Femtosecond currents in transition metal dichalcogenides monolayers

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(Dated: November 17, 2020)

We theoretically study the interaction of an ultrafast intense linearly polarized optical pulse with monolayers of transition metal dichalcogenides (TMDCs). Such a strong pulse redistributes electrons between the bands and generates femtosecond currents during the pulse. Due to the large bandwidth of the incident pulse, this process is completely off-resonant. While in TMDCs the time-reversal symmetry is conserved, the inversion symmetry is broken and these monolayers have the axial symmetry along armchair direction but not along the zigzag one. Therefore, the pulse polarized along the asymmetric direction of TMDC monolayer generates both longitudinal, i.e., along the direction of polarization, and transverse, i.e., in the perpendicular direction, currents. Such currents result in charge transfer through the system. We study different TMDC materials and show how the femtosecond transport in TMDC monolayers depend on their parameters, such as lattice constant and bandgap.

I. INTRODUCTION

Nowadays, the femtosecond and strong field driven phenomena, e.g., high harmonic generations, the ultrafast ionization and metalization, the nonlinear current generations, and the nonlinear optical absorption in solids attract growing interest due to their possible applications in ultrafast optical switches, optoelectronic devices and ultimately in ultrafast information processing[12]. Among solids, transition metal dichalcogenides (TMDC) have unique optical and electrical properties. The bulk TMDCs are stacks of monolayers, which are bounded by the van der Waals forces[29,30]. Due to natural weakness of these forces, the bulk can be easily exfoliated to atomically thin monolayers[31]. Each monolayer is made of one layer of transition metal atoms like Mo and W, which is sandwiched between two chalco- gen (S, Se, Te) layers. The monolayers can be found in different phases, while the semiconducting phase is the most common one. It has trigonal prismatic crystalline structures with $D_{3h}$ point symmetry group[32].

The TMDC monolayers are direct bandgap semiconductors with the bandgaps of 1.1-2.1 eV[32]. Similar to graphene, TMDC monolayers have honeycomb crystal structure but they are not centrosymmetric and the inversion symmetry is broken. Due to the broken inversion symmetry, the Berry curvature is not singular but has finite values with opposite signs in two valleys, $K$ and $K'$. The finite Berry curvature gives rise to an anomalous Hall effect in the absence of external magnetic field[33]. Another difference of these materials from graphene is the existence of strong intrinsic spin orbit coupling[32] which results in the spin splitting of the valence band (VB) and the conduction band (CB)[32] and makes TMDC monolayers suitable for spintronic applications.

Previously, we have shown that a single cycle of a circularly polarized optical pulse induces a large valley polarization, $\eta_v \geq 40\%-60\%$, in TMDC monolayers, MoS$_2$ and WS$_2$[32]. The mechanism of producing fundamentally fastest valley polarization in these monolayers is independent of electron spin and has topological origin. Predominant population of one of the valleys in TMDC monolayer is not due to the optical selection rule as in case of a continuous wave but due to topological resonance, which is a competition of the dynamic phase and the topological phase that is accumulated during ultrashort and strong pulse[32]. It has been also recently predicted that the valley polarization can be tuned by the bandgap in gapped graphene monolayers[32]. In graphene, the inversion symmetry can be broken by placing graphene on different substrates, e.g., SiC, which reduces the point group symmetry of graphene from $D_{6h}$ to $D_{3h}$[34,35].

In the field of intense optical pulse the valence and conduction band states are strongly coupled, which results in generation of strong nonlinear electric currents and finite transfer of electric charge through the system. Thus ultrafast optical pulses allow to control the transport properties of electron systems and enhance the conductivity of solids on the femtosecond time scale. Understanding of the extent of such control is important for possible device application of different solids. In this article we study the femtosecond currents driven by a single-cycle of an intensive laser pulse in monolayers of different TMDC materials. Different characteristics, e.g., the energy dispersion and the lattice constants, of these materials strongly affect the generated electric current and correspondingly transferred charge. The generated electric current also depends on the direction of polarization of the optical pulse[37].

