Supporting Information for

Tunable valley-selective circular polarization in vdW multilayer consist of inversion-symmetric monolayers

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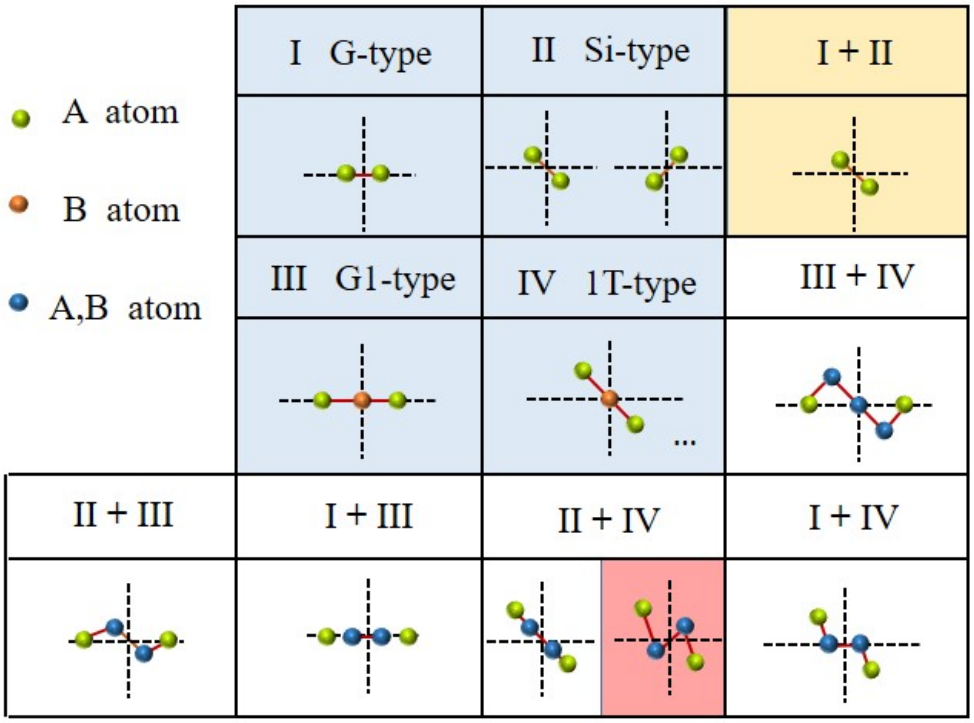

Fig. S1 Schematic diagram of inversion symmetry structure. The green and orange balls represents the A and B atoms, respectively. The blue balls represents the atom that replaces B atom when A exist. Four structures in the blue background (I, II, III and IV) represent the simplest inversion symmetry configurations, and others are combinations of them. The yellow background means that it is impossible to get out-of-plane electrical polarization by constructing homostructure due to it has only one type of atom. Red background represent the structure that we will discuss below.

