Supplementary Material

Origins of Electrostriction of MoS_2 and HfS_2 in 2 and 3 dimensional *1T* and *2H* structures

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I. Electrostriction as stress/strain derivative of permitivity/dielectric susceptibility

To confirm the thermodynamic validity of the methodology, we derive the coefficients Q_{ijkl} starting from the elastic Gibbs free energy density for a bulk material. The elastic Gibbs free energy density G_1 is expressed as

$$G_1 = u - Ts - X_{kl}x_{kl} - E_iP_i,$$

where u is the internal energy per unit volume (J/m^3), T is the temperature (K), s is the entropy density (J/m^3), X_{kl} is the stress tensor (Pa), x_{kl} is the strain tensor (dimensionless), E_i is the electric field (V/m), and P_i is the polarization (C/m^2).

Taking the differential of G_1, we have

$$dG_1 = -s\,dT - x_{kl}\,dX_{kl} - P_i\,dE_i - E_i\,dP_i.$$

Considering isothermal processes and focusing only on electromechanical deformations, the relevant terms are

$$dG_1 = -x_{kl}\,dX_{kl} - E_i\,dP_i.$$

The relationships between the elastic Gibbs free energy and the stress or electric field are then given by:

$$\frac{\partial G_1}{\partial X_{ij}} = -x_{ij}, \quad \frac{\partial G_1}{\partial P_i} = -E_i.$$

Substituting the functional forms, we have

$$x_{ij} = g_{ijk}P_k + Q_{ijkl}P_kP_l, \quad E_i = \eta_{ij}P_j,$$

where η_{ij} is the dielectric stiffness tensor.

To derive the coupling between polarization and stress, we calculate the third derivatives of G_1. From the strain expression,

$$x_{kl} = g_{klm}P_m + Q_{klmn}P_mP_n,$$

we compute the second derivatives with respect to P_i and P_j:

$$\frac{\partial^2 x_{kl}}{\partial P_i \partial P_j} = 2Q_{ijkl}.$$

Thus:

$$-\frac{\partial^2 x_{kl}}{\partial P_i \partial P_j} = -2Q_{ijkl}.$$

The same result can be obtained by computing the third derivative of G_1 directly:

$$\frac{\partial^3 G_1}{\partial P_j \partial P_i \partial X_{kl}} = -\frac{\partial^2 x_{kl}}{\partial P_i \partial P_j} = -2Q_{ijkl}.$$

Similarly, for the dielectric stiffness:

$$\frac{\partial^3 G_1}{\partial X_{kl}\partial P_j \partial P_i} = \frac{\partial \eta_{ij}}{\partial X_{kl}}.$$

By Schwarz's theorem, the mixed derivatives of G_1 are equal:

$$\frac{\partial^3 G_1}{\partial P_j \partial P_i \partial X_{kl}} = \frac{\partial^3 G_1}{\partial X_{kl}\partial P_j \partial P_i}.$$

Equating the two expressions, we find

$$-2Q_{ijkl} = \frac{\partial \eta_{ij}}{\partial X_{kl}}.$$

Finally, the electrostrictive coefficient is expressed as:

$$Q_{ijkl} = -\frac{1}{2}\frac{\partial \eta_{ij}}{\partial X_{kl}}.$$

II. Born effective charges of bulk $d1T$-MoS$_2$ and $2H$-HfS$_2$

Table 1: Born effective charges ($Z^*_{\alpha,\beta}$, α, β= x,y) of bulk $d1T$-MoS_2.

Atom	direction	x	y	z
Mo	x	2.3	0.0	2.3
	y	0.0	-5.8	0.0
	z	0.6	0.0	-0.5
Mo	x	-3.8	-3.5	-1.1
	y	-3.5	0.3	2.0
	z	-0.3	0.5	-0.5
Mo	x	-3.8	3.5	-1.1
	y	3.5	0.3	-2.0
	z	-0.3	-0.5	-0.5
S	x	1.1	0.7	0.0
	y	-0.7	1.1	0.0
	z	0.0	0.0	0.5
S	x	1.1	-0.7	0.0
	y	0.7	1.1	0.0
	z	0.0	0.0	0.5
S	x	-2.3	0.0	0.0
	y	0.0	-2.3	0.0
	z	0.0	0.0	-0.4
S	x	2.3	0.0	-0.3
	y	0	1.2	0.0
	z	0.1	0.0	0.4
S	x	1.5	-0.5	0.1
	y	-0.5	2.0	-0.2
	z	0.0	0.1	0.4
S	x	1.5	0.5	0.1
	y	0.5	2.0	0.2
	z	0.0	-0.1	0.4

Table 2: Born effective charges ($Z^*_{\alpha,\beta}$, α, β= x,y) of bulk 2H-HfS_2.

Atom	direction	x	y	z
Hf	x	4.4	0.0	0.0
	y	0.0	4.4	0.0
	z	0.0	0.0	3.3
Hf	x	4.4	0.0	0.0
	y	0.0	4.4	0.0
	z	0.0	0.0	3.3
S	x	-2.2	0.0	0.0
	y	0.0	-2.2	0.0
	z	0.0	0.0	-1.7
S	x	-2.2	0.0	0.0
	y	0.0	-2.2	0.0
	z	0.0	0.0	-1.7
S	x	-2.2	0.0	0.0
	y	0.0	-2.1	0.0
	z	0.0	0.0	-1.7
S	x	-2.2	0.0	0.0
	y	0.0	-2.1	0.0
	z	0.0	0.0	-1.7