Integrating Ferromagnetism and Ferroelectricity in an Iron Chalcogenide Monolayer: A First-Principle Study

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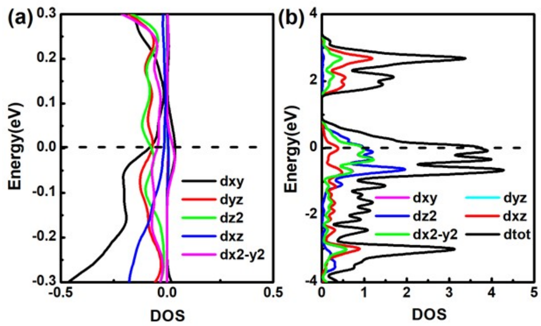

Figure.S1 The density of states (DOS) of FeSSe. (a) spin-polarized (b) non-spin-polarized.

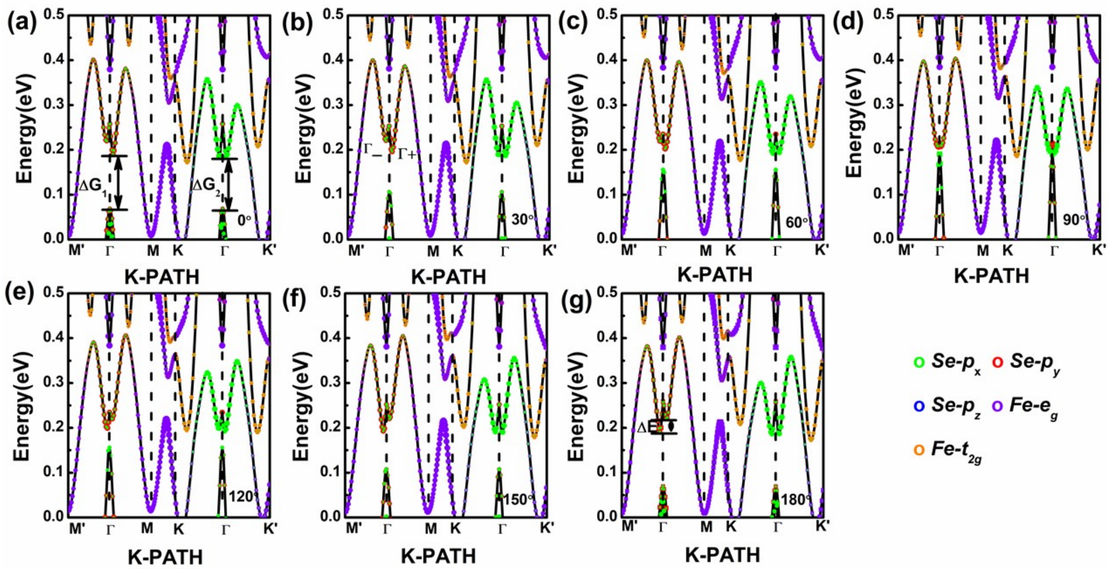

Figure.S2 The orbital-resolved band structure of FeSSe as a function of out-of-plane angle θ with SOC (φ=90º). The green, red, blue, orange and purple represent Se-p_x, Se-p_y, Se-p_z, Fe-t_{2g} (d_{xy}, d_{yz}, d_{xz}) and Fe-e_g (dx^2-y^2, dz^2). (a) [0º, 90º]; (b) [30º, 90º]; (c) [60º, 90º]; (d) [90º, 90º]; (e) [120º, 90º]; (f) [150º, 90º]; (g) [180º, 90º].