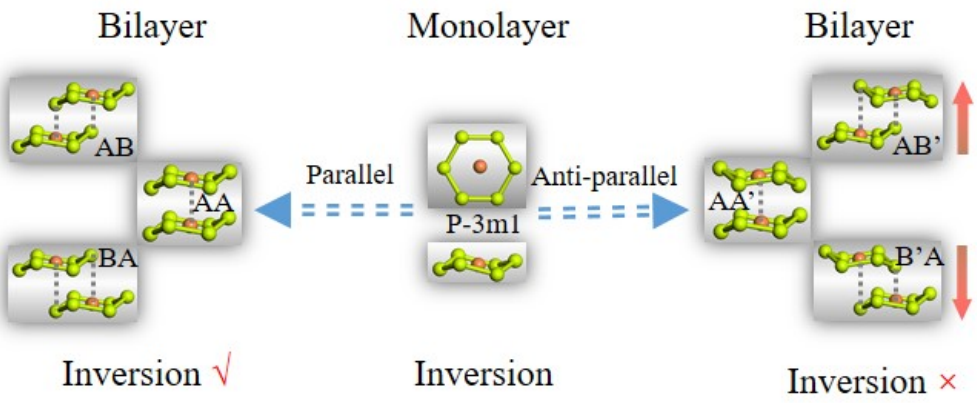

Fig. S2 Schematic diagram of inversion symmetry monolayer with space group of P-3m1 and possible bilayer. Two monolayers parallel stacking directly without relative rotation is AA bilayer, and under the translation operation t(1/3,-1/3,0)/t(1/3,2/3,0)/t(-2/3,-1/3,0) and t(2/3,1/3,0)/t(-1/3,1/3,0)/t(-1/3,-2/3,0) obtain the AB and BA, respectively. The parallelly-stacked bilayers keeps the inversion symmetry. Similarly, two monolayers anti-parallel stacking with 180° rotation is AA' bilayer, and under the translation operation t(1/3,-1/3,0)/t(1/3,2/3,0)/t(-2/3,-1/3,0) and t(2/3,1/3,0)/t(-1/3,1/3,0)/t(-1/3,-2/3,0) obtain the AB' and B'A, respectively. The anti-parallelly-stacked bilayers breaks the inversion symmetry. The red arrows represent the interlayer electric polarization.

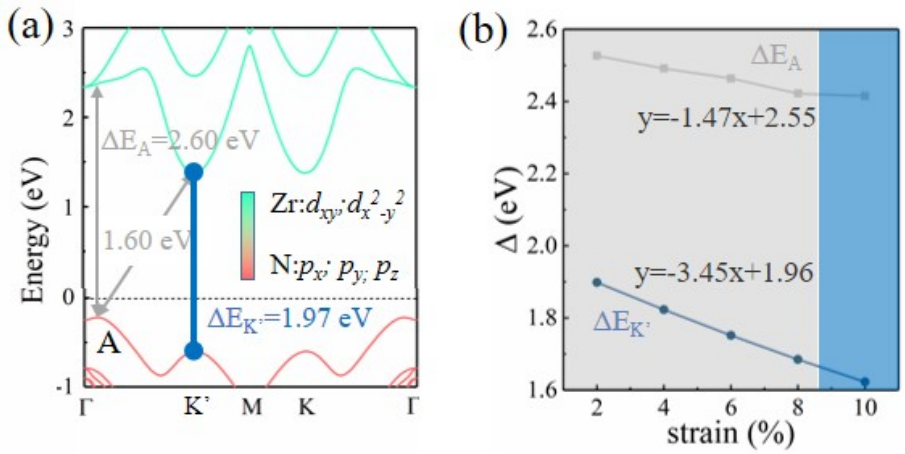

Fig. S3 (a) Band structure of ZrNBr monolayer. The direct band gap at A and K' is $\triangle E_{K'}$=1.97 eV and $\triangle E_A$=2.60 eV, and the indirect band gap is 1.60 eV. The light blue and orange line represent the contribution of Zr (d_{xy}, $d_{x^2-y^2}$) and N (p_x, p_y, p_z). (b) The direct band gap at A and K' point as the function of biaxial strain. A direct-band-gap semiconductor was shown in blue background.

