Supporting Information of

Origin and regulation of triaxial magnetic anisotropy in the

Ferromagnetic Semiconductor CrSBr monolayer

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1. Derivation formula of second-order perturbation theory.

In order to better explain the MAE of the CrSBr monolayer, we will start with the perturbation theory. Assuming that the system Hamiltonian can be divided into two parts, one part is $\hat{H}^{(0)}$ which without perturbation, its energy eigenvalue is $E_n^{(0)}$, and its eigenwave function is $\psi^{(0)}$; the other part is the perturbation part with \hat{H}' , and \hat{H}' is relative $\hat{H}^{(0)}$ is much smaller. Then the Schrödinger equation the of the system with total \hat{H} can be written as,

$$\hat{H} = \hat{H}^{(0)} + \hat{H}', \quad \hat{H}^{(0)}\psi^{(0)} = E_n^{(0)}\psi^{(0)}$$
(1)
$$\hat{H}\psi_n = E_n\psi_n$$
(2)

To show perturbation term is so small, \hat{H}' can be written as $\hat{H}' = \lambda \hat{H}^{(1)}$, λ is a small real parameter. Since both E_n and ψ_n are related to the perturbation and are functions of the degree of perturbation λ , expand them to the power function of λ as:

$$E_n = E_n^{(0)} + \lambda E_n^{(1)} + \lambda^2 E_n^{(2)} + \dots$$
(3)

$$\psi_n = \psi_n^{(0)} + \lambda \psi_n^{(1)} + \lambda^2 \psi_n^{(2)} + \dots$$
(4)

 $\lambda E_n^{(1)}$ and $\lambda \psi_n^{(1)}$ are the first-level correction of energy and wave function, respectively. And $\lambda^2 E_n^{(2)}$ and $\lambda^2 \psi_n^{(2)}$ are the second-level correction. Therefore, the following formulas can be obtained,

$$\left(\hat{H}^{(0)} - E^{(0)}_{n}\right)\psi^{(0)}_{n} = 0 \tag{5}$$

$$\left(\hat{H}^{(0)} - E^{(0)}_{n}\right)\psi^{(1)}_{n} = -\left(\hat{H}^{(1)} - E^{(1)}_{n}\right)\psi^{(0)}_{n} \tag{6}$$

$$\left(\hat{H}^{(0)} - E_{n}^{(0)}\right)\psi_{n}^{(2)} = -\left(\hat{H}^{(1)} - E_{n}^{(1)}\right)\psi_{n}^{(1)} + E_{n}^{(2)}\psi_{n}^{(0)}$$
(7)

To get $E_n^{(1)}$, multiply $\psi_n^{*(0)}$ left simultaneously on the both sides of equation (6) and integrate over the whole space,

$$\int \psi_{n}^{*(0)} (\hat{H}^{(0)} - E_{n}^{(0)}) \psi_{n}^{(1)} d\tau = E_{n}^{(1)} \int \psi_{n}^{*(0)} \psi_{n}^{(0)} d\tau - \int \psi_{n}^{*(0)} \hat{H}^{(1)} \psi_{n}^{(0)} d\tau$$
(8)

$$E_{n}^{(1)} = \int \psi_{n}^{*(0)} \hat{H}^{(1)} \psi_{n}^{(0)} d\tau = H_{nn}^{(1)}$$
(9)

Given $E_n^{(1)}$, the first-order correction term $\Psi_n^{(1)}$ of the wave function can be obtained from equation (6). Expand the eigenfunction system of $\Psi_n^{(1)}$ according to $\hat{H}^{(0)}$ as, $\Psi_n^{(1)} = \sum_l a_l^{(1)} \Psi_l^{(0)}$ $(l \neq n)$ into equation (6) and left multiplied by $\Psi_m^{(0)}$ on both sides

of the equation at the same time to integrate in the whole space, we get,

$$\int \psi_{m}^{*(0)} \left(\sum_{l} E_{l}^{(0)} a_{l}^{(1)} \psi_{l}^{(0)} - E_{n}^{(0)} \sum_{l} a_{l}^{(1)} \psi_{l}^{(0)} \right) d\tau = \int \psi_{n}^{*(0)} (E_{n}^{(1)} \psi_{n}^{(0)} - \hat{H}^{(1)} \psi_{n}^{(0)}) d\tau$$

$$(10)$$

$$\sum_{l \neq n} E_{l}^{(0)} a_{l}^{(1)} \delta_{ml} - E_{n}^{(0)} \sum_{l \neq n} a_{l}^{(1)} \delta_{ml} = -\int \psi_{m}^{*(0)} \hat{H}^{(1)} \psi_{n}^{(0)} d\tau = H_{mn}^{(1)}$$

$$(11)$$

....

