

Supplemental Information for "Electronic coupling between the FeSe monolayer film and SrTiO₃ substrate"

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(Dated: December 29, 2016)

I. ADDITIONAL DATA ON STRUCTURES

The positions of all atoms in our simulation cell are provided in Table I.

The van der Waals bonding between FeSe and SrTiO₃, with the uncertainty for DFT functionals for such layers, has made the separation of the two components uncertain. In Fig. 1 the band structures are shown for three distances(d_s) between Fe and TiO₂ termination layer that have been suggested in the literature. There are differences in Fe valence d bands, and also in the lowest conduction band, around Γ point near E_F . Note however that in each case. the O1 p_x, p_y interface band is slightly unoccupied at the M point.

II. ADDITIONAL DATA ON EFFECTS OF SUBSTRATE DOPING

Virtual crystal doping is simple and may be realistic for STO. Several corresponding fatbands plots are provided for 0, 5%, and 10% electron doping (VCA nuclear charges equal to 22.00, 22.05, and 22.10, respectively). Figure 2 displays the Fe d character, Fig. 3 the Ti1, Ti2, and Ti3

d character, and Fig. 4 the O1, O2, and O3 p character.

III. ADDITIONAL DATA ON ISOVALENT CHALCOGENIDE SUBSTITUTION

In the FeS_{0.5}Se_{0.5} and FeTe_{0.5}Se_{0.5} "alloy" we studied, an S or Te layer replaces the Se layer nearest the STO substrate. Corresponding band structure and Fermi surface as shown in Fig. 5 to allow assessment of the small differences.

IV. NITROGEN SUBSTITUTION FOR OXYGEN

Oxygen in STO can also be replaced to some extent by N.^{1,2} To provide some comparison along the lines we did for chalcogenide substitution, we provide orbital occupation differences and charge density rearrangement in the electron-rich substitutions of nitrogen on the oxygen site, denoted 1UC FeSe+STO_{1-x}N_x, $x=0.2$ and 0.8 , as shown in Table II, Fig. 6, and Fig. 7. For $x=0.2$, the O1 sites were replaced by N, for $x=0.8$, sites O2-O5 were replaced by N.

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² J. Xu, Y. Wei, Y. Huang, J. Wang, X. Zheng, Z. Sun, L. Fan, and J. Wu, Solvothermal synthesis nitrogen doped SrTiO₃ with high visible light photocatalytic activity, Ceramics International **40**, 10583 (2014).

TABLE I. The positions of each atom in the simulation cell when $d_s=4.19 \text{ \AA}$. The tetragonal lattice constants are $a=b=3.905 \text{ \AA}$, $c=23.28 \text{ \AA}$.

kinds of atoms	Se1	Fe	Se2	Ti1	O1	Sr1	O2	Ti2	O3	Sr2	O4	Ti3	O5
x	0.00	0.00	0.50	0.50	0.00/0.50	0.00	0.50	0.50	0.00/0.05	0.00	0.50	0.50	0.00/0.50
y	0.00	0.50	0.50	0.50	0.50/0.00	0.00	0.50	0.50	0.50/0.00	0.00	0.50	0.50	0.50/0.00
z	0.00	0.05	0.11	0.23	0.23	0.32	0.32	0.40	0.40	0.49	0.49	0.57	0.57