II. MAIN EQUATIONS

We consider coherent ultrafast electron dynamics in the field of the pulse, assuming that the relaxation and scattering times in TMDC monolayers are longer than 10 fs[38,39]. The time dependent Hamiltonian of the system has the following form

$$ H(t) = H_0 - e\mathbf{rF}(t), $$

(1)
where \( u(TDSE) \) is the corresponding time dependent Schrödinger equation and \( \hat{H} \) is the Hamiltonian. The electric field in the pulse has both \( x \) and \( y \) components, \( F_x \) and \( F_y \).

where \( H_0 \) is the field-free Hamiltonian of a TMDC monolayer, \( e \) is an electron charge, \( \mathbf{r} \) is the position vector, and \( \mathbf{F}(t) \) is the electric field of the pulse. We consider the three-band tight binding model for TMDC monolayer, which gives three bands: one valence band and two conduction bands. For the pulse, linearly polarized in the \( x \)-direction, the electric field is given by the following expression

\[
F_x(t) = F_0(1 - 2u^2)e^{-u^2}, \quad F_y(t) = 0, \quad (2)
\]

where \( u = t/\tau \), and \( \tau = 1 \) fs is the pulse duration.

The electron dynamics is determined by the solution of the corresponding time dependent Schrödinger equation (TDSE)

\[
\frac{ih}{\hbar}\frac{d\Psi(t)}{dt} = H(t)\Psi(t), \quad (3)
\]

It is convenient to express this solution in the basis of time dependent Houston functions\(^{23}\)

\[
\phi_{\alpha \mathbf{k}}^{\mathbf{q}}(\mathbf{r}, t) = \Psi_{\mathbf{k}; \mathbf{q}, \alpha}^{(\alpha)}(\mathbf{r}) \exp\left(i\phi_{\alpha}^{(D)}(q, t) + i\phi_{\alpha}^{(B)}(q, t)\right), \quad (4)
\]

where \( \Psi_{\mathbf{k}; \mathbf{q}, \alpha}^{(\alpha)}(\mathbf{r}) \) are the eigenfunctions of the time independent part of Hamiltonian \( H_0 \) and \( \phi_{\alpha}^{(D)}(q, t) = \frac{1}{\hbar}\int dt'E_{\alpha}[\mathbf{k}(\mathbf{q}, t')] \) is the dynamic phase, \( E_{\alpha} \) are the eigenvalues of \( H_0 \), \( \phi_{\alpha}^{(B)}(q, t) = \frac{1}{\hbar}\int dt'\mathbf{F}(t')A_{\alpha \alpha}[\mathbf{k}(\mathbf{q}, t')] \) is the Berry phase, \( A_{\alpha \alpha} \) is the Berry connection, which is defined below by Eq. \( (4) \), and \( \alpha \in \{ \upsilon, c_1, c_2 \} \) where \( \upsilon, c_1, c_2 \) denote the VB and two CBs, respectively. The electron trajectory in the reciprocal space, \( \mathbf{k}(\mathbf{q}, t) \), is given by the Bloch acceleration theorem\(^{24}\)

\[
\mathbf{k}(\mathbf{q}, t) = \mathbf{q} + \frac{e}{\hbar} \int_{-\infty}^{t} \mathbf{F}(t')dt' \quad (5)
\]

where \( \mathbf{q} \) is the initial crystal wave vector.