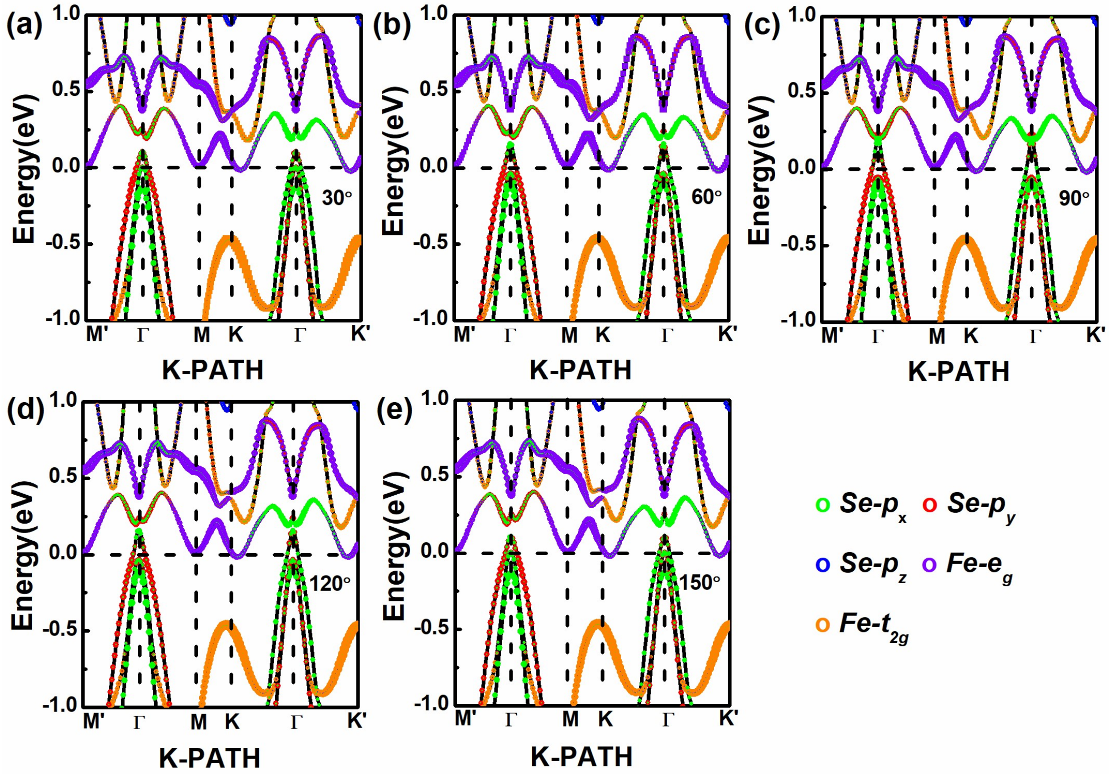

Figure.S3 (a-f) The orbital-resolved band structure of FeSSe as a function of out-of-plane angle θ with SOC (φ=0º). The green, red, blue, orange and purple represent Se-p_x, Se-p_y, Se-p_z, Fe-t_{2g} (d_{xy}, d_{yz}, d_{xz}) and Fe-e_g (dx^2-y^2, dz^2). (a) [30º, 0º]; (b) [60º, 0º]; (c) [90º, 0º]; (d) [120º, 0º]; (e) [150º, 0º].