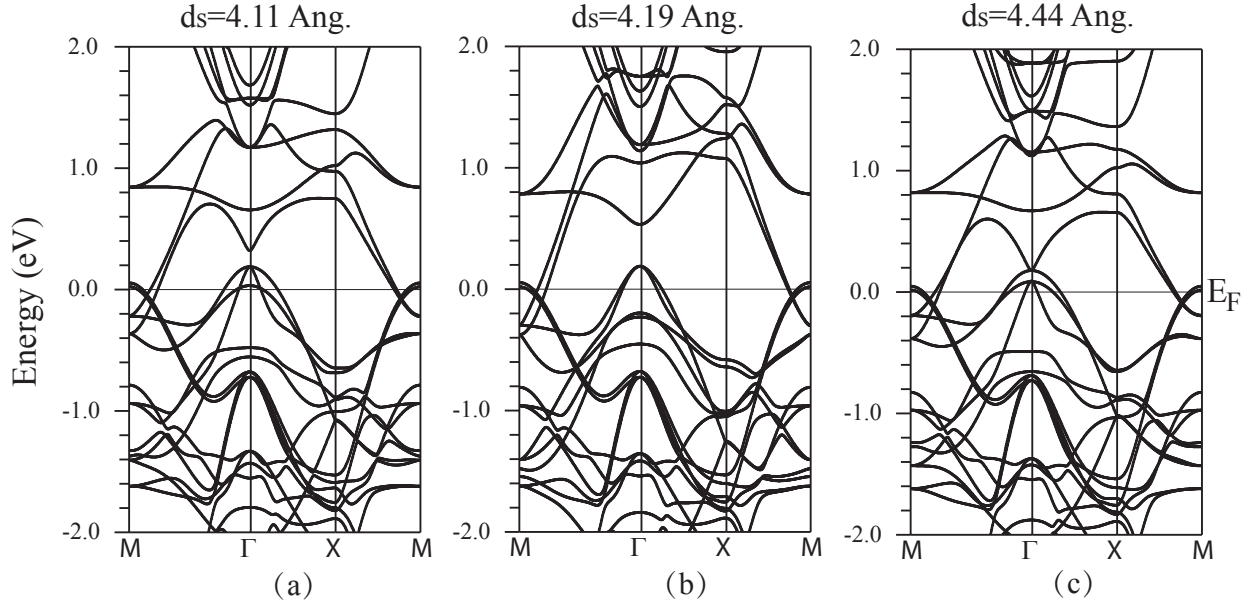


FIG. 1. (Color online) Band structures of three different structures with distances(d_s) equal to 4.11 \AA , 4.19 \AA , and 4.44 \AA , respectively.

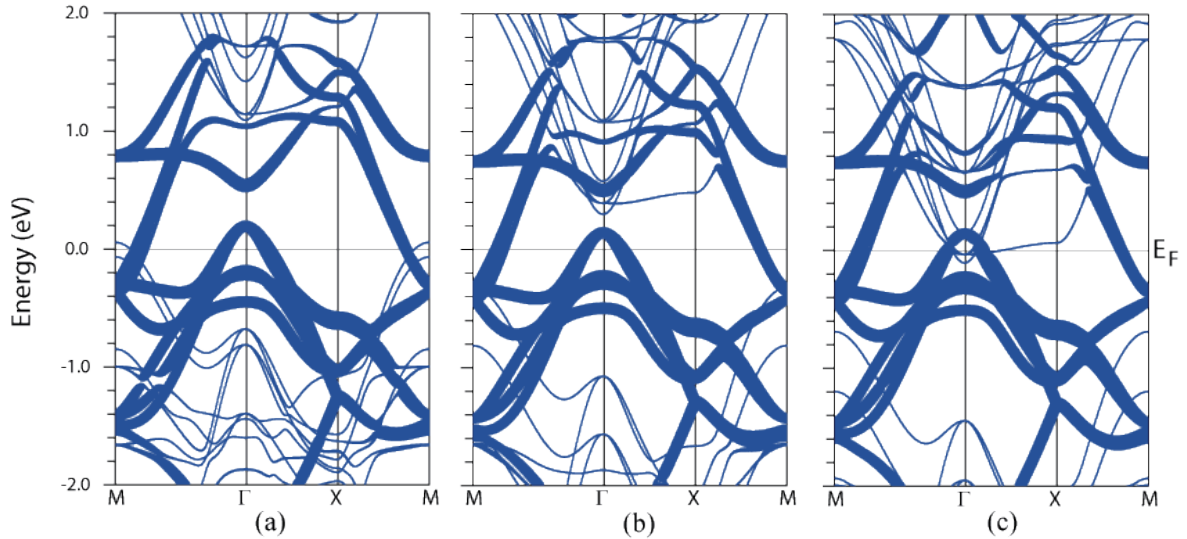


FIG. 2. (Color online) Fat bands of Fe 3d orbital for VCA nuclear charge equal to 22.00, 22.05, 22.10 from left to right, respectively.