In the basis of Houston functions, solutions of the time dependent Schrödinger equation \( (3) \) are parameterized by initial crystal wave vector \( \mathbf{q} \) and are given by the following expression

\[
\Psi_{\mathbf{q}}(\mathbf{r}, t) = \sum_{\alpha = \upsilon, c_1, c_2} \beta_{\alpha \mathbf{q}}(t)\Psi_{\mathbf{q}, \alpha}(\mathbf{r}, t), \quad (6)
\]

where \( \beta_{\alpha \mathbf{q}}(t) \) are expansion coefficients, which satisfy the following system of differential equations

\[
\frac{i\hbar}{\hbar}\frac{\partial B_{\mathbf{q}}(t)}{\partial t} = H'(\mathbf{q}, t)B_{\mathbf{q}}(t). \quad (7)
\]

Here

\[
B_{\mathbf{q}}(t) = \begin{bmatrix} \beta_{\upsilon \mathbf{q}}(t) \\ \beta_{c_1 \mathbf{q}}(t) \\ \beta_{c_2 \mathbf{q}}(t) \end{bmatrix}, \quad (8)
\]

\[
H'(\mathbf{q}, t) = -e\mathbf{F}(t)\mathbf{A}(\mathbf{q}, t), \quad (9)
\]

\[
\mathbf{A}(\mathbf{q}, t) = \begin{bmatrix} 0 & \mathcal{D}_{c_1 \upsilon}(\mathbf{q}, t) & \mathcal{D}_{c_2 \upsilon}(\mathbf{q}, t) \\ \mathcal{D}^\ast_{c_1 \upsilon}(\mathbf{q}, t) & 0 & \mathcal{D}^\ast_{c_2 \upsilon}(\mathbf{q}, t) \\ \mathcal{D}^\ast_{c_1 \upsilon}(\mathbf{q}, t) & \mathcal{D}^\ast_{c_2 \upsilon}(\mathbf{q}, t) & 0 \end{bmatrix}. \quad (10)
\]

where

\[
\mathcal{D}_{\alpha \alpha}(\mathbf{q}, t) = \mathcal{A}_{\alpha \alpha}[\mathbf{k}(\mathbf{q})] = \left( \Psi_{\mathbf{q}}^{(\alpha)} | \frac{\partial}{\partial \mathbf{q}} | \Psi_{\mathbf{q}}^{(\alpha)} \right), \quad (14)
\]

Here, \( \mathcal{A}_{\alpha \alpha}(\mathbf{k}) \) is the non-Abelian Berry connection\(^{25,26}\), and \( \mathcal{D}_{\alpha \alpha} \) is the interband dipole matrix, which determines the optical transitions between the VB and CBs.

The crystal structure of TMDC monolayer is shown in Fig. 1. It has \( D_{3h} \) symmetry and consists of two sublattices \( A \) and \( B \), which are occupied by transition metal atoms (sublattice \( A \)) and chalcogen atoms (sublattice \( B \)). The first Brillouin zone of TMDC monolayer is a hexagon with two valleys, \( K \) and \( K' \) - see Fig. 1(c). We describe TMDC monolayer within the three band tight binding model\(^{22}\). In this model only the couplings between the nearest neighbor \( d \) orbitals \( (d_{xy}, d_{xz}, \text{and } d_{x^2-y^2}) \) of transition metal atoms are considered. The corresponding Hamiltonian is the sum of the nearest neighbor tight-binding Hamiltonian \( H^{(TNN)} \), and spin orbit coupling (SOC) contribution \( H^{(SOC)} \)

\[
H_0(\mathbf{k}) = I \otimes H^{(TNN)} + H^{(SOC)} = \begin{bmatrix} H^{(TNN)}(\mathbf{k}) + \frac{1}{2} \mathcal{L}_z & 0 \\ 0 & H^{(TNN)}(\mathbf{k}) - \frac{1}{2} \mathcal{L}_z \end{bmatrix}
\]

\[
= \begin{bmatrix} H_{3\times3}^{(TNN)}(\mathbf{k}) & 0 \\ 0 & H_{3\times3}^{(SOC)}(\mathbf{k}) \end{bmatrix}, \quad (15)
\]
TABLE I. Lattice constant, spin orbit coupling constant, and bandgap (for spin up and spin down) at the $K$ and $K'$ points for different TMDC monolayers.\(^{32}\)