When $m = l_{1} \left(E_{n}^{(0)} - E_{m}^{(0)} \right) a_{m}^{(1)} = H_{mn, \text{ so}}^{(1)}$

$$a_{m}^{(1)} = \frac{H_{mn}^{(1)}}{E_{n}^{(0)} - E_{m}^{(0)}}$$
(12)
$$\psi_{n}^{(1)} = \sum_{m \neq n} \frac{H_{mn}^{(1)}}{E_{n}^{(0)} - E_{m}^{(0)}} \psi_{m}^{(0)}$$
(13)
correction of energy $\frac{E_{n}^{(2)}}{E_{n}^{(0)}}$ Put $\psi_{n}^{(1)} = \sum_{l} a_{l}^{(1)} \psi_{l}^{(0)} \ (l \neq n)$

Now find the second-order correction of energy $E_n^{(2)}$. Put

into equation (7), and use $\psi_{m}^{*(0)}$ to multiply both sides of the equation to integrate the full space,

$$\int \psi_{m}^{*(0)} (\hat{H}^{(0)} - E_{n}^{(0)}) \psi_{n}^{(2)} d\tau = -\sum_{l \neq n} a_{l}^{(1)} H_{nl}^{(1)} + E_{n}^{(1)} \sum_{l \neq n} a_{l}^{(1)} \delta_{nl} + E_{n}^{(2)}$$
(14)
$$E_{n}^{(2)} = \sum_{l \neq n} a_{l}^{(1)} H_{nl}^{(1)} = \sum_{m \neq n} \frac{H_{mn}^{(1)} H_{nm}^{(1)}}{E_{n}^{(0)} - E_{m}^{(0)}} = \sum_{m \neq n} \frac{|H_{nm}^{(1)}|^{2}}{E_{n}^{(0)} - E_{m}^{(0)}} (l = m)$$
(15)

The energy perturbed by the system is,

$$\Delta E = H_{nn}^{(1)} + \sum_{m \neq n} \frac{\left|H_{nm}^{(1)}\right|^2}{E_n^{(0)} - E_m^{(0)}} + \dots$$
(16)

For 3d transition metals, the energy level splitting caused by spin-orbit coupling is

much smaller than the energy level splitting caused by electron correlation interaction, so it is reasonable to regard spin-orbit coupling as perturbation. Therefore, according to the perturbation theory, the Hamiltonian including SOC can be written as,

$$\hat{H} = \hat{H}^{(0)} + \lambda \langle n \left| \hat{S} \cdot \hat{L} \right| n \rangle + \lambda^2 \sum_{m \neq n} \frac{|\langle n \left| \hat{S} \cdot \hat{L} \right| m \rangle|^2}{E_n^{(0)} - E_m^{(0)}}$$
(17)

Then the corresponding energy is,

$$E^{SOC} = \lambda \langle n \mid \Im \cdot \mathcal{L} \mid n \rangle + \lambda^2 \sum_{m \neq n} \frac{|\langle n \mid \Im \cdot \mathcal{L} \mid m \rangle|^2}{E_n^{(0)} - E_m^{(0)}}$$
(18)

where $|n\rangle$ and $|m\rangle$ respectively represent the Hamiltonian quantum state and $E_n^{(0)}$ and $E_m^{(0)}$ are the intrinsic energy values of these two states. If spin states σ (both spin-up states $|\uparrow\rangle$ and spin-down states $|\downarrow\rangle$) is considered in the wave function, then,

$$E^{SOC} = \lambda(\langle \uparrow, n \mid \mathring{S} \cdot \mathring{L} \mid \uparrow, n \rangle + \langle \uparrow, n \mid \mathring{S} \cdot \mathring{L} \mid \downarrow, n \rangle + \langle \downarrow, n \mid \mathring{S} \cdot \mathring{L} \mid \uparrow, n \rangle + \langle \downarrow, n \mid \mathring{S} \cdot \mathring{L} \mid \downarrow, n \rangle)$$

$$\lambda^{2} (\sum_{m \neq n} \frac{|\langle \uparrow, n \mid \mathring{S} \cdot \mathring{L} \mid \uparrow, m \rangle|^{2}}{E_{n}^{(0)} - E_{m}^{(0)}} + \sum_{m \neq n} \frac{|\langle \uparrow, n \mid \mathring{S} \cdot \mathring{L} \mid \downarrow, m \rangle|^{2}}{E_{n}^{(0)} - E_{m}^{(0)}}$$

$$+ \sum_{m \neq n} \frac{|\langle \downarrow, n \mid \mathring{S} \cdot \mathring{L} \mid \uparrow, m \rangle|^{2}}{E_{n}^{(0)} - E_{m}^{(0)}} + \sum_{m \neq n} \frac{|\langle \downarrow, n \mid \mathring{S} \cdot \mathring{L} \mid \downarrow, m \rangle|^{2}}{E_{n}^{(0)} - E_{m}^{(0)}})$$

$$(19)$$

Now discuss the representation of Hamiltonian quantum states. The Hamiltonian quantum state $|n\rangle$ can be written as a wave function such as,

$$\Psi = \psi_{nlm}(\vec{r},\theta,\varphi) = R_{nl}(\vec{r})Y_{lm}(\theta,\varphi)$$
(20)