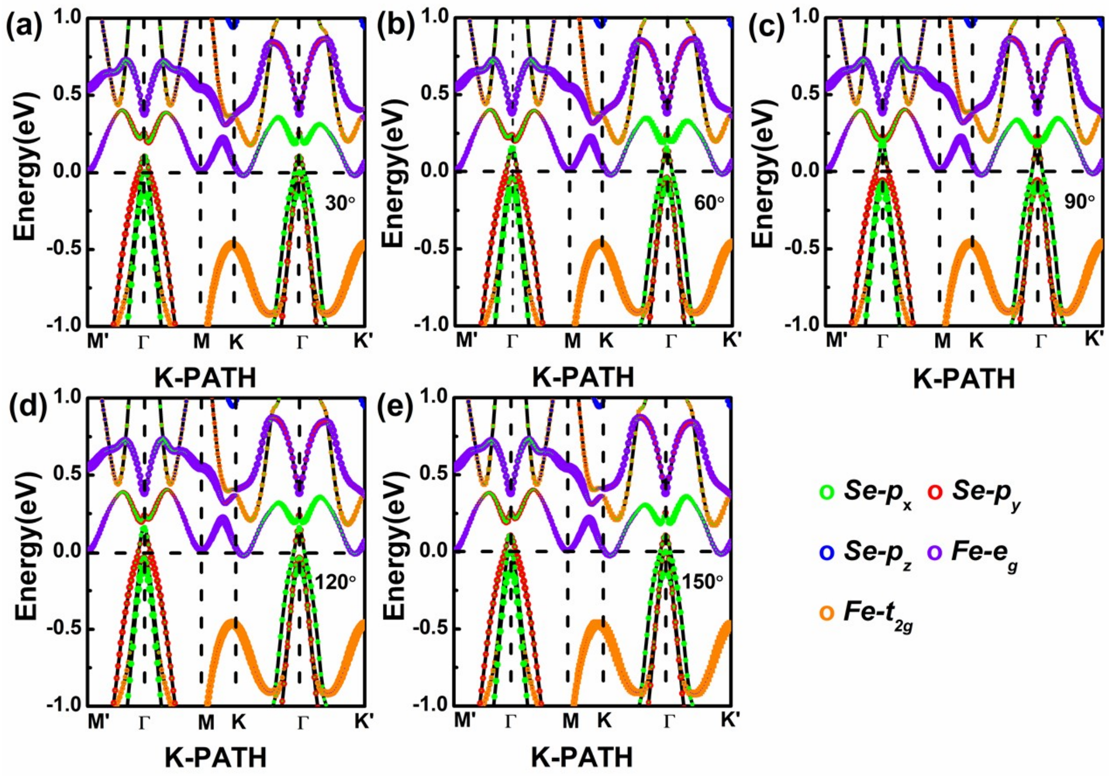

Figure.S4 (a-f) The orbital-resolved band structure of FeSSe as a function of out-of-plane angle θ with SOC (φ=30º). The green, red, blue, orange and purple represent Se-p_x, Se-p_y, Se-p_z, Fe-t_{2g} (d_{xy}, d_{yz}, d_{xz}) and Fe-e_g (dx^2-y^2, dz^2). (a)

[30º, 30º]; (b) [60º, 30º]; (c) [90º, 30º]; (d) [120º, 30º]; (e) [150º, 30º].