| Material | $a$ (Å) | $\lambda$ (eV) | $\Delta_{K}^{Up}$ (eV) | $\Delta_{K}^{Down}$ (eV) |
|----------|--------|----------------|------------------|------------------|
| MoS\(_2\) | 3.19 | 0.073 | 1.590 | 1.736 |
| WS\(_2\) | 3.191 | 0.211 | 1.600 | 2.023 |
| MoSe\(_2\) | 3.326 | 0.091 | 1.346 | 1.590 |
| WSe\(_2\) | 3.325 | 0.228 | 1.325 | 0.967 |
| MoTe\(_2\) | 3.557 | 0.237 | 0.211 | 3.190 |
| WTe\(_2\) | 3.560 | 0.107 | 0.835 | 1.180 |

where the tight binding matrix $H^{(TNN)}$ is given in Appendix A, $\lambda$ is the SOC constant,\(^{32}\) and

$$L_z = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 2i \\ 0 & -2i & 0 \end{bmatrix}.$$  \hspace{1cm} (16)

Since, the two spin components are not coupled by external electric field, we solve TDSE for each spin component independently.

The main parameters of TMDC monolayers, which are the bandgap, lattice constant, and SOC constant, are shown in Table I. The lattice constant is in the range of 3.19 - 3.56 Å, while the bandgap lies between 0.8 eV and 2.0 eV.

The femtosecond field-driven currents in solids generally have two main contributions, which come from intraband and interband dynamics. While these contributions are not gauge invariant quantities, the total current is given by the following expression

$$J_{tot} = J_{interband} + J_{intraband},$$

where $J_{intraband}$ and $J_{interband}$ are the intraband and interband currents, respectively.

$$J_{intraband}(t) = \frac{e}{\hbar^2} \sum_{\gamma} \sum_{\alpha} \frac{\partial}{\partial t} E_{\alpha \gamma}(\mathbf{k}) v_{\alpha \gamma}(\mathbf{k}, t),$$

$$J_{interband}(t) = \frac{i e}{\hbar a^2} \sum_{\gamma} \sum_{\alpha, \alpha'} \beta_{\alpha' \alpha \gamma}(\mathbf{k}, t) \beta_{\alpha \gamma}(\mathbf{k}, t) \times \exp\{i \phi_{\alpha' \alpha \gamma}(\mathbf{k}, t) + i \phi_{\alpha \gamma}(\mathbf{k}, t)\} \times [E_{\alpha' \gamma}(\mathbf{k}(\mathbf{q}, t)) - E_{\gamma}(\mathbf{k}(\mathbf{q}, t))] A_{\alpha' \alpha \gamma}(\mathbf{k}(\mathbf{q}, t)).$$

$$\hspace{1cm} (17)$$

III. RESULTS AND DISCUSSION

Below we consider the following TMDC materials: MoS\(_2\), WS\(_2\), MoSe\(_2\), WSe\(_2\), MoTe\(_2\), and WTe\(_2\). The parameters for these materials are taken from Ref.\(^{32}\) The crystal structure of the corresponding TMDC monolayer with the first Brillouin zone is shown Fig. 1. Within the three band tight binding model the monolayer has one valence band (VB) and two conduction bands (CBs). Initially, i.e., before the pulse, the valence band is occupied and the conduction bands are empty. We apply a linearly polarized pulse propagating along $z$ direction with the amplitude of $\sim 0.1 - 0.5$ VÅ\(^{-1}\) and the duration of $\sim 5$ fs.

One of the characteristics of electron dynamics in the field of the pulse is CB population distribution in the reciprocal space, $N_{CB}(\mathbf{k}) = |\beta_{C_{\alpha \gamma}}(\mathbf{k})|^2$. Such distribution is nonzero during the pulse and its residual value, $N_{CB}^{(res)}(\mathbf{k})$, determines the valley polarization distribution after a circularly polarized pulse.

