# Supplemental Material for: Substrate Effects on Spin Relaxation in Two-Dimensional Dirac Materials with Strong Spin-Orbit Coupling

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## SI. COMPUTATIONAL DETAILS

The ground-state electronic structure, phonons, as well as electron-phonon and electron-impurity (e-i) matrix elements are firstly calculated using density functional theory (DFT) with relatively coarse $k$ and $q$ meshes in the DFT plane-wave code JDFTx[1]. Since all substrates have hexagonal structures and their lattice constants are close to germanene's, the heterostructures are built simply from unit cells of two systems. The lattice mismatch values are within 1% for Ge-GeH, Ge-GaTe and Ge-InSe but about 3.5% for Ge-SiH. All heterostuctures use the lattice constant 4.025 Å of free-standing Ge relaxed with Perdew-Burke-Ernzerhof exchange-correlation functional[2]. The internal geometries are fully relaxed using the DFT+D3 method for van der Waals dispersion corrections[3]. We use Optimized Norm-Conserving Vanderbilt (ONCV) pseudopotentials[4] with self-consistent spin-orbit coupling throughout, which we find converged at a kinetic energy cutoff of 44, 64, 64, 72 and 66 Ry for free-standing Ge, Ge-GeH, Ge-SiH, Ge-GaTe and Ge-InSe respectively. The DFT calculations use 24×24 $k$ meshes. The phonon calculations employ $3 \times 3$ supercells through finite difference calculations. We have checked the supercell size convergence and found that using $6 \times 6$ supercells lead to very similar results of phonon dispersions and spin lifetimes. For all systems, the Coulomb truncation technique is employed to accelerate convergence with vacuum sizes.

The e-i matrix $g^i$ between state $(k, n)$ and $(k', n')$ is

$$g^i_{kn,k'n'} = \langle kn| \Delta V^i |k'n'\rangle , \tag{S1}$$

$$\Delta V^i = V^i - V^0, \tag{S2}$$

where $V^i$ is the potential of the impurity system and $V^0$ is the potential of the pristine system. $V^i$ is computed with SOC using a large supercell including a neutral impurity. To speed up the supercell convergence, we used the potential alignment method developed in Ref. 5. We use 5×5 supercells, which have shown reasonable convergence (a few percent error of the spin lifetime).

We then transform all quantities from plane wave basis to maximally localized Wannier function basis[6], and interpolate them[7–12] to substantially finer k and q meshes. The fine $k$ and $q$ meshes are $384 \times 384$ for simulations at 300 K and are finer at lower temperature, e.g., $1440 \times 1440$ for simulations at 50 K.

The real-time dynamics simulations are done with our own developed DMD code interfaced to JDFTx. The energy-conservation smearing parameter $\sigma$ is chosen to be comparable or smaller than $k_B T$ for each calculation.