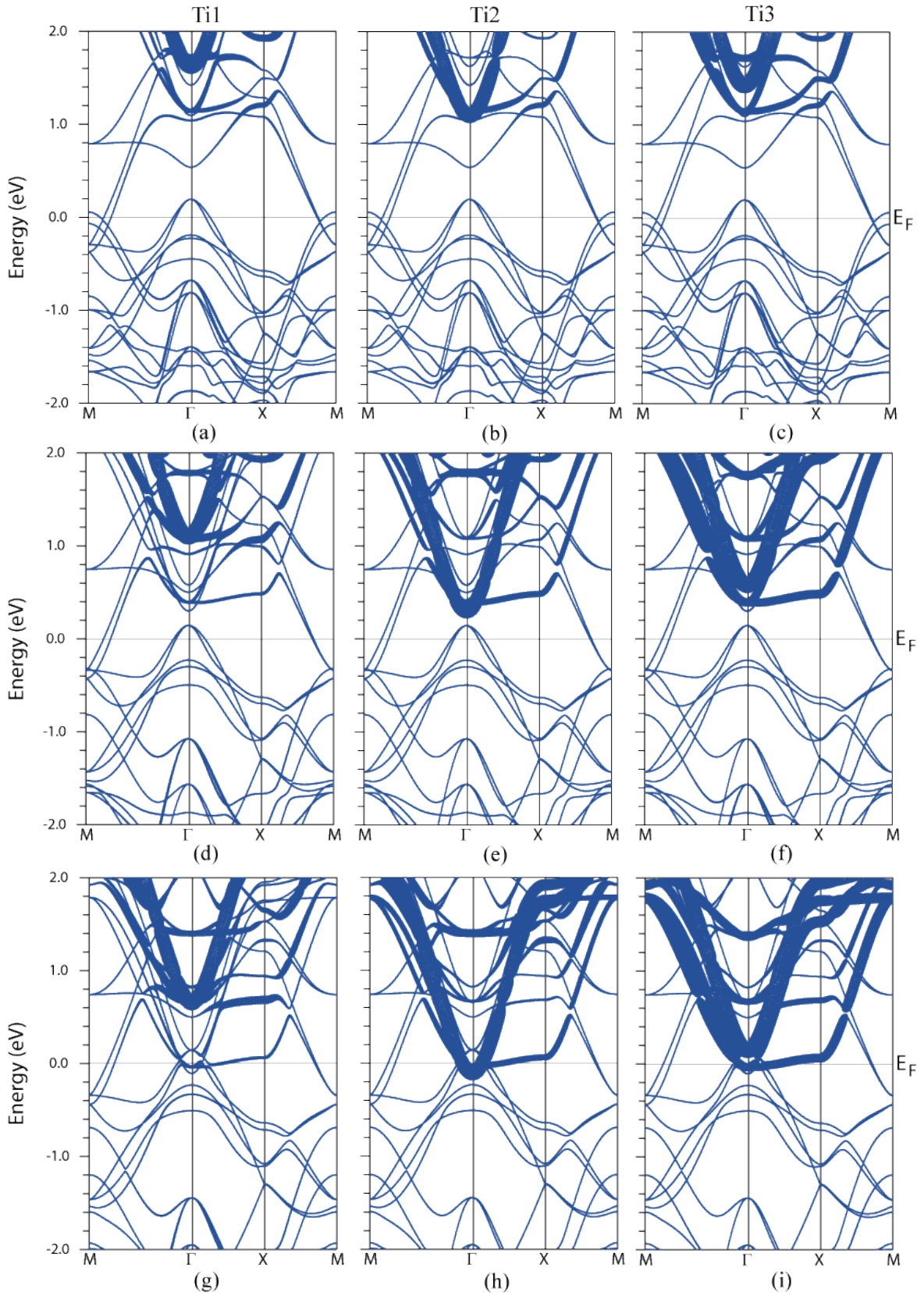


FIG. 3. (Color online) Fat bands of Ti 3d orbital for VCA nuclear charge equal to 22.00, 22.05, 22.10 from top to bottom, respectively.