Typical for TMDC monolayers, the residual CB population distribution in the reciprocal space is shown in Fig. 2 for two spin components, down (a) and up (b). The pulse is linearly polarized in $x$-direction with the amplitude of 0.25 VÅ\(^{-1}\). The CB population is large near the $K$ and $K'$ valleys, which is due to large interband coupling at these two points. For such small field amplitude, the population distribution does not show any interference fringes. For a given spin component, up or down, one valley is more populated than another one. For example, for spin down (see Fig. 2(a)), the CB population of $K'$ valley is higher than the corresponding population of the $K$ valley. However, the total CB population, summed over both spin components, is the same for both valleys. This is because the linear polarized pulse preserves the
FIG. 3. (Color online) The same as Fig. 2 but for the pulse polarized in the $y$ direction.

time reversal symmetry and does not induce any valley polarization. The axis $x$ is not the axis of symmetry of TMDC monolayer and the residual CB population distribution, shown in Fig. 2, clearly shows such asymmetry. Because the CB population distribution is not symmetric with respect to the $x$ axis, the electric current is generated in both $x$ and $y$ directions.

The CB population distribution for the applied pulse polarized in $y$ direction is shown in Fig. 3. Similar to the $x$ polarized pulse, the CB population is concentrated near the $K$ and $K'$ valleys with zero residual valley polarization. The $y$ axis is the axis of symmetry of the system and the CB population distribution is symmetric with respect to the $y$ axis. Because of this symmetry, the electric current is generated during the pulse only in $y$ direction.

The ultrafast field driven intraband and interband electron dynamics generates an electric current. For the pulse polarized in $x$ direction, i.e., along the zigzag direction, the electric current is generated in both $x$ and $y$ directions. The current along $y$ direction, i.e., the direction perpendicular to the polarization of the pulse, strongly depends on the bandgap of TMDC monolayer. It disappears at zero bandgap, e.g., for pristine graphene, when the $x$ axis is the axis of symmetry. For the pulse polarized in $y$ axis, which is the axis of symmetry of TMDC monolayer, the electric current is generated only along the direction of polarization of the pulse. Below we consider only the electric pulse polarized in $x$ direction, which covers electron transport both in the direction of the pulse polarization and in the perpendicular direction.

The generated electric currents for different TMDC materials are shown in Fig. 4. The pulse is linearly polarized in the $x$ direction and its amplitude is $0.25$ V/Å. The $x$ component of the current for all TMDC materials shows the same profile during the pulse, i.e., $-2fs < t < 2fs$, but after the pulse, $J_x$ has oscillatory behavior with the frequency of oscillations that depends on the bandgap of TMDC monolayer, which is in the range of $0.8 - 2$ eV for the TMDC materials shown in the figure. Such oscillations in the residual current $J_x$ is due to the fact that the main contribution to the current is the interband one, while the intraband contribution, which depends only on the population of the conduction and valence bands and thus do not show oscillation after the pulse, is small.

The generated current in the $y$ direction is almost three times smaller than the current in the $x$ direction. It also shows the oscillatory behavior as a function of time with well pronounced bandgap-dependent oscillations after the pulse. Although the profile of current $J_y$ during the pulse ($-2fs < t < 2fs$) is almost the same for all TMDC monolayers, one TMDC material, namely MoTe$_2$, shows completely different time dependence.

The unique behavior of MoTe$_2$ monolayer can be understood from the corresponding CB population distribution in the reciprocal space. The residual CB populations are shown in Fig. 5 for different TMDC monolayers. For all monolayers except MoTe$_2$, $N_{\text{CB}}(\mathbf{k})$ is concentrated at the $K$ and $K'$ points along both $k_x$ and $k_y$ directions. As a result they all show the same time dependence of the generated current for both $x$ and $y$ directions. At the same time, for MoTe$_2$ monolayer, the CB population distribution is completely different. While along the direc-
FIG. 5. (Color online) Residual CB population distributions in the reciprocal space for different TMDC monolayers: (a) MoS$_2$, (b) MoSe$_2$, (c) MoTe$_2$, (d) WS$_2$, (e) WSe$_2$, and (f) WTe$_2$. The optical pulse is linearly polarized in the $x$ direction and its amplitude is $0.25 \text{ V} \mu\text{A}^{-1}$. The black solid lines show the edges of the first Brillouin zone. For all TMDC monolayers, except MoTe$_2$, the CB population is concentrated near the $K$ and $K'$ points.