Where n = 1, 2, 3, ... is called principal quantum number; l = 0, 1, 2, ... is called the angular quantum number which characterize the magnitude of angular momentum; m = l, l - 1, l - 2, ..., -l + 2, -l + 1, -l is called magnetic quantum number. $R_{nl}(\vec{r})$ is a radial function, related only to the position vector \vec{r} whose expression is,

$$R_{nl}(\vec{r}) = N_{nl}e^{-\frac{Z}{na_0}r} \left(\frac{2Z}{na_0}r\right)^l F(-n+l+1, 2l+2, \frac{2Z}{na_0}r)$$
(21)

$$N_{nl} = \frac{2}{(2l+1)!} \sqrt{\frac{(n+l)!Z^3}{(n-l-1)!a_0^3}}$$
$$a_0 = \frac{\hbar^2}{m a_0^2}$$

Where $m_e e_s^2$ and Z is the atomic number. The values of several radial functions are listed below:

$$R_{1,0}(r) = \left(\frac{Z}{a_0}\right)^{\frac{3}{2}} 2e^{\left(-\frac{Zr}{a_0}\right)}, \qquad (22)$$

$$R_{2,0}(r) = \left(\frac{Z}{2a_0}\right)^{\frac{3}{2}} (2 - \frac{Zr}{a_0})e^{\left(-\frac{Zr}{2a_0}\right)}, \qquad (22)$$

$$R_{2,1}(r) = \left(\frac{Z}{2a_0}\right)^{\frac{3}{2}} \frac{Zr}{a_0\sqrt{3}}e^{\left(-\frac{Zr}{2a_0}\right)}, \qquad (22)$$

$$R_{3,0}(r) = \left(\frac{Z}{3a_0}\right)^{\frac{3}{2}} \left[2 - \frac{4Zr}{3a_0} + \frac{4}{27}(\frac{Zr}{a_0})^2\right] e^{\left(-\frac{Zr}{3a_0}\right)}, \qquad (22)$$

$$R_{3,1}(r) = \left(\frac{2Z}{a_0}\right)^{\frac{3}{2}} \left(\frac{2}{27\sqrt{3}} - \frac{Zr}{81a_0\sqrt{3}}\right) \frac{Zr}{a_0}e^{\left(-\frac{Zr}{3a_0}\right)}, \qquad (22)$$

$$R_{3,2}(r) = \left(\frac{2Z}{a_0}\right)^{\frac{3}{2}} \frac{1}{81\sqrt{15}} \left(\frac{Zr}{a_0}\right)^2 e^{\left(-\frac{Zr}{3a_0}\right)}, \qquad (22)$$

The spherical harmonic function $Y_{lm}(\theta, \varphi)$ is a single-valued, continuous, and bounded complex function containing θ ($\theta \in [0, \pi]$) and φ ($\varphi \in [0, 2\pi]$). It can be expressed by the following formula.

$$Y_{lm}(\theta,\varphi) = (-1)^m N_{lm} P_l^m(\cos\theta) e^{im\varphi}$$

Where $P_l^m(\cos\theta)$ is associated Legendre polynomial. $N_{lm} = \sqrt{\frac{(l-m)!(2l+1)}{(l+m)!4\pi}}$ is the normalization factor. Next, list a few spherical harmonic functions

