补充材料

$MoSi_2N_4$ 的本征点缺陷以及掺杂特性的第一性原理计算研究

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表 S1 固体 Si，Mo 原子以及 N2 分子和其他二元化合物的形成能计算结果

Table S1. Formation energy values of bulk Si atom, N_2 molecule, Mo atom and their binary compounds.

材料	空间群 (pace group)	化合物总能量/eV	形成能/eV
Mo	*P6/mmm*	-13.00	0
N	*Pnma*	-10.23	0
Si	*Pm3n*	-6.29	0
MoN	*P63mc*	-24.05	-0.81
Si_3N_4	*P63/m*	-68.44	-8.63
$MoSi_2N_4$		-73.78	-7.28
$SiMo_3$	*Pm3n*	-50.97	-5.66
$Mo_{15}N_{16}$	*Cc*	-370.82	-11.99
Si_3Mo_5	*I4/mcm*	-89.72	-5.82
Si_2Mo	*I4/mmm*	-22.41	-1.49

表 S2 不同缺陷的形成能量随晶格参数改变而变化的趋势，包括参不参与校正能量的情况（所有结果均来源于 hse 计算）

Table S2. Formation energy of different defects with or without correction energy varying with lattice parameter, with all the results derived from hse calculations.

缺陷种类	电荷	不同晶格放大倍数情况下的能量 $(1/\alpha, \alpha = 4,6,8)$/eV					
		0.125		0.166		0.25	
		无校正	有校正	无校正	有校正	无校正	有校正
V_{Mo}	-*q*	9.93	10.00	10.03	10.09	10.26	10.11
	-2*q*	11.56	12.14	11.28	11.82	11.18	10.52
	q	9.21	9.20	8.97	8.91	8.93	8.72
	2*q*	10.03	10.37	9.74	10.23	9.62	9.03
	Neutral	10.83	10.83	10.78	10.78	10.91	10.91
V_{Si}	-*q*	10.65	10.44	10.84	10.89	11.18	11.06
	-2*q*	11.40	11.39	12.00	11.81	12.19	11.73
	q	10.91	11.04	10.85	10.80	10.64	10.43
	2*q*	12.14	12.06	11.84	11.72	11.63	11.10
	Neutral	10.72	10.72	10.75	10.75	10.84	10.84

V_{N1}	-q	7.22	7.22	7.26	7.06	7.16	7.05
	-2q	10.56	10.64	10.40	9.69	10.28	9.52
	q	4.65	4.75	4.71	4.59	4.63	4.38
	2q	4.96	5.23	5.34	4.83	5.26	4.28
	Neutral	7.18	7.18	7.12	7.12	7.26	7.26
V_{N2}	-q	5.64	5.66	5.52	5.54	5.48	5.50
	-2q	8.15	8.16	7.62	8.01	7.58	7.97
	q	3.01	2.98	2.87	2.84	2.81	2.73
	2q	8.12	8.17	7.99	8.04	7.94	8.02
	Neutral	6.68	6.68	6.72	6.72	6.81	6.81
Mo_N	-q	9.33	9.48	9.40	9.57	9.54	9.84
	-2q	12.93	13.46	12.99	13.60	12.80	11.96
	q	6.68	6.76	6.72	6.78	6.53	6.23
	2q	7.16	7.53	7.19	7.72	7.15	7.13
	Neutral	6.85	6.85	6.91	6.91	7.02	7.02
Si_N	-q	5.90	5.97	5.98	6.05	6.05	6.09
	-2q	8.95	9.38	9.10	9.57	9.20	8.66
	q	5.37	5.54	5.42	5.60	5.36	5.47
	2q	6.36	6.83	6.52	7.00	6.01	5.42
	Neutral	5.33	5.33	5.41	5.41	5.44	5.44
Si_{Mo}	-q	5.29	5.42	5.31	5.47	5.17	5.33
	-2q	7.14	7.50	7.19	7.60	6.86	7.63
	q	3.82	3.94	3.84	3.95	3.81	3.96
	2q	4.56	4.89	4.61	4.99	4.38	4.93
	Neutral	4.18	4.18	4.22	4.22	4.27	4.27
Mo_{Si}	-q	6.44	6.51	6.61	6.68	13.71	13.74
	-2q	9.70	10.05	9.89	10.27	9.55	9.90
	q	3.72	3.82	3.90	4.02	3.80	3.92
	2q	4.36	4.80	4.54	5.01	4.09	4.85
	Neutral	4.08	4.08	4.21	4.21	4.32	4.32
$N1_{Si}$	-q	11.39	11.43	11.49	11.53	10.94	10.97
	-2q	13.13	13.45	13.23	13.58	12.75	13.10
	q	10.39	10.52	10.63	10.76	10.29	10.43
	2q	11.16	11.49	11.40	11.74	10.89	11.66
	Neutral	10.46	10.46	10.56	10.56	10.26	10.26
$N2_{Si}$	-q	9.46	9.58	9.34	9.38	9.30	9.34
	-2q	12.34	12.48	12.23	12.38	12.14	12.28
	q	8.41	8.52	8.35	8.42	8.25	8.32
	2q	7.92	8.01	7.86	7.94	7.66	7.74
	Neutral	9.26	9.26	9.26	9.26	9.26	9.26
$N1_{Mo}$	-q	11.76	12.01	11.85	12.00	11.60	11.86
	-2q	14.42	14.95	14.51	15.19	14.02	14.46
	q	10.13	10.33	10.23	10.34	10.30	10.36