The dependence of the electric current on the field amplitude, $F_0$, is shown in Fig. 6 for MoS$_2$ monolayer. For other TMDC materials the dependence on $F_0$ is similar. As expected, the generated current monotonically increases with $F_0$. In residual current, the frequency of oscillations, which is determined by the bandgap, does not depend on $F_0$.

One of the characteristics of nonlinearity of electron response to an ultrashort pulse is a transferred charge through the system during the pulse, which can be also measured experimentally. The transferred charge is defined by the following expression

$$Q = \int_{-\infty}^{\infty} J(t)dt'.$$

Since the residual current shows an oscillating behavior, to eliminate the dependence on the upper limit in the above integral we introduce a relaxation time of 5 fs when calculating the transferring charge. The transferred charge is also the residual polarization of the system.

The transferred charge as a function of the field amplitude, $F_0$, is shown in Fig. 7 for different TMDC monolayers. The transferred charge along the $y$ direction, $Q_y$, monotonically increases with $F_0$ - see Fig. 7(a). For all TMDC monolayers, except MoTe$_2$, the charge is transferred in the positive direction of the $y$ axis, while for MoTe$_2$ the transfer of the charge occurs in the negative direction of the $y$ axis. Such direction of the transfer corresponds to the condition that the field maximum of the pulse is in the positive direction of the $x$ axis. The magnitude of the transferred charge increases with decreasing the bandgap of TMDC monolayer. The largest charge
FIG. 6. (Color online) Ultrafast field driven currents in MoS$_2$ monolayer as a function of time for different field amplitudes. The $y$ component (a) and the $x$ component (b) of the current are shown. The optical pulse is linearly polarized in the $x$ direction.

transfer occurs for WTe$_2$ monolayer, while the smallest - for MoTe$_2$ monolayer.

Along the $x$ axis [see Fig. 7(b)], the charge is transferred in the direction of the field maximum for all TMDC monolayers. The dependence of $Q_x$ on the pulse amplitude, $F_0$, is nonmonotonic. The transferred charge reaches its maximum at some value of $F_0 = F_{\text{max}}$ and then decreases with $F_0$. The value of $F_{\text{max}}$ is partially correlated with the condition that at this field an electron, which is initially at one valley, say valley $K$, reaches the second valley, $K'$, during the pulse. For example, for TMDC monolayers with large lattice constants, MoTe$_2$: 3.557 Å and WTe$_2$: 3.560 Å, the maxima occur at lower field amplitudes. Although the lattice constant is not the only parameter, which determines $Q_x$ dependence on $F_0$, the transferred charge also depends on the bandgap and spin orbit coupling of TMDC monolayer. In terms of applications, the data in Fig. 7(b) illustrate that MoTe$_2$ monolayer is the most sensitive to the pulse amplitude, i.e., for MoTe$_2$ monolayer, the transferred charge, $Q_x$, shows relatively sharp maximum with strong dependence on $F_0$. 

FIG. 7. (Color online) Charge transferred through the system during the pulse as a function of the field amplitude, $F_0$, for different TMDC monolayers. The transferred charge along the $y$ direction (a) and the $x$ direction (b) is shown. The optical pulse is linearly polarized in the $x$ direction.
IV. CONCLUSION

The ultrafast field driven currents in solids are governed by interband and intraband electron dynamics, resulting in two contributions, interband and interband, to the electric current. In TMDC monolayers, the generated electric current is mainly determined by the interband contribution. As a result, the residual current as a function of time shows oscillations, the frequency of which is determined by the bandgap of the corresponding TMDC monolayer. The TMDC monolayers have broken inversion symmetry, their symmetry group is $D_{3h}$, and they have only three axises of symmetry, which are along the armchair directions. If the optical pulse is polarized along the direction of symmetry of the monolayer then the electric current is generated only along the direction of polarization. But if the polarization of the optical pulse is along a non-symmetric direction, for example, along the zigzag direction, then the electric current is generated both along the direction of polarization and in the perpendicular direction.