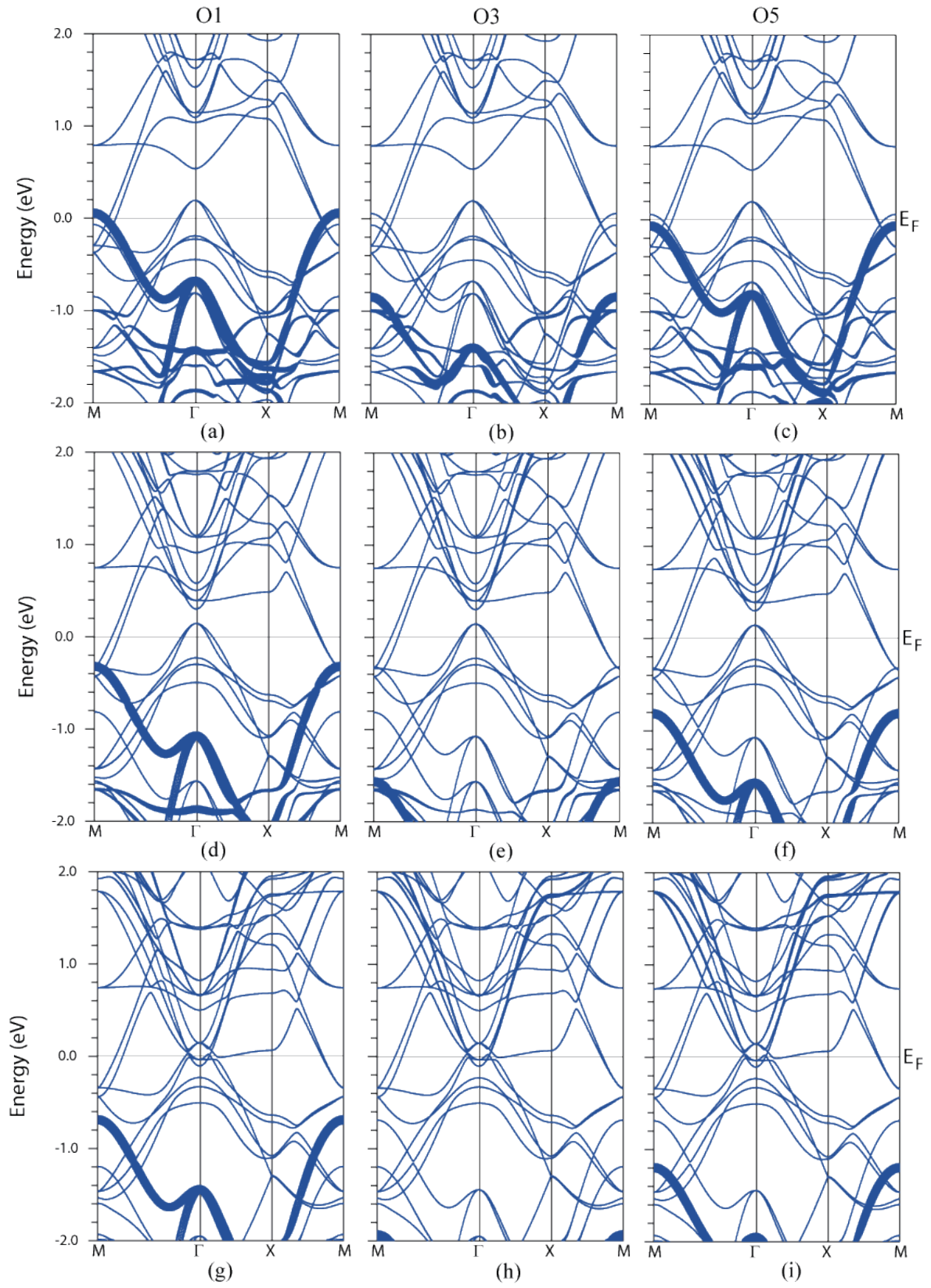


FIG. 4. (Color online) Fat bands of O 2p orbital for VCA nuclear charge from top to bottom, respectively.

