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TITLE:

Probe Type II Band Alignment in One-Dimensional Van Der Waals Heterostructures Using First-Principles Calculations

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SUMMARY:

Calculations performed by the Vienna Ab initio Simulation Package can be used to identify the intrinsic electronic properties of nanoscale materials and predict the potential water-splitting photocatalysts.

ABSTRACT:

Computational tools based on density-functional theory (DFT) enable the exploration of the qualitatively new, experimentally attainable nanoscale compounds for a targeted application. Theoretical simulations provide a profound understanding of the intrinsic electronic properties of functional materials. The goal of this protocol is to search for photocatalyst candidates by computational dissection. Photocatalytic applications require suitable band gaps, appropriate band edge positions relative to the redox potentials. Hybrid functionals can provide accurate values of these properties but are computationally expensive, whereas the results at the Perdew-Burke-Ernzerhof (PBE) functional level could be effective for suggesting strategies for band structure engineering via electric field and tensile strain aiming to enhance the photocatalytic performance. To illustrate this, in the present manuscript, the DFT based simulation tool VASP is used to investigate the band alignment of nanocomposites in combinations of nanotubes and nanoribbons in the ground state. To address the lifetime of photogenerated holes and electrons in the excited state, nonadiabatic dynamics calculations are needed.

INTRODUCTION:

The worldwide demand for clean and sustainable energy has spurred research for promising

materials to reduce dependence on finite petroleum resources. Simulations are more efficient and economical than experiments in accelerating the search for new functional materials[1]. Material design from a theoretical perspective[2–4] is now more and more popular due to rapid advances in computational resources and theory developments, making computational simulations more reliable[5]. The density functional theory (DFT) calculations implemented in many codes are becoming more robust and yield reproducible results[6].

The Vienna Ab initio Simulation Package (VASP)[7] presents one of the most promising DFT codes for predicting molecular and crystalline properties and more than 40,000 studies making use of this code have been published. Most work is performed at the Perdew-Burke-Ernzerhof (PBE) functional level[8], which underestimates the band gap sizes, but captures the essential trends in band alignment and band offsets[3]. This protocol aims to outline the details of investigating the band edge profiles and bandgaps of nanoscale materials for clean and renewable energy using this computational tool. More examples using VASP are available at https://www.vasp.at.

This report presents the computational screening of one-dimensional (1D) vdW heterostructures with type II band alignments[9] for a promising application in photocatalytic water splitting[4]. Specifically, nanoribbons (NRs) encapsulated inside nanotubes (NTs) are examined as an example[10]. To address noncovalent interactions, vdW corrections using the DFT-D3 method are included[11]. The DFT calculations in steps 1.2, 2.2, 3.2, 3.5.2, and section 4 by VASP are performed using a Portable Batch System (PBS) script by the high-performance research computers in the CenTOS system. An example of a PBS script is shown in the **Supplementary Materials**. The data postprocessing by the P4VASP software in step 3.3 and the figure plot by the xmgrace software in step 3.4 are carried on a local computer (laptop or desktop) in the Ubuntu system.

PROTOCOL:

1. Optimize the atomic structure.

1.1. Prepare four input files for structure relaxation calculation by VASP: INCAR, POSCAR, POTCAR, and KPOINTS.

NOTE: There are specified parameters in the INCAR file that define the calculation. The line "EDIFFG= 0.02" in the INCAR file indicates that all atoms are relaxed until the force on each atom is <0.02 eV/Å. The POSCAR file contains the atomic geometry information. The initial lattice parameters in the POSCAR file can be chosen from theoretical[3] or experimental references[12,13]. The KPOINTS file defines the k point mesh and POTCAR is the pseudopotential file. The order of atom types in POSCAR should be the same as that in POTCAR. Examples of input files for structure relaxation are shown in the **Supplementary Materials** (except the pseudopotential file, which needs a license from VASP).

1.1.1. Generate the initial structure of boron nitride (BN) nanoribbons (NR) for "POSCAR".

1.1.1.1. Download the POSCAR file for the BN bulk unit from https://materialsproject.org.

1.1.1.2. Use v2xsf to convert the POSCAR file to a file in xsf format that can be read by xcrysden. Type **v2xsf POSCAR** on the terminal in the Ubuntu system to get "**POSCAR.xsf.gz**". Type **gunzip POSCAR.xsf.gz** and output the POSCAR.xsf file.

