Supplementary Information

Strong Coupling Mechanism between Ferromagnetism and Piezoelectricity in 2D Ferroelectric Cr*X*S*Y*Br*Z* with High Carrier Mobility

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Transport and piezoelectric properties of CrSBr and Cr*X*Br*Y*S*Z* monolayers

The relaxed ion piezoelectric stress tensor is determined by combining the contributions from both ions and electrons:[1]

$$e_{ijk} = \frac{dP_i}{d\varepsilon_{jk}} = e_{ijk}^{ion} + e_{ijk}^{ele}, \tag{S1}$$

where ε_{jk} is the stress tensor and P_i is the intrinsic polarization tensor. i, j, and k represent the x, y, and z axes, respectively. Similarly, the piezoelectric strain coefficient d_{ijk} can be defined as the derivative of the P_i with respect to the strain tensor σ_{jk}:

$$d_{ijk} = \frac{dP_i}{d\sigma_{jk}}, \tag{S2}$$

For simplicity, in the condensed Voigt notation, the third-order tensors d_{ijk} and e_{ijk} are commonly denoted as d_{il} and e_{il}. Here, the subscript i corresponds to the x, y, or z axes, indicated by the numbers 1, 2, and 3. The subscript l represents the second-order tensor xx, yy, zz, yz, zx, xy, which are denoted by the numbers 1, 2, 3, 4, 5, and 6, respectively.[2] The fourth-order tensor elastic stiffness coefficient C_{kl} serves as a link connecting e_{il} and d_{ik}:

$$e_{il} = d_{ik} C_{kl}, \tag{S3}$$

where the subscript i in the piezoelectric strain coefficient indicates the polarization direction.

The e_{il}, d_{ik} and C_{kl} are all determined by the lattice symmetry, with each being expressed as a matrix. The CrSBr monolayer possesses a D_{2h} space point group (Pmmn spatial symmetry), indicating a center of symmetry in the vertical direction (z-direction). Consequently, it lacks out-of-plane piezoelectricity, leading to a matrix element of 0 for

i = 3 in e_{il} and d_{ik}. When atomic substitutions create the Cr$XSYZ$Br monolayers from the CrSBr monolayer, the resulting structures exhibit C_{2v} space point group (Pmm2 spatial symmetry). This alteration breaks the vertical symmetry of the CrSBr monolayer, allowing for the expression of e_{il}, C_{kl} and d_{ik} in Cr$XSYZ$Br monolayers as follows:

$$e_{il} = \begin{pmatrix} 0 & 0 & 0 & 0 & e_{15} & 0 \\ 0 & 0 & 0 & e_{24} & 0 & 0 \\ e_{31} & e_{32} & e_{33} & 0 & 0 & 0 \end{pmatrix}, \tag{S4}$$

$$C_{kl} = \begin{pmatrix} C_{11} & C_{12} & C_{13} & 0 & 0 & 0 \\ C_{12} & C_{22} & C_{23} & 0 & 0 & 0 \\ C_{13} & C_{23} & C_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & C_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & C_{55} & 0 \\ 0 & 0 & 0 & 0 & 0 & C_{66} \end{pmatrix}, \tag{S5}$$

$$d_{ik} = \begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{pmatrix}, \tag{S6}$$

The relational expressions for the out-of-plane piezoelectric coefficients are derived:

$$d_{31} = \frac{Ae_{31} + Be_{32} + Ce_{33}}{G}, \tag{S7}$$

$$d_{32} = \frac{Be_{31} + De_{32} + Ee_{33}}{G}, \tag{S8}$$

$$d_{33} = \frac{Ce_{31} + Ee_{32} + Fe_{33}}{G}, \tag{S9}$$

where $A = C_{22}C_{33} - C_{23}^2$, $B = C_{13}C_{23} - C_{12}C_{33}$, $C = C_{12}C_{23} - C_{13}C_{22}$, $D = C_{11}C_{33} - C_{13}^2$, $E = C_{12}C_{13} - C_{11}C_{23}$, $F = C_{11}C_{22} - C_{12}^2$ and $G = C_{12}(C_{13}C_{23} - C_{12}C_{33}) + C_{22}(C_{11}C_{33} - C_{13}^2) + C_{23}(C_{13}C_{12} - C_{11}C_{23})$.