$$\begin{split} Y_{00}(\theta,\varphi) &= \frac{1}{2\sqrt{\pi}}, \end{split} (23) \\ Y_{1+1}(\theta,\varphi) &= -\frac{1}{2\sqrt{2\pi}} \frac{\sqrt{3}}{2\pi} \sin \theta e^{i\varphi}, \\ Y_{10}(\theta,\varphi) &= \frac{1}{2\sqrt{2\pi}} \frac{\sqrt{3}}{2\pi} \sin \theta e^{-i\varphi}, \\ Y_{10}(\theta,\varphi) &= \frac{1}{2\sqrt{2\pi}} \frac{\sqrt{3}}{2\pi} \sin \theta e^{-i\varphi}, \\ Y_{1-1}(\theta,\varphi) &= \frac{1}{2\sqrt{2\pi}} \frac{\sqrt{3}}{2\pi} \sin \theta e^{-i\varphi}, \\ Y_{2+2}(\theta,\varphi) &= \frac{1}{4\sqrt{\frac{3}{2\pi}}} \frac{\sqrt{3}}{2\pi} \cos \theta \sin \theta e^{i\varphi} = -\frac{1}{4\sqrt{\frac{3}{2\pi}}} \frac{\sqrt{3}}{2\pi} \sin 2\theta e^{i\varphi}, \\ Y_{2+1}(\theta,\varphi) &= -\frac{1}{2\sqrt{\frac{3}{2\pi}}} \frac{\sqrt{3}}{2\pi} \cos \theta \sin \theta e^{i\varphi} = -\frac{1}{4\sqrt{\frac{3}{2\pi}}} \frac{\sqrt{3}}{2\pi} \sin 2\theta e^{i\varphi}, \\ Y_{20}(\theta,\varphi) &= \frac{1}{4\sqrt{\frac{\pi}{\pi}}} \frac{\sqrt{3}}{2\pi} \cos \theta \sin \theta e^{-i\varphi} = \frac{1}{4\sqrt{\frac{3}{2\pi}}} \frac{\sqrt{3}}{2\pi} \sin 2\theta e^{-i\varphi}, \\ Y_{2-1}(\theta,\varphi) &= \frac{1}{2\sqrt{\frac{3}{2\pi}}} \frac{\sqrt{3}}{2\pi} \cos \theta \sin \theta e^{-i\varphi} = \frac{1}{4\sqrt{\frac{3}{2\pi}}} \frac{\sqrt{3}}{2\pi} \sin 2\theta e^{-i\varphi}, \\ Y_{2-1}(\theta,\varphi) &= \frac{1}{4\sqrt{\frac{3}{2\pi}}} \frac{\sqrt{3}}{2\pi} \sin^{2} \theta e^{-i2\varphi} = \frac{1}{8\sqrt{\frac{3}{2\pi}}} (1 - \cos 2\theta) e^{-i2\varphi}, \\ Y_{3+3}(\theta,\varphi) &= -\frac{1}{4\sqrt{\frac{3}{2\pi}}} \frac{\sqrt{3}}{\pi} \sin^{3} \theta e^{i3\varphi} = -\frac{1}{32\sqrt{\frac{5}{\pi}}} \frac{\sqrt{3}}{\pi} (\sin \theta - \sin 3\theta) e^{i3\varphi}, \\ Y_{3+2}(\theta,\varphi) &= \frac{1}{4\sqrt{\frac{3}{2\pi}}} \frac{\sqrt{3}}{2\pi} \cos \theta \sin^{2} \theta e^{i2\varphi} = \frac{1}{16\sqrt{\frac{3}{2\pi}}} \frac{\sqrt{3}}{2\pi} (\cos \theta - \cos 3\theta) e^{i2\varphi}, \\ Y_{30}(\theta,\varphi) &= -\frac{1}{8\sqrt{\frac{\pi}{\pi}}} (5 \cos^{2} \theta - 1) \sin \theta e^{i\varphi} = -\frac{1}{32\sqrt{\frac{\pi}{\pi}}} (\sin \theta + 5 \sin 3\theta) e^{i\varphi}, \\ Y_{30}(\theta,\varphi) &= \frac{1}{4\sqrt{\frac{\pi}{\pi}}} (5 \cos^{2} \theta - 1) \sin \theta e^{-i\varphi} = -\frac{1}{32\sqrt{\frac{\pi}{\pi}}} (\sin \theta + 5 \sin 3\theta) e^{i\varphi}, \\ Y_{3-1}(\theta,\varphi) &= \frac{1}{4\sqrt{\frac{\pi}{\pi}}} \frac{\sqrt{3}}{2\pi} \cos \theta \sin^{2} \theta e^{-i2\varphi} = \frac{1}{16\sqrt{\frac{\pi}{\pi}}} \frac{\sqrt{3}}{\pi} (\sin \theta + 5 \sin 3\theta) e^{i\varphi}, \\ Y_{3-2}(\theta,\varphi) &= \frac{1}{4\sqrt{\frac{\pi}{\pi}}} \frac{\sqrt{3}}{\pi} (5 \cos^{2} \theta - 1) \sin \theta e^{-i\varphi} = -\frac{1}{32\sqrt{\frac{\pi}{\pi}}} (\sin \theta + 5 \sin 3\theta) e^{i\varphi}, \\ Y_{3-1}(\theta,\varphi) &= \frac{1}{4\sqrt{\frac{\pi}{\pi}}} \frac{\sqrt{3}}{\pi} (\cos^{2} \theta - 1) \sin \theta e^{-i\varphi} = -\frac{1}{32\sqrt{\frac{\pi}{\pi}}} (\sin \theta + 5 \sin 3\theta) e^{-i\varphi}, \\ Y_{3-2}(\theta,\varphi) &= \frac{1}{4\sqrt{\frac{\pi}{\pi}}} \frac{\sqrt{3}}{\pi} \cos^{2} \theta e^{-i2\varphi} = \frac{1}{16\sqrt{\frac{\pi}{\pi}}} \frac{\sqrt{3}}{\pi} (\cos \theta - \cos 3\theta) e^{-i\varphi}, \\ Y_{3-2}(\theta,\varphi) &= \frac{1}{4\sqrt{\frac{\pi}{\pi}}} \frac{\sqrt{3}}{\pi} \cos^{2} \theta - \frac{1}{\pi} \cos^{2} \theta -$$

$$Y_{3-3}(\theta,\varphi) = \frac{1}{8} \sqrt{\frac{5\cdot7}{\pi}} \sin^3 \theta e^{-i3\varphi} = \frac{1}{32} \sqrt{\frac{5\cdot7}{\pi}} (3\sin\theta - \sin 3\theta) e^{-i3\varphi},$$

Thus, the total wave function and the orbits it represents are therefore listed Table S1.