1.1.1.3. Use xcrysden to build the BN supercell.

1.1.1.3.1. Type **xcrysden --xsf POSCAR.xsf** on the terminal in the Ubuntu system. Select the menu **Modify/Number of Units Drawn** and extend the cell in the X and Y directions.

1.1.1.3.2. Select the menu **File/Save XSF Structure** to export the supercell structure, named "**supercell**".

NOTE: The name of the structure is an arbitrary definition.

1.1.1.4. Use xmakemol to open the supercell. Type **xmakemol -f supercell** on the terminal in the Ubuntu system. Select the menu **Edit/Visible**. Click **Toggle** to delete the atoms inside the region and cut the NR to the desired width and chirality.

1.1.2. Generate the initial structure of the BN nanotube (NT) for POSCAR. Download "**NanotubeModeler**" from http://www.jcrystal.com/products. Open NanotubeModeler.exe in the Windows system. Select the menu **Select type/B-N** and specify the chirality. Select the menu **File/Save XYZ table** to export the structure.

1.1.3. Generate the initial structure of the nanocomposite by encapsulating the NR (from step 1.1.1) inside the NT (from step 1.1.2).

NOTE: The encapsulation can be finished by adjusting the Cartesian coordinates of the NR and the NT[10,14,15].

1.1.4. Use the vmd software to check the atomic structure before submitting the calculation job.

1.1.4.1. Type **vmd** on the terminal in the Ubuntu system. In the opened vmd main window, select the menu **File/New Molecule** and find the POSCAR file through the **Browse** window. Load POSCAR by typing **VASP_POSCAR**.

1.1.4.2. Display the structure in different styles in the **Graphical Representations/Drawing Method** window.

NOTE: For example, once the CPK is chosen, each atom (bond) is represented by a sphere (stick). The installation guide and full tutorial of vmd are available at http://www.ks.uiuc.edu/Research/vmd.

1.2. Type **qsub job.pbs** on the terminal in the Linux system to submit the job to the computer

cluster.

NOTE: "**job.pbs**" represents the name of the PBS script. The name of the PBS script is an arbitrary definition. The four input files together with the PBS script should be at the working directory. The command **qsub job.pbs** will be used in steps 2.2, 3.2, 3.5.2, and section 4. An example of a PBS script can be found in the supplementary coding file. After the submitted job is finished, if "**reached required accuracy - stopping structural energy minimization**" appears at the end of the output log, the converged result is obtained. The resulting CONTCAR file will be used as the input file POSCAR in steps 2.1, 3.1, 3.5.1, 3.5.3.1, 4.1.1, 4.1.4, and section 4.2.

2. Calculate the encapsulation energy.

2.1. Type **mkdir nanocomposite isolated-nanoribbon isolated-nanotube** to create three folders for the nanocomposite, the NR, and the NT on a terminal in a Linux system. Prepare one PBS script "**job.pbs**" and four input files INCAR, POSCAR, POTCAR, and KPOINTS for the energy calculation in each folder.

NOTE: The input file POSCAR is the file named CONTCAR with the relaxed structure from step 1. Examples of input files are given in **Supplementary Materials** (except POTCAR).

2.2. Go to each folder and type **qsub job.pbs** on the terminal in the Linux system.

NOTE: The three submitted jobs will perform the static self-consistent energy calculations for the nanocomposite, isolated NR, and isolated NT, respectively.

2.3. Extract the total energy from the file OUTCAR for each system after finishing the static self-consistent calculations. Type **grep "free energy TOTEN" ./nanocomposite/OUTCAR | tail -n 1**, **grep "free energy TOTEN" ./isolated-nanoribbon/OUTCAR | tail -n 1**, and **grep "free energy TOTEN" ./isolated-nanotube/OUTCAR | tail -n 1**. Define the three displayed values as E_{NT+NR}, E_{NR}, and E_{NT}, respectively. Calculate the encapsulation energy per angstrom: $E_L = (E_{NT+NR} - E_{NT} - E_{NR})/L$[14,15].

NOTE: The periodical direction in each system is along the Z axis and L is the lattice constant of the unit cell along the Z axis. Test calculations of the energy dependence on the plane wave cutoff energy and the k point mesh are needed. The encapsulation energy can be used as an estimate for the energetic stability of the nanocomposite.