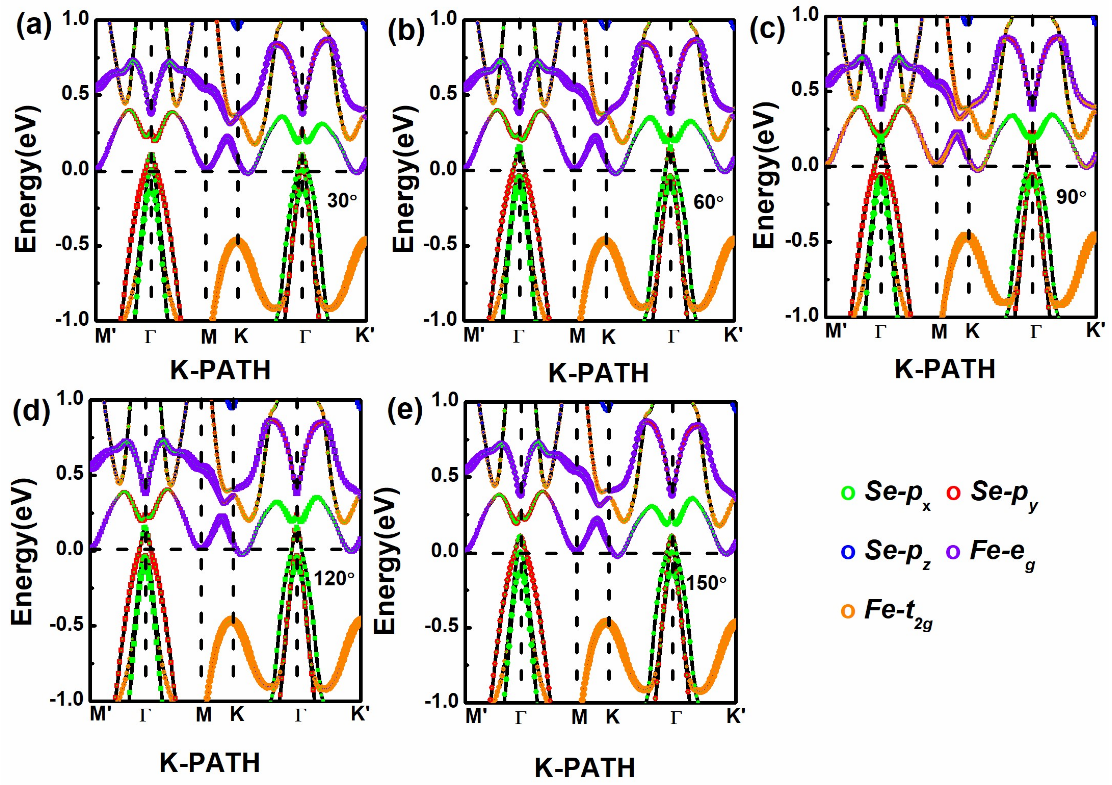

Figure.S5 (a-f) The orbital-resolved band structure of FeSSe as a function of out-of-plane angle θ with SOC (φ=60º). The green, red, blue, orange and purple represent Se-p_x, Se-p_y, Se-p_z, Fe-t_{2g} (d_{xy}, d_{yz}, d_{xz}) and Fe-e_g (dx^2-y^2, dz^2). (a) [30º, 60º]; (b) [60º, 60º]; (c) [90º, 60º]; (d) [120º, 60º]; (e) [150º, 60º].

Table.S1 The value G_1, G_2 and ΔE for different [θ,φ].

	[0º,0º]	[30º,0º]	[60º,0º]	[90º,0º]	[120º,0º]	[150º,0º]	[180º,0º]
ΔG_1(meV)	121.31	89.62	46.75	16.82	44.71	86.71	121.36
ΔG_2(meV)	117.38	84.79	37.31	0	36.00	78.78	116.43
ΔE(meV)	29.24	27.66	21.73	0	-14.94	-23.62	-29.00
	[0º,30º]	[30º,30º]	[60º,30º]	[90º,30º]	[120º,30º]	[150º,30º]	[180º,30º]
ΔG_1(meV)	121.31	90.43	47.04	17.20	43.70	85.96	121.36
ΔG_2(meV)	117.38	84.21	36.94	0	37.75	80.17	116.43
ΔE(meV)	29.24	26.69	20.52	0	-16.29	-24.63	-29.00
	[0º,60º]	[30º,60º]	[60º,60º]	[90º,60º]	[120º,60º]	[150º,60º]	[180º,60º]
ΔG_1(meV)	121.31	90.88	47.32	17.60	43.04	85.78	121.36
ΔG_2(meV)	117.38	84.62	36.69	0	40.26	82.07	116.43
ΔE(meV)	29.24	25.41	18.58	0	-18.30	-25.76	-29.00

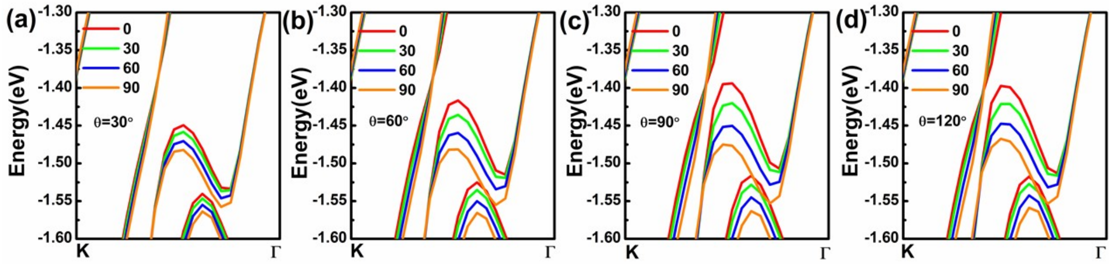

Figure.S6 (a-d) The band structure of FeSSe as a function of in-plane angle with SOC. The red, green, orange and blue represent φ = 0, 30º, 60º, 90º. (a) [30º, φ]; (b) [60º, φ]; (c) [90º, φ]; (d) [120º, φ].

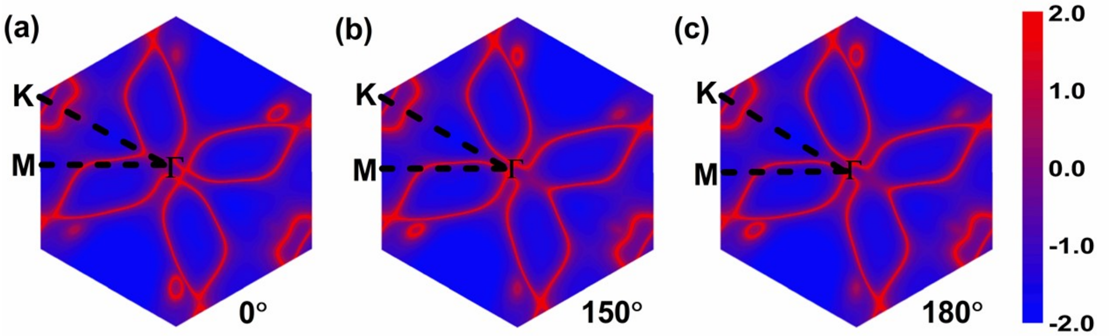

Figure.S7 Magnetization direction-dependent Fermi surfaces of FeSSe. (a) [0º, 90º]; (b) [150º, 90º]; (c) [180º, 90º].