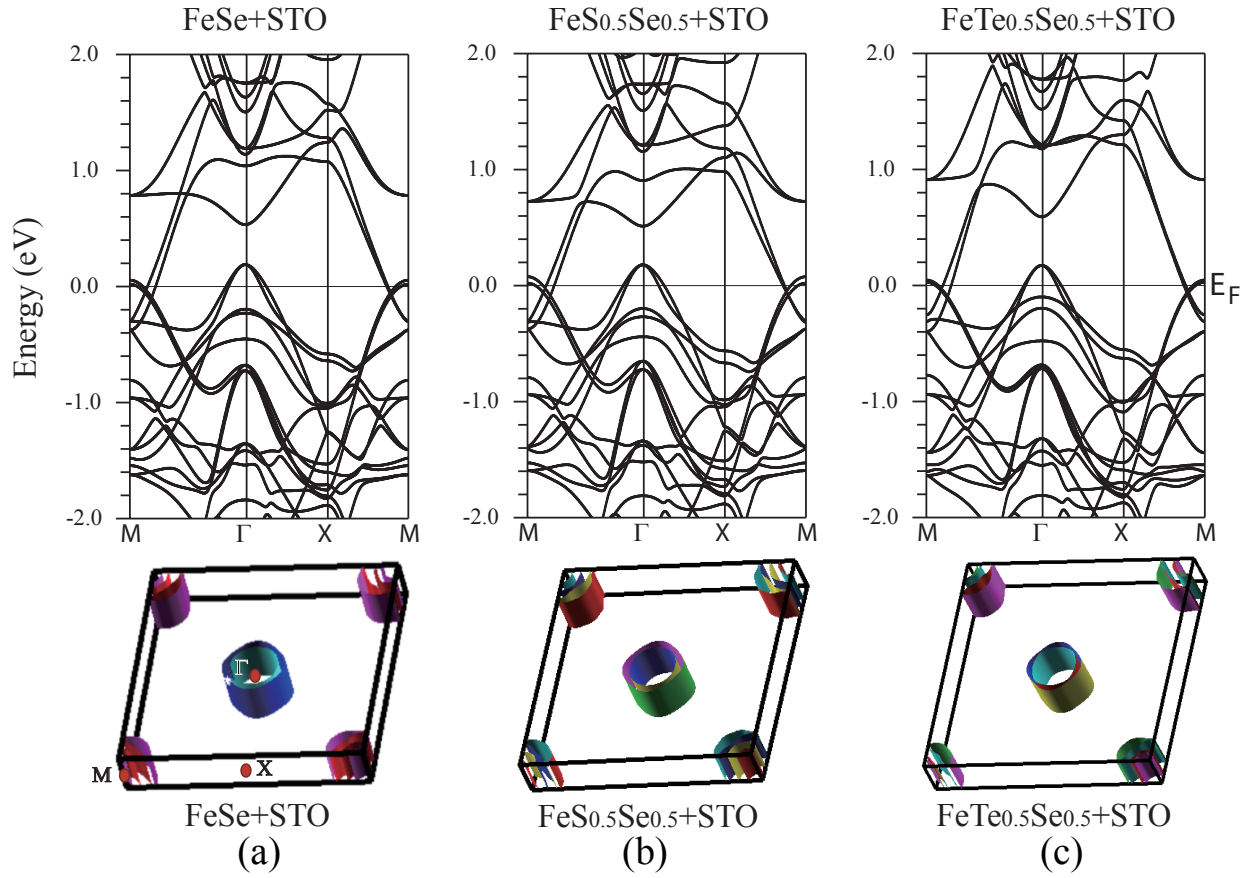


FIG. 5. (Color online) Band structures and Fermi surfaces of FeSe+STO, FeS_{0.5}Se_{0.5}+STO, and FeTe_{0.5}Se_{0.5}+STO, respectively.

TABLE II. The orbital occupation differences (in 10^{-3} a.u.) of Fe orbitals quantified by subtracting corresponding orbital occupations.