3. Extract the electronic properties from the band structure.

3.1. Prepare one PBS script "**job.pbs**" and six input files: INCAR, POSCAR, POTCAR, KPOINTS, CHGCAR, and CHG for band calculation. Set **ICHARG = 11** in INCAR.

NOTE: The preconverged CHGCAR and CHG files are from the static self-consistent calculations in step 2.2. The band analysis is at the PBE level. The k point sampling in the KPOINTS file is in

line-mode. Examples of input files for this step can be found in the **Supplementary Materials** (except POTCAR).

3.2. Type **qsub job.pbs** on the terminal in the Linux system to submit the job.

3.3. Use P4VASP to generate the projected band.

3.3.1. Load "**vasprun.xml**" by typing **p4v vasprun.xml** on terminal in the Ubuntu system.

NOTE: "p4v" is used to start P4VASP. The file "vasprun.xml" should be at the working directory.

3.3.2. Select the menu **Electronic/Local DOS+bands control** and then **Select/Bands**.

3.3.2.1. Specify the atomic numbers of NT in the section **Atom selection**. Get the atomic number by pointing to the corresponding atoms using vmd as mentioned in step 1.1.4. Specify the color, type, and size of the symbol for the projected band structure through the menu **Symbol** and **Symbol size**. Press the menu **Add new line**.

NOTE: The graph will show the band structure with contributions from the NT.

3.3.2.2. Repeat the same procedure following step 3.3.2.1 to get the projected band with contributions from the NR.

3.3.3. Select the menu **Graph/Export**. Export the graph into a file with an agr format (for example, as "11-4.agr").

NOTE: The output data of the projected bands by P4VASP are in three columns where the third one represents the weighting.

3.4. Use xmgrace to edit the projected band.

3.4.1. Type **xmgrace 11-4.agr** on the terminal to start xmgrace in the Ubuntu system. Select the menu **Plot/Axis properties** to edit the label and range of the axis.

3.4.2. Select the menu **Plot/Set appearance** to read the energy value at the specified band number and k point.

NOTE: The valence band maximum (VBM) and conduction band minimum (CBM) of NR/NT can be read from the projected band with contributions on NR/NT, respectively. According to the band alignments, heterostructures can be classified into three types: type I ($VBM_{NT} < VBM_{NR} < CBM_{NR} < CBM_{NT}$ or $VBM_{NR} < VBM_{NT} < CBM_{NT} < CBM_{NR}$), type II ($VBM_{NT} < VBM_{NR} < CBM_{NT} < CBM_{NR}$ or $VBM_{NR} < VBM_{NT} < CBM_{NR} < CBM_{NT}$), or type III ($VBM_{NT} < VBM_{NT} < CBM_{NR} < CBM_{NR}$ or $VBM_{NR} < VBM_{NR} < CBM_{NT} < CBM_{NT}$)[9].

3.4.3. Calculate the valence band offset (VBO), conduction band offset (CBO), and the band gap following Kang et al.[16].

3.4.4. Select the menu **File/Print** to export the graph with eps format.

3.5. Calculate the band decomposed charge density for the VBM and CBM.

3.5.1. Prepare one PBS script "**job.pbs**" and seven input files: INCAR, POSCAR, POTCAR, KPOINTS, WAVECAR, CHGCAR, and CHG. Specify the band numbers for the CBM and VBM by the tag IBAND in INCAR. Use the single corresponding k point for each band edge.

NOTE: The preconverged CHGCAR, CHG, and WAVECAR files are from the static self-consistent calculations in step 2.2. Examples of input files for this step are given in the **Supplementary Materials** (except POTCAR).

3.5.2. Type **qsub job.pbs** on the terminal in the Linux system to submit the job.

3.5.3. Use vmd to plot the VBM and CBM in real space after the job is finished.

3.5.3.1. Start a vmd session and load the POSCAR file as in step 1.1.4.

3.5.3.2. Select the menu **File/New Molecule** in the vmd main window. Find the PARCHG file through the **Browse** window. Load PARCHG by typing **VASP_PARCHG**.

3.5.3.3. Select the menus **Draw/Solid Surface** and **Show/Isosurface** in the **Graphical Representations** window. Change the isovalue to an appropriate value (for example, 0.02). Change the color of the isosurface through the menu **Coloring Method**.