The deformation potential approximation is a widely used method for determining carrier mobility.[3-5] In the case of 2D structures, the calculation of carrier mobility follows a specific formula:[6]

$$\mu_{2D} = \frac{e\hbar^3 C_{2D}}{k_B T m^* \sqrt{m_x m_y} E_d^2}, \tag{S10}$$

where e is the elementary charge, C_{2D} denotes for the elastic modulus, E_d signifies the deformation potential constant, k_B and $\hbar$ correspond to the Boltzmann constant and the reduced Planck constant, m^* represents for effective mass and the average effective mass of carriers, and T = 300 K is the temperature utilized in these calculations.

The effective masses of electrons (m_e^*) and holes (m_h^*) can be determined by fitting a parabolic function to the band edges (CBM and VBM) using the following expression:

$$\frac{1}{m^*} = \frac{1}{\hbar}\left|\frac{\partial^2 E(k)}{\partial k^2}\right|, \tag{S11}$$

where $E(k)$ represents the energy dependence on the wave vector k at the CBM/VBM in the k_x-k_y plane. The carrier transport characteristics are studied in two distinct directions within the reciprocal lattice space.

The 2D materials' elastic modulus C_{2D} is expressed as following:

$$C_{2D} = \frac{1}{S_{uni}} \frac{\partial^2 E_{tot}}{\partial \varepsilon_{uni}^2}, \tag{S12}$$

where S_{uni}, E_{tot} and ε_{uni} symbolize the optimized unit cell area, total energy and uniaxial strain along the x and y carrier transport directions, respectively. The deformation potential constant E_d is determined using the formula:

$$E_d = \frac{\Delta E_{edge}}{\varepsilon_{uni}}, \tag{S13}$$

the term ΔE_{edge} denotes the energy shifting of the band edges relative to the vacuum level.

Table S1 Band gaps (eV) of CrSBr and Cr*X*S*Y*Br*Z* in GGA+*U* calculation and GGA+*U* calculation with spin-orbit coupling (SOC).

Band gap	CrSBr	Cr_2S_2BrH	Cr_2S_2BrF	Cr_2S_2BrCl	Cr_2SSeBr_2	$CrMoS_2Br_2$
GGA+*U*	0.65	0.47	0.56	0.69	0.32	0.002
SOC	0.61	0.46	0.19	0.23	0.10	Metal

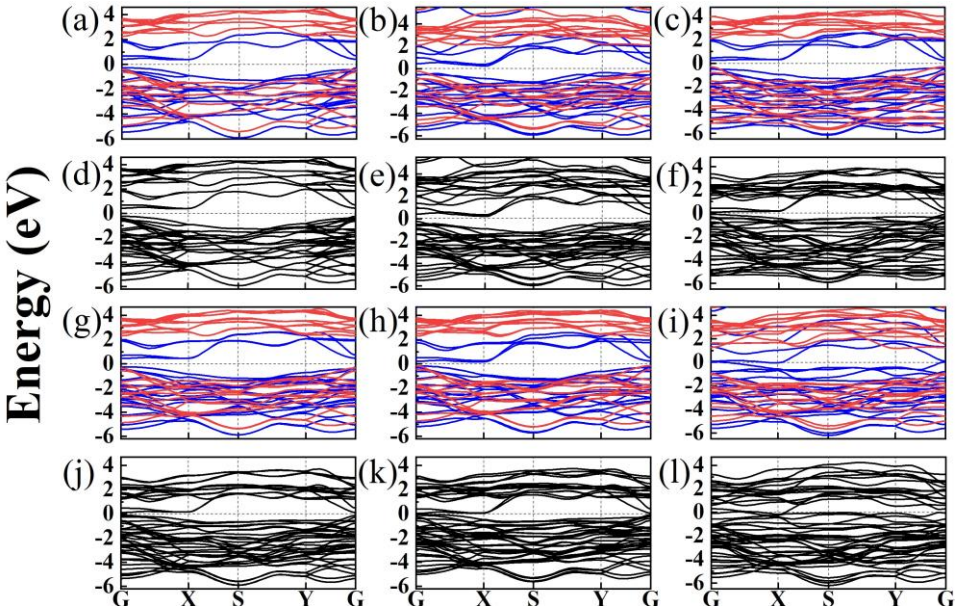

Fig. S1 Electronic band structures of (a), (d) CrSBr, (b), (e) Cr_2S_2BrH, (c), (f) Cr_2S_2BrF, (g), (j) Cr_2S_2BrCl, (h), (k) Cr_2SSeBr_2, and (i), (l) $CrMoS_2Br_2$ in GGA + *U* calculation ((a) – (c) and (g) – (i)) and GGA + *U* calculation with SOC ((d) – (f) and (j) – (l)). U^{Cr} = 3 eV and U^{Mo} = 2 eV, blue and red lines manifest spin-up and spin-down, respectively.