	$2q$	10.90	11.30	11.00	11.60	10.95	11.31
	Neutral	10.40	10.40	10.49	10.49	10.50	10.50
$N2_{Mo}$	$-q$	9.63	9.81	9.56	9.75	9.44	9.63
	$-2q$	13.46	13.64	13.37	13.57	13.29	13.43
	q	8.17	8.09	8.11	8.03	8.03	7,96
	$2q$	6.89	6.75	6.83	6.70	6.74	6.62
	Neutral	8.89	8.89	8.89	8.89	8.89	8.89

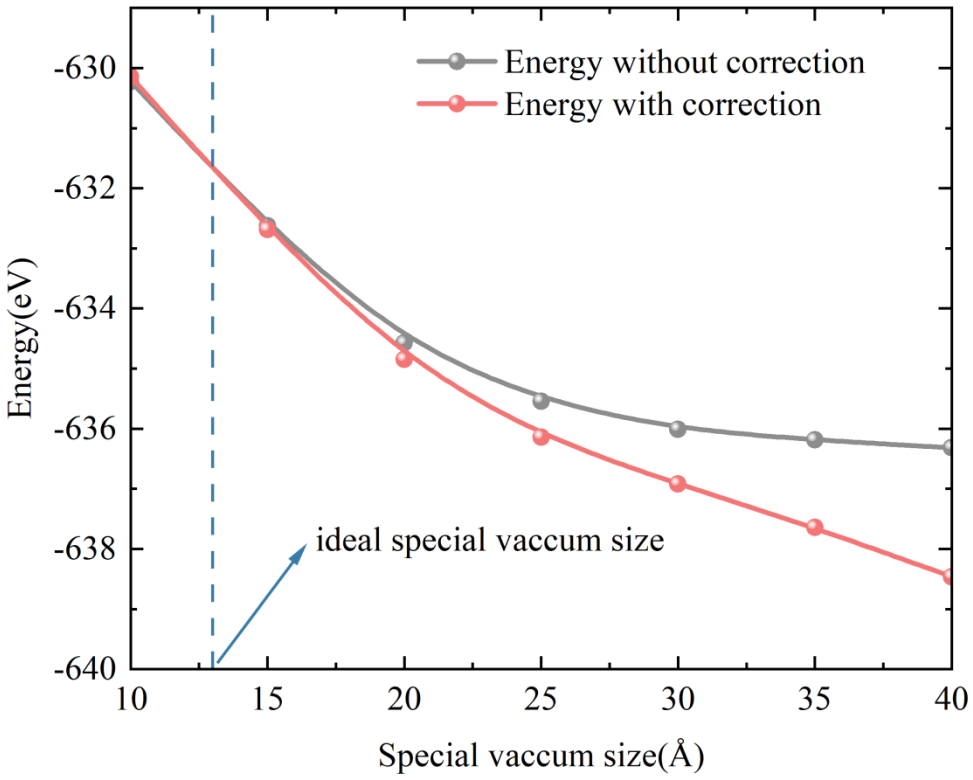

图 S1 硅空位缺陷的总能量随着特殊真空尺寸（SVS）增加的变化（包括是否进行校正）。在此我们选择 SVS 的尺寸为 13Å，以消除缺陷的真空空间带来的影响[1]

Fig. S1. Variations of total energywith special vacuum size(SVS) of Si vacancy defect with or without correction energy, with SVS chosen as 13 Å to minimize the effect of vacuum space of all kinds of defects[1].