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Table S1. The principal quantum number, angular quantum number, magnetic quantum number, total wave function, and corresponding orbit.

n	l	т	$\Psi = \psi_{nlm}(\vec{r},\theta,\varphi)$	orbit
1	0	0	$\psi_{100}(\vec{r},\theta,\varphi) = R_{10}(\vec{r})Y_{00}(\theta,\varphi)$	1s
	0	0	$\psi_{200}(\vec{r},\theta,\varphi) = R_{20}(\vec{r})Y_{00}(\theta,\varphi)$	2s
2		-1	$\psi_{21-1}(\vec{r},\theta,\varphi) = R_{21}(\vec{r})Y_{1-1}(\theta,\varphi)$	2py
Z	1	0	$\psi_{210}(\vec{r},\theta,\varphi) = R_{21}(\vec{r})Y_{10}(\theta,\varphi)$	2pz
		1	$\psi_{211}(\vec{r},\theta,\varphi) = R_{21}(\vec{r})Y_{11}(\theta,\varphi)$	2p _x
	0	0	$\psi_{300}(\vec{r},\theta,\varphi) = R_{30}(\vec{r})Y_{00}(\theta,\varphi)$	3s
		-1	$\psi_{31-1}(\vec{r},\theta,\varphi) = R_{31}(\vec{r})Y_{1-1}(\theta,\varphi)$	3py
	1	0	$\psi_{310}(\vec{r},\theta,\varphi) = R_{31}(\vec{r})Y_{10}(\theta,\varphi)$	3pz
		1	$\psi_{311}(\vec{r},\theta,\varphi) = R_{31}(\vec{r})Y_{11}(\theta,\varphi)$	3p _x
3		-2	$\psi_{32-2}(\vec{r},\theta,\varphi) = R_{32}(\vec{r})Y_{2-2}(\theta,\varphi)$	$3d_{xy}$
		-1	$\psi_{32-1}(\vec{r},\theta,\varphi) = R_{32}(\vec{r})Y_{2-1}(\theta,\varphi)$	$3d_{yz}$
	2	0	$\psi_{320}(\vec{r},\theta,\varphi) = R_{32}(\vec{r})Y_{20}(\theta,\varphi)$	$3d_z^2$
		1	$\psi_{321}(\vec{r},\theta,\varphi) = R_{32}(\vec{r})Y_{21}(\theta,\varphi)$	$3d_{xz}$
		2	$\psi_{322}(\vec{r},\theta,\varphi) = R_{32}(\vec{r})Y_{22}(\theta,\varphi)$	$3d_{x}^{2}-y^{2}$

Then, the part of $\hat{S} \cdot \hat{L}$ can be written as the following form in spherical coordinates.

$$\begin{split} \hat{l}_{x} &= i\hbar(\sin\varphi\frac{\partial}{\partial\theta} + \frac{\cos\theta}{\sin\theta}\cos\varphi\frac{\partial}{\partial\varphi}), \end{split} \tag{24} \\ \hat{l}_{y} &= -i\hbar(\cos\varphi\frac{\partial}{\partial\theta} - \frac{\cos\theta}{\sin\theta}\sin\varphi\frac{\partial}{\partial\varphi}), \\ \hat{l}_{z} &= -i\hbar\frac{\partial}{\partial\varphi}, \end{aligned}$$
$$\begin{split} \hat{l}^{2} &= -\hbar^{2} \bigg[\frac{1}{\sin\theta\partial\theta}(\sin\theta\frac{\partial}{\partial\theta} + \frac{1}{\sin^{2}\theta\partial\varphi^{2}}) \bigg] \end{split}$$

Now, taking equations (20 - 24) into the first term of equation (19) and get the Table S2, noting that all diagonal elements in Table 2 are zero. Therefore, the effect of the first order perturbation term on the SOC is zero, so we can just consider the second-order perturbation term when calculating the SOC (shown in Table S3).

Table S2. Matrix elements $\langle \sigma_{i\nu} p_i | \hat{S}_n \cdot \hat{L}_n | \sigma_{j\nu} p_j \rangle$ and $\langle \sigma_{i\nu} d_i | \hat{S}_n \cdot \hat{L}_n | \sigma_{j\nu} d_j \rangle$ of the spin-orbit coupling operator with respect to spin state in direction $\vec{n} = (x, y, z)$, in units of \hbar .