For all TMDC monolayers the longitudinal electric current, i.e., the current along the direction of polarization of the pulse, show similar behavior as a function of time. Such current transfers an electric charge along the direction of the field maximum. As a function of the field amplitude, the transferred charge has a maximum, the position of which depends on the lattice constant of TMDC monolayer. MoTe$_2$ monolayer is the most sensitive to the parameters of the optical pulse.

The transverse current also results in the charge transfer through the system during the pulse. The magnitude of the transferred charge monotonically increases with the field amplitude, while the direction of the transfer depends on the TMDC material. Control of an electron transport on a femtosecond time scale pave the way for ultrafast electronic application of TMDCs monolayers.

\[ V_0 = \epsilon_1 + 2t_0(2 \cos \alpha \cos \beta + \cos 2\alpha) \]
\[ + 2r_0(2 \cos 3\alpha \cos \beta + \cos 2\beta) \]
\[ + 2u_0(2 \cos 2\alpha \cos 2\beta + \cos 4\alpha), \]
\[ \text{Re}[V_1] = -2\sqrt{3}t_2 \sin \alpha \sin \beta + 2(r_1 + r_2) \sin 3\alpha \sin \beta \]
\[ - 2\sqrt{3}u_2 \sin 2\alpha \sin 2\beta, \]
\[ \text{Im}[V_1] = 2t_1 \sin \alpha (2 \cos \alpha + \cos \beta) \]
\[ + 2(r_1 - r_2) \sin 3\alpha \cos \beta \]
\[ + 2u_1 \sin 2\alpha (2 \cos 2\alpha + \cos 2\beta), \]
\[ \text{Re}[V_2] = 2t_2(\cos 2\alpha - \cos \alpha \cos \beta) \]
\[ - \frac{2}{\sqrt{3}}(r_1 + r_2)(\cos 3\alpha \cos \beta - \cos 2\beta) \]
\[ + 2u_2(\cos 4\alpha - \cos 2\alpha \cos 2\beta), \]
\[ \text{Im}[V_2] = 2\sqrt{3}t_1 \cos \alpha \sin \beta \]
\[ + \frac{2}{\sqrt{3}} \sin \beta (r_1 - r_2)(\cos 3\alpha + 2\cos \beta) \]
\[ + 2\sqrt{3}u_1 \cos 2\alpha \sin 2\beta, \]
\[ V_{11} = \epsilon_2 + (t_{11} + 3t_{12}) \cos \alpha \cos \beta \]
\[ + 2t_{11} \cos 2\alpha + 4r_{11} \cos 3\alpha \cos \beta \]
\[ + 2(r_{11} + \sqrt{3}r_{12} \cos 2\beta) \]
\[ + (u_{11} + 3u_{12}) \cos 2\alpha \cos 2\beta + 2u_{11} \cos 4\alpha, \]
\[ \text{Re}[V_{12}] = \sqrt{3}(t_{12} - t_{11}) \sin \alpha \sin \beta \]
\[ + 4r_{12} \sin 3\alpha \sin \beta \]
\[ + \sqrt{3}(u_{12} - u_{11}) \sin 2\alpha \sin 2\beta, \]
\[ \text{Im}[V_{12}] = 4t_{12} \sin \alpha (\cos \alpha - \cos \beta) \]
\[ + 4u_{12} \sin 2\alpha (\cos \alpha - \cos 2\beta), \]
\[ V_{22} = \epsilon_2 + (3t_{11} + t_{12}) \cos \alpha \cos \beta + 2t_{22} \cos 2\alpha \]
\[ + 2r_{11} (2 \cos 3\alpha \cos \beta + \cos 2\beta) \]
\[ + \frac{2}{\sqrt{3}}r_{12} (4 \cos 3\alpha \cos \beta - \cos 2\beta) \]
\[ + (3u_{11} + u_{12}) \cos 2\alpha \cos 2\beta + 2u_{22} \cos 4\alpha, \]
\[ (A2) \]