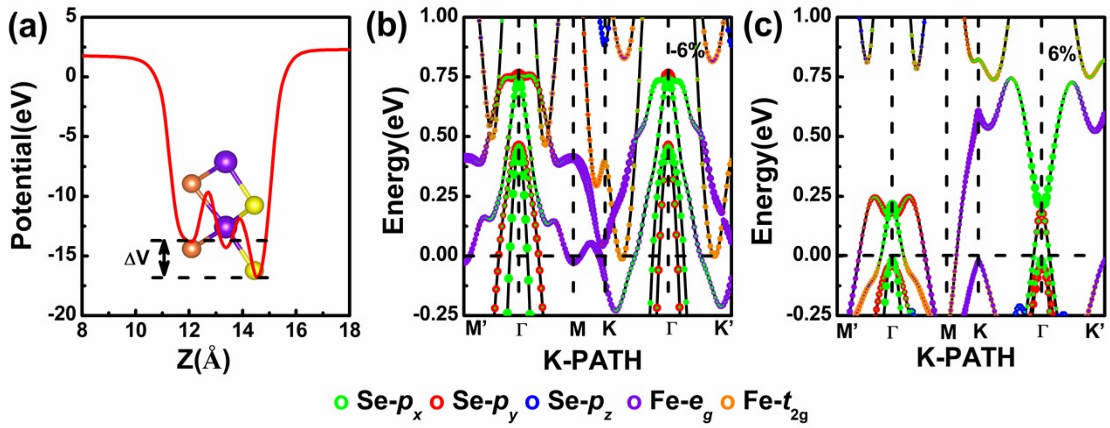

Figure.S8 (a) The average electrostatic potential perpendicular to the surface (ΔV) of FeSSe with FM state without strain. (b-c) The orbital-resolved band structure of FeSSe as a function of biaxial strain with SOC (θ=90º, φ=90º). The green, red, blue, orange and purple represent Se-p_x, Se-p_y, Se-p_z, Fe-t_{2g} (d_{xy}, d_{yz}, d_{xz}) and Fe-e_g (dx^2-y^2, dz^2). (b) -6%; (c) 6%. The color intensity denotes the amplitude of the orbital-resolved character.