	$ \uparrow, p_y\rangle$	$ \uparrow,p_z angle$	$ \uparrow, p_x\rangle$	$ \downarrow,p_{y}\rangle$	$ \downarrow,p_z angle$	$ \downarrow, p_x\rangle$
$ \uparrow, p_y\rangle$	0	0	iz	0	-ix	0
$ \uparrow,p_z\rangle$	0	0	0	ix	0	-y
$ \uparrow, p_x\rangle$	-iz	0	0	0	у	0
$ \downarrow, p_y\rangle$	0	-ix	0	0	0	-iz
$ \downarrow,p_z\rangle$	ix	0	у	0	0	0
$ \downarrow, p_x\rangle$	0	-у	0	iz	0	0

	$\left \uparrow,d_{xy} ight angle$	$ \uparrow, d_{yz}\rangle$	$ \uparrow, d_{z^2}\rangle$	$\left \uparrow,d_{xz} ight angle$	$ \uparrow, d_{x^2 - y^2}\rangle$	$\left \downarrow,d_{xy}\right\rangle$	$ \downarrow, d_{yz}\rangle$	$ \downarrow, d_{z^2}\rangle$	$\left \downarrow,d_{\chi\chi} ight angle$	$ \downarrow, d_{x^2 - y^2}\rangle$
$ \uparrow, d_{xy}\rangle$	0	0	0	0	2iz	0	у	0	-ix	0
$ \uparrow, d_{yz}\rangle$	0	0	0	iz	0	-y	0	$\sqrt{3}ix$	0	-ix
$ \uparrow, d_{z^2}\rangle$	0	0	0	0	0	0	$\sqrt{3}ix$	0	$\sqrt{3}y$	0
$ \uparrow, d_{xz}\rangle$	0	-iz	0	0	0	ix	0	$\sqrt{3}y$	0	-у
$ \uparrow, d_{x^2 - y^2}\rangle$	-2iz	0	0	0	0	0	ix	0	у	0
$ \downarrow, d_{xy}\rangle$	0	-у	0	-ix	0	0	0	0	0	-2iz
$ \downarrow, d_{yz}\rangle$	У	0	$\sqrt{3}ix$	0	-ix	0	0	0	-iz	0
$ \downarrow, d_{z^2}\rangle$	0	$\sqrt{3}ix$	0	$\sqrt{3}y$	0	0	0	0	0	0
$ \downarrow, d_{xz}\rangle$	ix	0	$\sqrt{3}y$	0	у	0	iz	0	0	0

Table S3. Matrix elements $|\langle \sigma_i, p_i | S_n \cdot \hat{L}_n | \sigma_j, p_j \rangle|^2$ and $|\langle \sigma_i, d_i | S_n \cdot \hat{L}_n | \sigma_j, d_j \rangle|^2$ of the spin-orbit coupling operator with respect to spin state in direction $\vec{n} = (x, y, z)$, in units of \hbar .

	$ \uparrow,p_{y}\rangle$	$ \uparrow,p_z angle$	$ \uparrow, p_x\rangle$	$ \downarrow,p_{y}\rangle$	$ \downarrow,p_z angle$	$ \downarrow, p_x\rangle$
$ \uparrow, p_y\rangle$	0	0	z^2	0	x^2	0
$ \uparrow, p_z\rangle$	0	0	0	x^2	0	y^2
$ \uparrow, p_x\rangle$	z^2	0	0	0	y^2	0
$ \downarrow, p_y\rangle$	0	x^2	0	0	0	z^2
$ \downarrow, p_z\rangle$	x^2	0	y^2	0	0	0
$ \downarrow, p_x\rangle$	0	y^2	0	z^2	0	0

	$ \uparrow, d_{xy}\rangle$	$\left \uparrow,d_{yz}\right\rangle$	$ \uparrow, d_{z^2}\rangle$	$ \uparrow, d_{xz}\rangle$	$ \uparrow, d_{x^2 - y^2}\rangle$	$ \downarrow, d_{xy}\rangle$	$ \downarrow, d_{yz}\rangle$	$ \downarrow, d_{z^2}\rangle$	$ \downarrow, d_{xz}\rangle$	$ \downarrow, d_{x^2 - y^2}\rangle$
$ \uparrow, d_{xy}\rangle$	0	0	0	0	$4z^{2}$	0	y^2	0	x^2	0
$ \uparrow, d_{yz}\rangle$	0	0	0	z^2	0	\mathcal{Y}^2	0	$3x^{2}$	0	x^2
$ \uparrow, d_{z^2}\rangle$	0	0	0	0	0	0	$3x^{2}$	0	$3y^{2}$	0
$ \uparrow, d_{xz}\rangle$	0	z^2	0	0	0	x^2	0	$3y^{2}$	0	y^2
$ \uparrow, d_{x^2-y^2}\rangle$	$4z^{2}$	0	0	0	0	0	x^2	0	y^2	0
$ \downarrow, d_{xy}\rangle$	0	y^2	0	x^2	0	0	0	0	0	$4z^{2}$
$ \downarrow, d_{yz}\rangle$	\mathcal{Y}^2	0	$3x^{2}$	0	x^2	0	0	0	z^2	0
$ \downarrow, d_{z^2}\rangle$	0	$3x^{2}$	0	$3y^{2}$	0	0	0	0	0	0
$ \downarrow, d_{xz}\rangle$	x^2	0	$3y^{2}$	0	y^2	0	z^2	0	0	0
$ \downarrow, d_{x^2 - y^2}\rangle$	0	x^2	0	y^2	0	$4z^{2}$	0	0	0	0