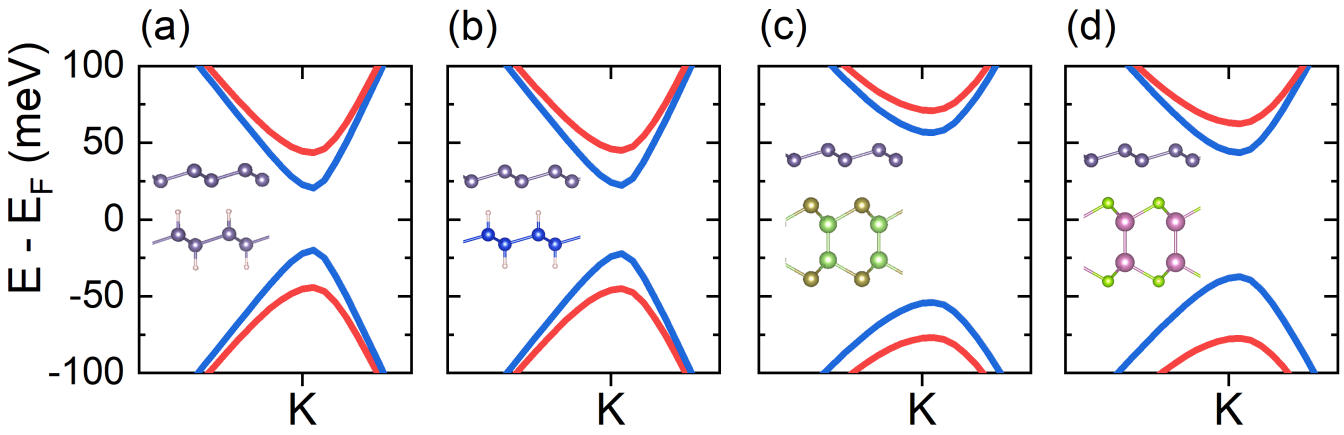


FIG. S1. Band structures around a Dirac cone of four supported germanene (Ge) - (a) Ge-GeH, (b) Ge-SiH, (c) Ge-GaTe and (d) Ge-InSe. The red and blue bands correspond to spin-up and spin-down states.

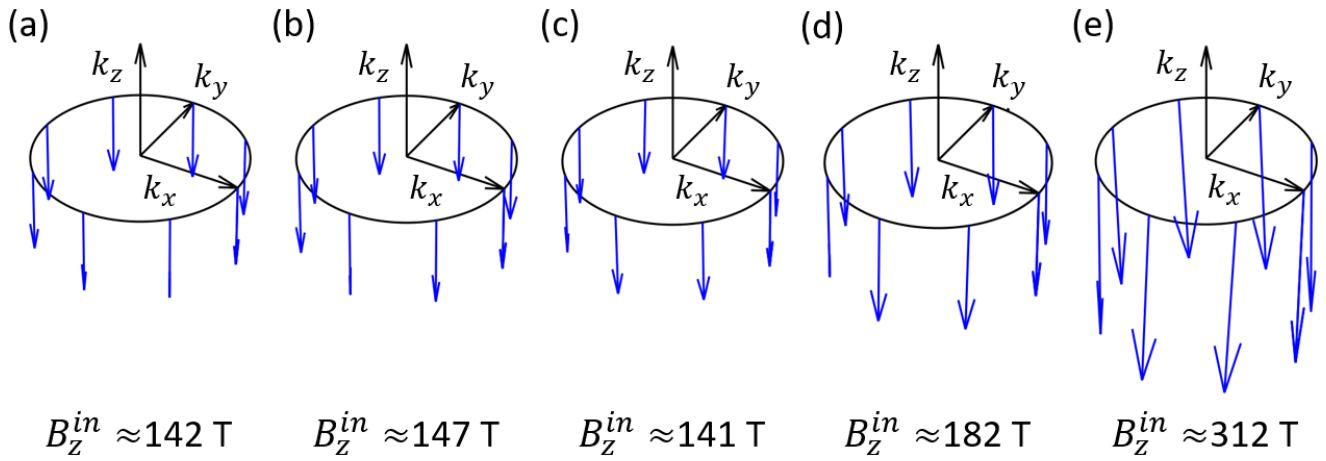


FIG. S2. 3D plots of internal magnetic fields $\mathbf{B}^{\text{in}}$ on the circle $|\overrightarrow{k}| = 0.0025$ bohr$^{-1}$ of the lowest conduction band around high-symmetry point $K$ of (a) Ge@-7V/nm, (b) Ge-GeH, (c) Ge-SiH, (d) Ge-GaTe and (e) Ge-InSe. The magnitudes of $z$-component of $\mathbf{B}^{\text{in}}$ - $B_z^{\text{in}}$ are also given in the unit of tesla (T). $\mathbf{B}_{kn}^{\text{in}} = 2\Delta_{kn}\mathbf{S}_{kn}^{\text{exp}}/(g_e\mu_B)$, where $k$ and $n$ are k-point and band indices, respectively. $g_e\mu_B$ is the electron spin gyromagnetic ratio. $\Delta_{kn}$ is energy difference between two conduction (valence) bands when $n$ is a conduction (valence) band. $\mathbf{S}^{\text{exp}}$ is a vector and $\mathbf{S}^{\text{exp}} \equiv \left(S_x^{\text{exp}}, S_y^{\text{exp}}, S_z^{\text{exp}}\right)$, where $S_i^{\text{exp}}$ is spin expectation value along direction $i$ and is the diagonal element of spin matrix $s_i$ in Bloch basis. The arrow length scales with the magnitude of $\mathbf{B}^{\text{in}}$.