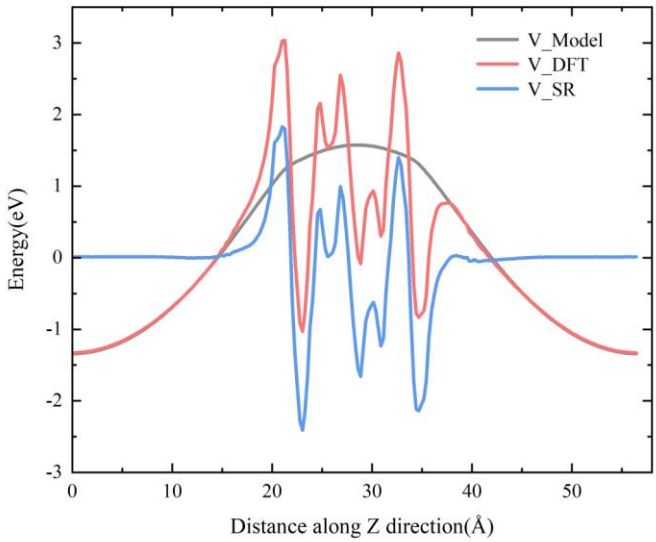

图 S2 Freysoldt-Neugebauer 方法的校正过程，通过调整校正参数以及增加校正能量，我们保证两个原胞边界上的短程势为零[2]

Fig. S2. The correction process of Freysoldt-Neugebauer method. Though adjusting the correction parameter and adding the correction energy, we hope to keep the short range electral potential zero (only long range electral potential exists) in the boundary between two volume[2].

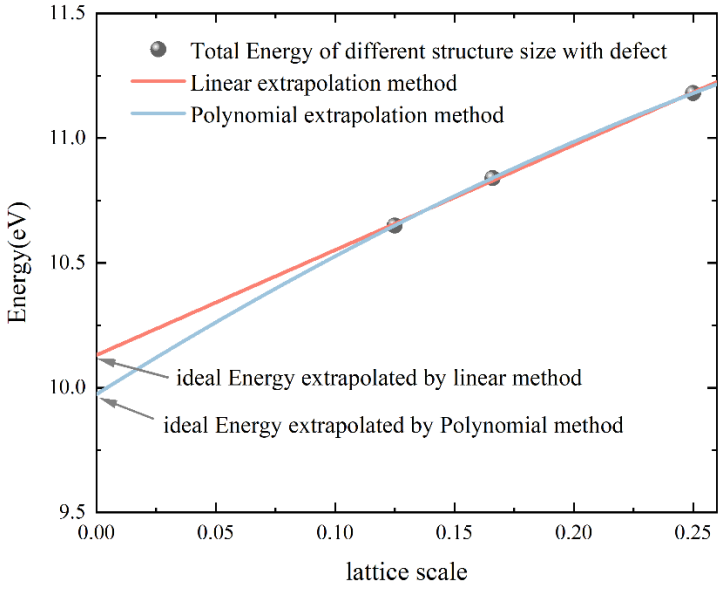

图 S3 在硅空位缺陷上比较 Kosma 等建议的高次外推方法和我们使用的线性方法的效果区别[3]

Fig. S3. The comparison of linear extropolation method suggested by Kosma and linear extrapolation method we used in our work of V_{Si} defects. It is obvious that there is no large difference between these two methods in $MoSi_2N_4$. In light of the limited data we can obtain from enlarging the structure and the deviation in calculation, we employ linear method to verify our results[3].

[1] Komsa H-P, Pasquarello A 2013 *Phys. Rev. Lett.* **110** 095505

[2] Freysoldt C, Neugebauer J 2018 *Phys. Rev. B* **97** 205425

[3] Komsa H P, Berseneva N, Krasheninnikov A V, Nieminen R M 2018 *Physi.Rev. X* **8** 031044