Table 4. The following three tables show the *d*-orbit coupling matrices in the *x*-, *y*- and

	$ \uparrow, d_{xy}\rangle$	$ \uparrow, d_{yz}\rangle$	$ \uparrow, d_{z^2}\rangle$	$ \uparrow, d_{xz}\rangle$	$ \uparrow, d_{x^2-y^2}\rangle$	$ \downarrow, d_{xy}\rangle$	$ \downarrow, d_{yz}\rangle$	$ \downarrow, d_{z^2}\rangle$	$ \downarrow, d_{xz}\rangle$	$ \downarrow, d_{x^2 - y^2}\rangle$
$ \uparrow, d_{xy}\rangle$	0	0	0	0	0	0	0	0	1	0
$ \uparrow, d_{yz}\rangle$	0	0	0	0	0	0	0	3	0	1
$ \uparrow, d_{z^2}\rangle$	0	0	0	0	0	0	3	0	0	0
$ \uparrow, d_{xz}\rangle$	0	0	0	0	0	1	0	0	0	0
$ \uparrow, d_{x^2-y^2}\rangle$	0	0	0	0	0	0	1	0	0	0
$ \downarrow, d_{xy}\rangle$	0	0	0	1	0	0	0	0	0	0
$ \downarrow, d_{yz}\rangle$	0	0	3	0	1	0	0	0	0	0
$ \downarrow, d_{z^2}\rangle$	0	3	0	0	0	0	0	0	0	0
$ \downarrow, d_{xz}\rangle$	1	0	0	0	0	0	0	0	0	0
$ \downarrow, d_{x^2-y^2}\rangle$	0	1	0	0	0	0	0	0	0	0

z- directions respectively.

	$ \uparrow, d_{xy}\rangle$	$\left \uparrow,d_{yz}\right\rangle$	$ \uparrow, d_{z^2}\rangle$	$ \uparrow, d_{xz}\rangle$	$ \uparrow, d_{x^2 - y^2}\rangle$	$ \downarrow, d_{xy}\rangle$	$ \downarrow, d_{yz}\rangle$	$ \downarrow, d_{z^2}\rangle$	$ \downarrow, d_{xz}\rangle$	$ \downarrow, d_{x^2 - y^2}\rangle$
$ \uparrow, d_{xy}\rangle$	0	0	0	0	0	0	1	0	0	0
$ \uparrow, d_{yz}\rangle$	0	0	0	0	0	1	0	0	0	0
$ \uparrow, d_{z^2}\rangle$	0	0	0	0	0	0	0	0	3	0
$ \uparrow, d_{xz}\rangle$	0	0	0	0	0	0	0	3	0	1
$ \uparrow, d_{x^2-y^2}\rangle$	0	0	0	0	0	0	0	0	1	0
$ \!\downarrow, d_{xy}\rangle$	0	1	0	0	0	0	0	0	0	0
$ \downarrow, d_{yz}\rangle$	1	0	0	0	0	0	0	0	0	0
$ \downarrow, d_{z^2}\rangle$	0	0	0	3	0	0	0	0	0	0
$ \downarrow, d_{xz}\rangle$	0	0	3	0	0	0	0	0	0	0
$ \downarrow, d_{x^2 - y^2}\rangle$	0	0	0	1	0	0	0	0	0	0