NOTE: This is an intuitive analysis for band types with respect to that in step 3.4. Generally, the atomic structure is arranged away from the boundary, otherwise the visualized charge density is not shown in a continuous manner. Please see **Supplemental Figure 1** for details.

4. Modulate the electronic properties of the nanocomposite (NT encapsulated inside NR) by external fields.

4.1. Add a transverse electric field to the nanocomposite[17].

4.1.1. Prepare one PBS script "**job.pbs**" and four input files: INCAR, POSCAR, POTCAR, and KPOINTS.

4.1.2. Define the strength of the electric field by the tag "**EFIELD**" in units of eV/Å.

4.1.3. Set LDIPOL = T. Specify IDIPOL with an exact value (1, 2, or 3).

NOTE: These two tags are added to include dipole corrections. The electric field will be applied along the X, Y, or Z axis by setting the value of IDIPOL to 1, 2, or 3.

4.1.4. Perform the static self-consistent calculations and band structure calculations following sections 2 and 3 without structural optimization.

NOTE: Previous studies indicate that electric fields over 5 V/Å can be used to modify the band gap of BN-NT and BN-NR without deforming the structure[18,19].

4.2. Add a longitudinal tensile strain to the nanocomposite.

4.2.1. Change the lattice parameters along the periodical direction to reflect the strain effect.

NOTE: For example, the optimized lattice parameter of the nanocomposite along the Z axis is 2.5045 Å. If 1% uniaxial tensile strain is applied along the Z direction, change the lattice parameter in POSCAR to 2.5045 x 1.01 = 2.529545 Å.

4.2.2. Relax the modified structure following section 1.

4.2.3. Perform static self-consistent calculations and band structure calculations following sections 2 and 3.

REPRESENTATIVE RESULTS:

Zigzag BN-NRs encapsulated inside armchair BN-NTs (11,11) were chosen as representative examples for a 1D vdW heterostructure. The lattice parameters were taken from Sahin et al.[20]. For convenience, zigzag NRs are abbreviated Z_n, where n represents the III–V dimers along the width[14]. The encapsulation energy E_L from step 2.3 was used as a rough estimate for the energetic stability of the nanocomposite. The E_L values of Z_2, Z_3, and Z_4 encapsulated inside BN-NT (11,11) were -0.033 eV/Å, -0.068 eV/Å, and -0.131 eV/Å, respectively[10], as shown in **Figure 1**. Although E_L varied by an order of magnitude with BN-NR size, all three nanocomposites presented type II band structures (from step 3.4) superior to the all-carbon cases[14], where type II only emerged for NR with only one appropriate size inserted in NT[14].

The band structure of the nanocomposite from step 3.2, BN-NT (11,11)+Z_4, is shown in **Figure 2**. VBM/CBM locates at NT/NR (from step 3.5), respectively. The staggered band alignment was beneficial for light harvesting. The main mechanism of charge transfer is as follows: the photo generates electrons and a hole in Z_4 at the X point, shown in Figure 3, and then the hole dissociates from Z_4 (k_X) to NT (11,11) (k_{VBM}, the k point of VBM for this nanocomposite), shown in **Figure 4**. The calculated VBO (from step 3.4.3) is 317 meV, larger than the thermal energy at 300 K (KT ~30 meV), and effectively decreases the recombination rate of the photogenerated carriers[10].

To enhance light harvesting through a wide spectrum, both transverse electric fields and longitudinal tensile strains are applied to BN-NT (11,11) + Z_4. The evolution of band edges relative

to the vacuum level from step 4 is shown in **Figure 5**. A substantial gap reduction up to near 0.95 eV is observed in this nanocomposite by external fields. More importantly, the staggered band alignment is preserved[10]. Based on these results, such a 1D system is expected to integrate photocatalytic hydrogen generation and safe capsule storage[21]. The photogenerated electrons could be collected by NR. Driven by electrostatic attraction, protons penetrate through the NT to generate a hydrogen molecule. The produced hydrogen is completely isolated within the nanotube to avoid an unwanted reverse reaction or explosion.

FIGURE LEGENDS:

Figure 1: Zigzag BN nanoribbons Z_2, Z_3, and Z_4 encapsulated inside a BN nanotube (11,11). The encapsulation energy (E_L) is listed under each structure.