FeSe+STO minus kinds of structures	d_{z^2}	$d_{x^2-y^2}$	d_{xy}	d_{xz}	d_{yz}	<i>net change</i>
1.FeSe and STO	+11.4	+3.9	+3.6	-24.6	+10.0	+4.3
7.FeSe+STO _{0.8} N _{0.2}	-5.8	+4.1	+0.4	-7.3	+10.8	+2.2
8.FeSe+STO _{0.2} N _{0.8}	+1.2	+1.4	+0.6	-6.7	+4.1	+0.6
9.FeSe+STN	+3.1	+0.9	+1.5	-9.8	+5.5	+1.2

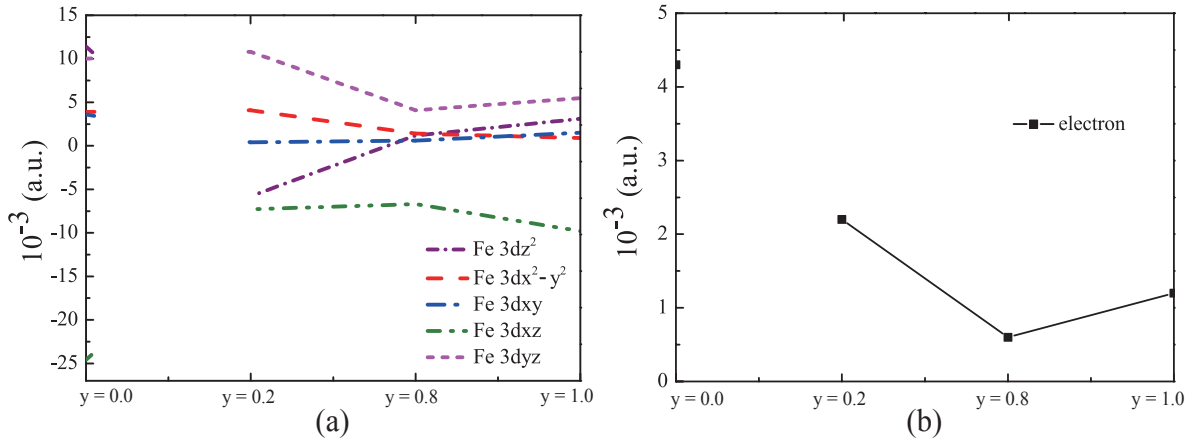


FIG. 6. (Color online) Orbital occupation difference of $\text{FeSe}+\text{STN}_y\text{O}_{1-y}$, which in units of 10^{-3} for (a) Fe five d orbitals occupations and (b) the net change of charge in all Fe d orbitals. All the data corresponding to 1, 7~9 kinds of structures as shown in Table II.

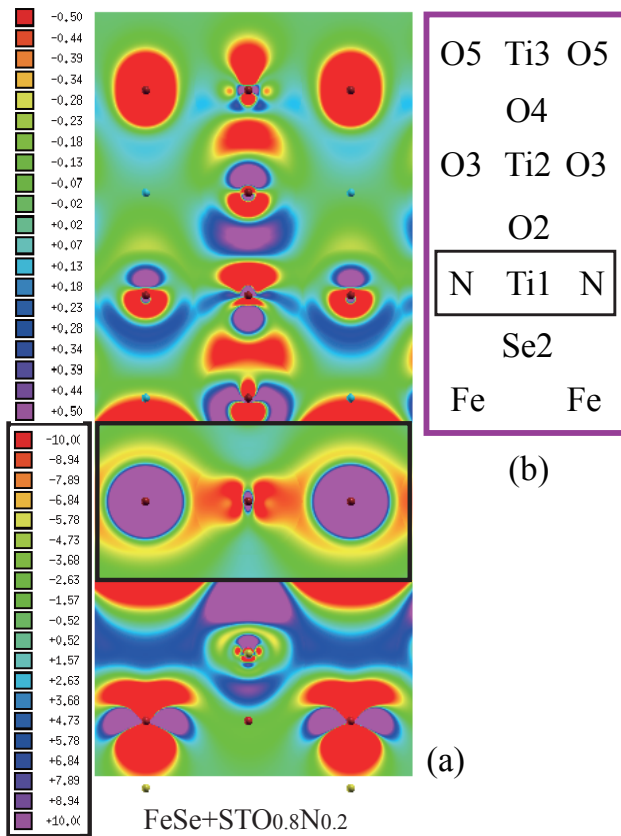


FIG. 7. (Color online) Difference density $\rho(\text{FeSe}+\text{STO})$ minus $\rho(\text{FeSe}+\text{STO}_{0.8}\text{N}_{0.2})$. The color scale is changed in the N region (between the black bars) where the bottom scale applies.