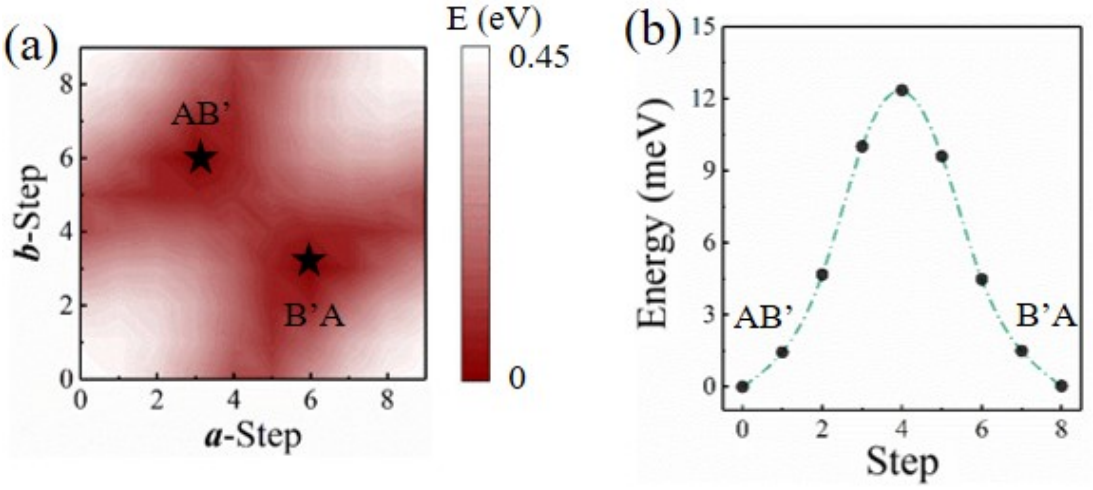

Fig. S4 The energy profile of anti-parallelly-stacked ZrNBr bilayer with interlayer sliding along two basic vectors. (c) The potential barrier from AB' to B'A along the minimum energy path by NEB calculation.

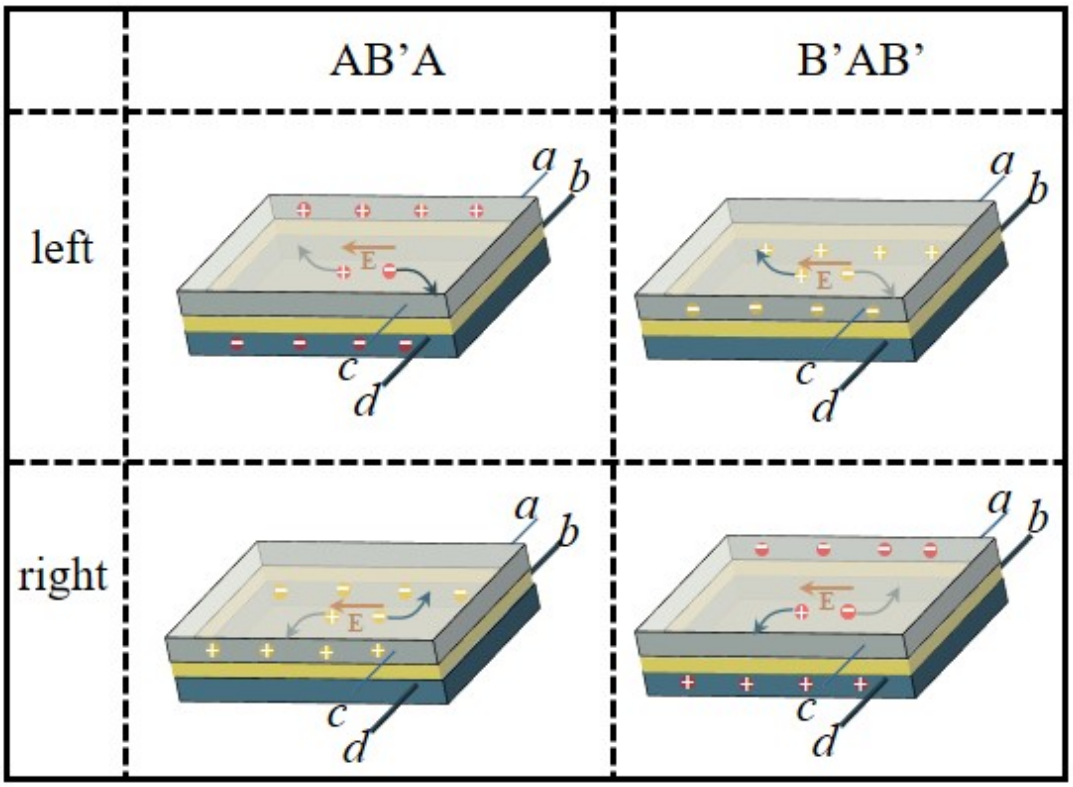

Fig. S5 The diagram of spatial separation of electrons and holes in AB'A and B'AB' trilayers under left- (left) and right-polarized (right) light, which same as **Fig. 5**.

Table SI The voltage between different sides in trilayer under left- and right-polarized light. The 0 represent no voltage.

	AB'A		B'AB'	
left	ab=0	cd=0	ab=0	cd=0
	ac=1	bd=1	ac=1	bd=1
right	ab=0	cd=0	ab=0	cd=-0
	ac=-1	bd=-1	ac=-1	bd=-1