and
\[ (\alpha, \beta) = \left( \frac{1}{2} k_x a, \frac{\sqrt{3}}{2} k_y a \right). \]
\[ (A3) \]

The values of the parameters in the above Hamiltonian for different TMDC materials are given in table Ref 32.

ACKNOWLEDGMENTS

Major funding was provided by Grant No. DE-FG02-01ER15213 from the Chemical Sciences, Biosciences and Geosciences Division, Office of Basic Energy Sciences,
TABLE II. Parameters of three band tight-binding Hamiltonian. Here the lattice constant, $a$, is in units of Å, while all other parameters are in units of eV.

|       | MoS$_2$ | WS$_2$ | MoSe$_2$ | WSe$_2$ | MoTe$_2$ | WTe$_2$ |
|-------|---------|--------|----------|---------|----------|---------|
| $a$   | 3.19    | 3.191  | 3.326    | 3.325   | 3.557    | 3.560   |
| $\epsilon_1$ | 0.683  | 0.717  | 0.684    | 0.728   | 0.588    | 0.697   |
| $\epsilon_2$ | 1.707  | 1.916  | 1.546    | 1.655   | 1.303    | 1.380   |
| $t_0$  | -0.146  | -0.152 | -0.146   | -0.146  | -0.226   | -0.109  |
| $t_1$  | -0.114  | -0.097 | -0.130   | -0.124  | -0.234   | -0.164  |
| $t_2$  | 0.506   | 0.590  | 0.432    | 0.507   | 0.036    | 0.368   |
| $t_{11}$ | 0.085  | 0.047  | 0.144    | 0.117   | 0.400    | 0.204   |
| $t_{12}$ | 0.162  | 0.178  | 0.117    | 0.127   | 0.098    | 0.093   |
| $t_{22}$ | 0.073  | 0.016  | 0.075    | 0.015   | 0.017    | 0.038   |
| $r_0$  | 0.06    | 0.069  | 0.039    | 0.036   | 0.003    | -0.015  |
| $r_1$  | -0.236  | -0.264 | -0.209   | -0.234  | -0.025   | -0.269  |
| $r_{11}$ | 0.016  | -0.003 | 0.052    | 0.044   | 0.082    | 0.115   |
| $r_{12}$ | 0.087  | 0.109  | 0.066    | 0.075   | 0.054    | 0.009   |
| $r_2$  | 0.067   | 0.107  | 0.069    | 0.107   | -0.109   | 0.107   |
| $u_0$  | -0.038  | -0.054 | -0.012   | -0.061  | 0.057    | -0.066  |
| $u_1$  | 0.046   | 0.045  | 0.036    | 0.032   | 0.103    | 0.011   |
| $u_2$  | 0.001   | 0.002  | 0.008    | 0.007   | 0.187    | -0.013  |
| $u_{11}$ | 0.266  | 0.325  | 0.272    | 0.329   | -0.045   | 0.312   |
| $u_{12}$ | -0.176 | -0.206 | -0.172   | -0.202  | -0.141   | -0.177  |
| $u_{22}$ | -0.15  | -0.163 | -0.150   | -0.164  | 0.087    | -0.132  |
| $\lambda$ | 0.073  | 0.211  | 0.091    | 0.228   | 0.107    | 0.237   |

Office of Science, US Department of Energy. Numerical simulations were performed using support by Grant No. DE-SC0007043 from the Materials Sciences and Engineering Division of the Office of the Basic Energy Sciences, Office of Science, US Department of Energy.

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