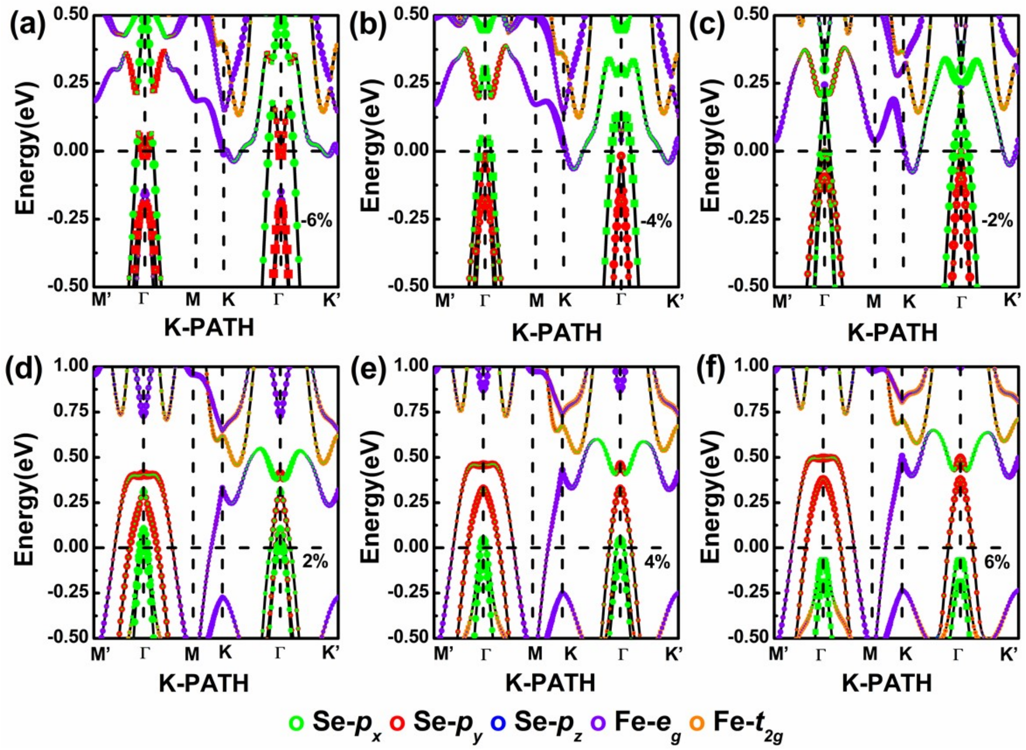

Figure.S9 The orbital-resolved band structure of FeSSe as a function of uni-axially strain with SOC (θ=90º, φ=90º). The green, red, blue, orange and purple represent Se-p_x, Se-p_y, Se-p_z, Fe-t_{2g} (d_{xy}, d_{yz}, d_{xz}) and Fe-e_g (dx^2-y^2, dz^2). (a) -6%; (b) -4%; (c) -2%; (d) 2%; (e) 4%; (f) 6%. The color intensity denotes the amplitude of the orbital-resolved character.

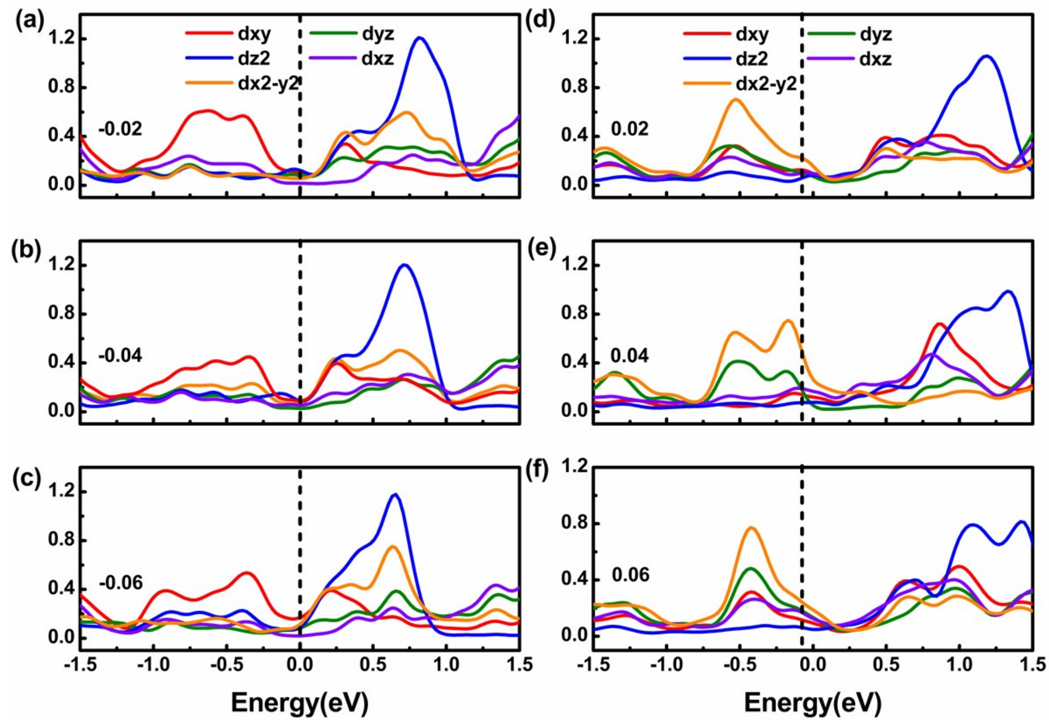

Figure.S10 Project density of state of FeSSe monolayer as a function of biaxial strain in the range of -6% to 6%. (a) -6%; (b) -4%; (c) -2%; (d) 2%; (e) 4%; (f) 6%.