expansion relative to equilibrium [Figs. 8(b)–8(d)]. We also examined the electronic structure for several in-plane displacements and confirmed that it shows no significant changes.

Figures 9 and 10 show the total spin magnetization [panel (a)] and spin transfer [panel (b)] of the HB under laser in-

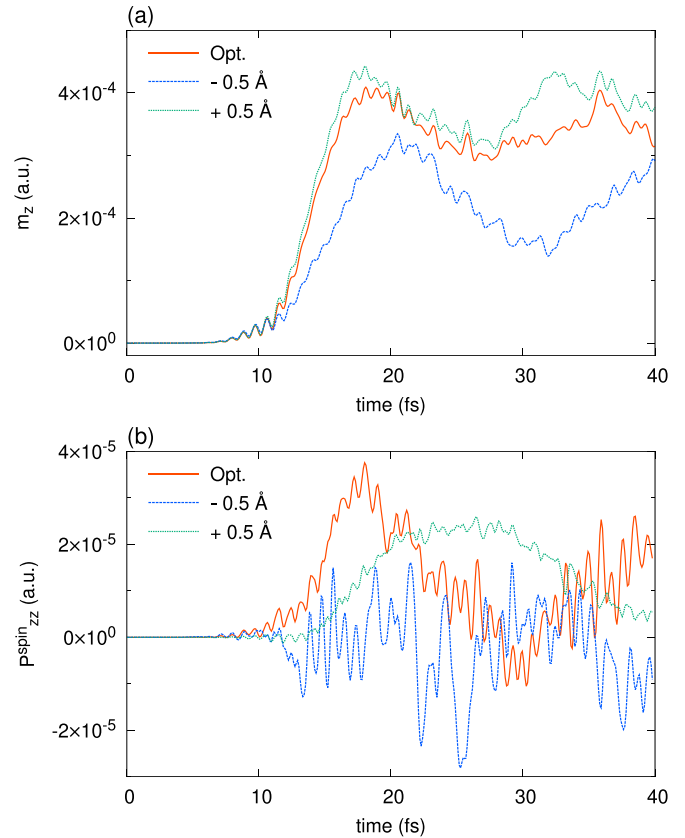


FIG. 10. (a) Total spin magnetization and (b) spin transfer of the HB under laser intensity of $I = 10^{12}$ W/cm². The optimized, -0.5 Å reduced, and $+0.5$ Å increased interlayer spacing results are plotted as the red, blue, and green lines, respectively.

intensities of $I = 10^9$ and 10^{12} W/cm², respectively. The red solid lines in panel (a) [(b)] correspond to the data used in Fig. 7(b) [Fig. 1(c)], representing the structure-optimized HB. The blue dashed and green dotted lines show the results for HBs with the interlayer spacing reduced and increased by 0.5 Å, respectively.

In the low-intensity case [Fig. 9(b)], spin transfer increases with decreasing interlayer spacing. In contrast, the high-intensity case [Fig. 10(b)] shows a decrease in spin transfer under the same compression. This reduction is attributed to the decrease in total spin magnetization [Fig. 10(a)] caused by the reduced spacing. An unrealistically small interlayer spacing may hinder spin excitation in the high-intensity regime.

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