Table S2 Relative total energies $\triangle E$ (meV/unit cell), local spin moments (μ_B) of atoms in Cr*X*S*Y*Br*Z* monolayers by GGA+*U*.

Material	states	$\triangle E$	$Z1^{-}$	$X1^{3+}$	$Y1^{2-}$	$S2^{2-}$	$Cr2^{3+}$	Br^{2-}
CrSBr	FM	-64	-0.06	3.25	-0.22	-0.22	3.25	-0.06
	AFM	49	0.05	3.19	0.03	-0.03	-3.19	-0.05
Cr_2S_2BrH	FM	-36	-0.06	3.28	-0.21	-0.25	3.22	-0.06
	AFM	108	0.05	-3.19	-0.03	0.07	3.13	-0.04
Cr_2S_2BrF	FM	-36	0.01	3.22	-0.21	-0.24	3.21	-0.05
	AFM	83	-0.04	-3.15	-0.01	-0.03	3.16	0.05
Cr_2S_2BrCl	FM	-37	-0.04	3.24	-0.22	-0.23	3.24	-0.06
	AFM	75	-0.05	-3.17	0.04	-0.03	-3.18	0.04
Cr_2SSeBr_2	FM	-82	-0.07	3.27	-0.26	-0.24	3.34	-0.06
	AFM	33	0.05	-3.24	-0.05	0.03	3.27	-0.05
$CrMoS_2Br_2$	FM	-77	0.04	2.65	-0.16	-0.13	3.40	-0.13
	AFM1	109	0.07	2.66	0.09	0.07	-3.19	0.04
	AFM2	109	-0.07	-2.66	-0.09	-0.07	3.19	-0.04

Table S3 Intralayer exchange parameters J_1, J_2 and J_3 of Cr_2S_2BrZ and Cr_2SSeBr_2 monolayers (in units of meV).

	Cr_2S_2BrH	Cr_2S_2BrF	Cr_2S_2BrCl	Cr_2SSeBr_2
J_1	2.52	2.23	3.97	4.33
J_2	6.90	5.80	5.45	5.56
J_3	0.84	1.87	2.78	0.54

Table S4 The contribution of the piezoelectric stress coefficients e_{33} electron part e_{33}-*ele* and the ion part e_{33}-*ion* (10^{-10} C/m) in Cr*X*S*Y*Br*Z* monolayers.

Material		e_{33}-*ele*	e_{33}-*ion*	e_{33}
CrSBr	GGA	0.0	0.0	0.0
	GGA+*U*	0.0	0.0	0.0
Cr_2S_2BrH	GGA	-0.57	-0.36	-0.93
	GGA+*U*	-0.52	-0.35	-0.87
Cr_2S_2BrF	GGA	-1.70	0.16	-1.54
	GGA+*U*	-1.57	0.11	-1.46
Cr_2S_2BrCl	GGA	-0.47	0.07	-0.40
	GGA+*U*	-0.46	0.07	-0.39
Cr_2SSeBr_2	GGA	0.34	-0.06	0.28
	GGA+*U*	0.34	-0.06	0.28
$CrMoS_2Br_2$	GGA	0.64	-0.005	0.64
	GGA+*U*	0.50	-0.004	0.49

REFERENCES

1. L. Dong, J. Lou and V. B. Shenoy, *Acs Nano*, 2017, **11**, 8242-8248.
2. H. N. Zhang, Y. Wu, C. H. Yang, L. H. Zhu and X. C. Wang, *Physical Review B*, 2021, **104**, 235437.
3. J. Bardeen and W. Shockley, *Physical Review*, 1950, **80**, 72-80.
4. N. N. Hieu, H. V. Phuc, A. I. Kartamyshev and T. V. Vu, *Physical Review B*, 2022, **105**, 075402.
5. T. Zhang, Y. D. Ma, L. Yu, B. B. Huang and Y. Dai, *Materials Horizons*, 2019, **6**, 1930-1937.
6. W. H. Wan, S. Zhao, Y. F. Ge and Y. Liu, *Journal of Physics-Condensed Matter*, 2019, **31**, 435501.