## SII. SPIN MIXING PARAMETER

Suppose the spin of a state "1" (which is the combined index of k-point and band) is highly polarized along $z$ direction. Then in general, the wavefunction of state "1" can be written as $\Psi_1(\mathbf{r}) = a_{z,1}(\mathbf{r})\,\alpha + b_{z,1}(\mathbf{r})\,\beta$, where $a$ and $b$ are the coefficients of the large and small components of the wavefunction, and $\alpha$ and $\beta$ are spinors (one up and one down for direction $z$). Define $a_{z,1}^2 = \int |a_{z,1}(\mathbf{r})|^2 d\mathbf{r}$ and $b_{z,1}^2 = \int |b_{z,1}(\mathbf{r})|^2 d\mathbf{r}$, then $a_{z,1}^2 > b_{z,1}^2$ and $b_{z,1}^2$ is just spin-mixing parameter of state "1" along direction $z$. With the definition of spin expectation value $S_{z,1}^{\text{exp}} = s_{z,11}$, which is the diagonal element for state "1" of spin Pauli matrix along direction $z$ in Bloch basis, we have

$$a_{z,1}^2 + b_{z,1}^2 = 1, \tag{S3}$$

$$0.5\left(a_{z,1}^2 - b_{z,1}^2\right) = S_{z,1}^{\text{exp}}, \tag{S4}$$

Therefore,

$$b_{z,1}^2 = 0.5 - S_{z,1}^{\text{exp}}. \tag{S5}$$

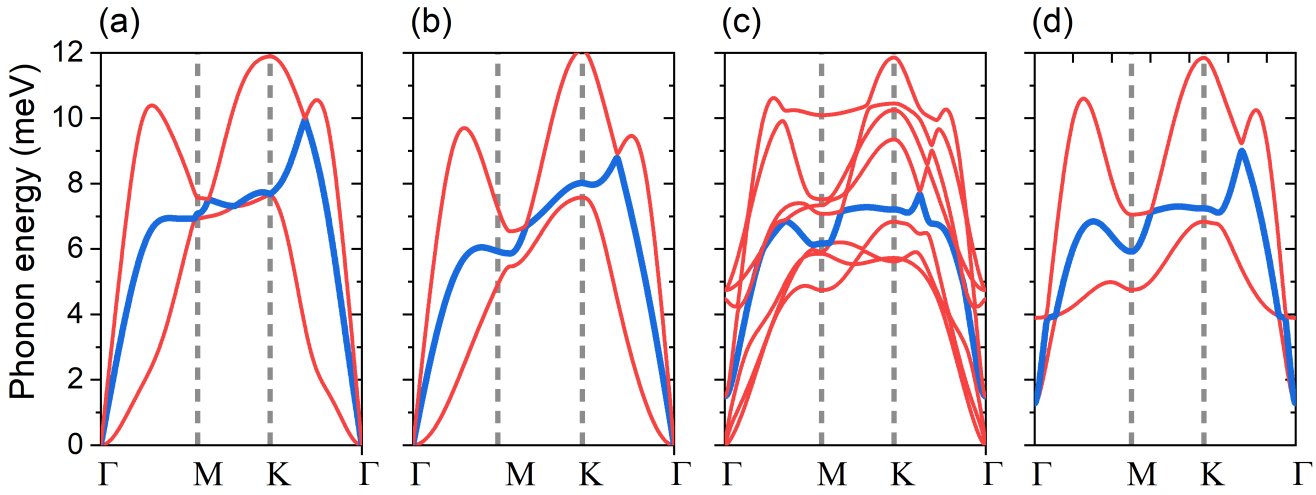


FIG. S3. Phonon dispersions of (a) free-standing Ge under zero $E_z$ and (b) finite $E_z$, (c) and (d) of Ge-InSe interface. For (d), substrate atoms from InSe are fixed at the equilibrium structure and only atoms in the material layer (Ge) are allowed to move.

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