	$ \uparrow, d_{xy}\rangle$	$ \uparrow, d_{yz}\rangle$	$ \uparrow, d_{z^2}\rangle$	$ \uparrow, d_{xz}\rangle$	$ \uparrow, d_{x^2-y^2}\rangle$	$\left \downarrow,d_{xy}\right\rangle$	$ \downarrow, d_{yz}\rangle$	$ \downarrow, d_{z^2}\rangle$	$ \downarrow, d_{xz}\rangle$	$ \downarrow, d_{x^2 - y^2}\rangle$
$ \uparrow, d_{xy}\rangle$	0	0	0	0	4	0	0	0	0	0
$ \uparrow, d_{yz}\rangle$	0	0	0	1	0	0	0	0	0	0
$ \uparrow, d_{z^2}\rangle$	0	0	0	0	0	0	0	0	0	0
$\left \uparrow,d_{xz} ight angle$	0	1	0	0	0	0	0	0	0	0
$ \uparrow, d_{x^2-y^2}\rangle$	4	0	0	0	0	0	0	0	0	0
$ \downarrow, d_{xy}\rangle$	0	0	0	0	0	0	0	0	0	4
$ \downarrow, d_{yz}\rangle$	0	0	0	0	0	0	0	0	1	0
$ \downarrow, d_{z^2}\rangle$	0	0	0	0	0	0	0	0	0	0
$ \downarrow, d_{xz}\rangle$	0	0	0	0	0	0	1	0	0	0
$ \downarrow, d_{x^2 - y^2}\rangle$	0	0	0	0	0	4	0	0	0	0

2. One FM state and Four AFM states of a 2×2 supercell CrSBr

monolayer.



Figure S1 The 2×2 supercell of CrSBr monolayer with one FM (a) and four AFM (b - e). Red and blue respectively represent different spin directions.

3. Density of states (DOS) for CrSBr monolayer



Figure S2 The DOS and PDOS for CrSBr monolayer under no strain.

4. Change of bond angle, the energy difference, and magnetic moment under biaxial and uniaxial strains.



Figure S3 Change of bond angle (a), the energy difference $(\Box E)$ between ferromagnetic and different antiferromagnetic states (b), and magnetic moment (c) with different strains, respectively.

To study the effect of strain on the lattice changes of CrSBr monolayers in detail, the change of bond angle θ_1 (Cr-S1-Cr), θ_2 (Cr-Br-Cr), θ_3 (Cr-S3-Cr) as shown in Figure 1a with different strains was shown in Figure S3a. The result shows that the bond angles of CrSBr monolayer change slightly with strains. Figure S3b shows the energy difference between ferromagnetic and different antiferromagnetic states, and indicates that the ferromagnetic ground state of CrSBr monolayer does not change under different strains. Besides, the total magnetic moment of CrSBr monolayer also changes little under different strains (Figure S3c).



5. The band gaps for CrSBr monolayer under different strains.

Figure S4 The band gaps of CrSBr monolayer under uniaxial strains along x direction.



Figure S5 The band gaps of CrSBr monolayer under uniaxial strains along y direction.



Figure S6 The band gaps of CrSBr monolayer under biaxial strains.

6. Magnetic anisotropy energy in whole space as well as in the three planes under different strains.



Figure S7 (a) and (b) SOC-MAE of CrSBr monolayer in the whole space and the *xy*, *yz*, *xz* planes. (c) and (d)Shape-MAE of CrSBr monolayer in the whole space with no strain.



Figure S8 The variation of SOC-MAE (a) and total-MAE (b) for CrSBr monolayer in the whole space under biaxial xy strains ($-5\% \sim 5\%$).



Figure S9 The variation of SOC-MAE (a) and total-MAE (b) for CrSBr monolayer in the whole space under uniaxial y strain (-5% \sim 5%).

7. The contribution to MAE from the SOC interaction between different d orbital channels for Cr atoms under different strains



Figure S10 The contributions of p orbitals and d orbitals of Cr, S, and Br atom to SOC-MAE along [100] direction (a) and [010] direction (b) for CrSBr monolayer under different strains. The energy is the reference along [001] direction.



Figure S11 The contribution to MAE from the SOC interaction between different d orbital channels along [100] (a) and [010] (b) directions for Cr atoms under different biaxial xy strains. The energy is the reference along [001] direction.



Figure S12 The contribution to MAE from the SOC interaction between different p orbital channels along [100] (a) and [010] (b) directions for Br atoms under different biaxial xy strains. The energy is the reference along [001] direction.

Figure S13 The contribution to MAE from the SOC interaction between different p orbital channels along [100] (a) and [010] (b) directions for Br atoms under different uniaxial x strains. The energy is the reference along [001] direction.

Figure S14 The contribution to MAE from the SOC interaction between different d orbital channels along [100] (a) and [010] (b) directions for Cr atoms under different uniaxial y strains. The energy is the reference along [001] direction.

Figure S15 The contribution to MAE from the SOC interaction between different p orbital channels along [100] (a) and [010] (b) directions for Br atoms under different uniaxial y strains. The energy is the reference along [001] direction.

8. The variations of Curie temperature (T_C) under different strains.

Figure S16 The verities of T_C for CrSBr monolayer under the biaxial strain.

Figure S17 The verities of T_C under for CrSBr monolayer the x-directional uniaxial strain

Figure S18 The verities of T_C for CrSBr monolayer under the y-directional uniaxial strain.