Figure 2: Band structure of BN nanotube (11,11) + BN nanoribbon Z_4. The contributions from the nanotube and nanoribbon to the energy bands are represented in red and blue spheres, respectively. The left insets show the charge density distributions of the CBM and the VBM states (isovalue 0.02 e/Å^3). This figure was adapted from Gong et al.[10] with permission from The Royal Society of Chemistry.

Figure 3: The photo generates electrons and a hole in the BN nanoribbon Z_4 at the X point.

Figure 4: The hole dissociates from the BN nanoribbon Z_4 (k_X) to the BN nanotube (11,11) (k_{VBM}, the k point of VBM for this nanocomposite).

Figure 5: Band edge modulation of the BN nanotube (11,11) and the BN nanoribbon Z_4 by external fields. Evolution of band edges relative to the vacuum level under (**A**) an electric field and (**B**) uniaxial tensile strain. The negative direction of electric field is denoted from the lower edge atom B to the upper edge atom N of Z_4. The reduction potential of H^+/H_2 and the oxidation potential of O_2/H_2O are -4.44 eV and -5.67 eV at pH = 0, respectively. The pH = 7 shifts the water's redox potentials (by pH x 0.059 eV) to -4.027 eV and -5.257 eV, respectively, shown as blue dashed lines. This figure was reproduced from Gong et al.[10] with permission from The Royal Society of Chemistry.

Supplemental Figure 1: (**A**) Atomic structure of a BN nanotube (11,11) + BN nanoribbon Z_4 arranged away from the boundary and its corresponding conduction band minimum (**B**). (**C**) Atomic structure of a BN nanotube (11,11) and BN nanoribbon Z_4 aligned with one boundary and its corresponding conduction band minimum (D).

DISCUSSION:

The calculations for electronic properties in sections 2, 3, and 4 would be similar among various nanoscale materials. The initial atomic model in step 1 should be carefully designed to extract meaningful information. For example, the factor for selecting the model could be the size or chirality of the materials. Also, the initial atomic model in step 1.1 should be reasonably prepared for low-cost structure relaxation. Taking the nanocomposite in the protocol as an example, the NR should be encapsulated inside the NT in a symmetrical manner. Otherwise, it will be time

consuming to search the optimized structure by VASP.

To consider the effect of an electric field, an artificial dipole sheet is added in the middle of the vacuum part in the periodic unit cell in VASP[22]. The vacuum region should not be too broad and the electric field should be weak enough to avoid artificial field emission[23].

Whereas the effect of the strain can be simply realized by changing the lattice parameter in POSCAR, in the nanocomposite the situation would be more complex. The elastic responses of the NR and NT may be different from each other, undergoing the same strength. This will lead to a disproportionate structure. For example, when the uniaxial tensile strain is applied along the periodical direction, the optimized lattice parameter of the NT and NR along this direction changes from an initial 1.8 Å to 2.0 Å, and 2.2 Å, respectively. Large supercells are required for modeling: at least 11 unit cells of NT and 10 unit cells of NR in this case (11 x 2.0 Å = 10 x 2.2 Å = 22 Å).

While ground state electronic properties of materials can be determined by VASP quite well, to address the lifetime of photogenerated holes and electrons existing in an excited state, it is better to perform nonadiabatic dynamics calculation[24]. This is important to design photocatalysts with long lifetime carriers[4].

The role of the computational approach performed by VASP plays into the discovery of novel materials and the screening for potential photocatalysts to assist experimental efforts. The band alignment at the PBE level in water splitting is not as convincing as quantitative experimental work. More accurate values of the band edges relative to the redox potentials, CBO, and VBO are needed. It would be best to use the Heyd-Scuseria-Ernzerhof (HSE) hybrid functional[25], but it is more time consuming than PBE. Nevertheless, the results at the PBE level could be efficient for suggesting strategies for the enhancement of photocatalytic activity.

It should be mentioned that the computational design by VASP will also enable the prediction of solar cell materials, thermoelectric materials, lithium battery materials, gas capture materials, etc.[2]. High-throughput calculations have been combined with the machine learning procedures for better materials prediction and lower computational cost[26,27].

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DISCLOSURES:

The authors have nothing to disclose.

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Figure 1 (eps)

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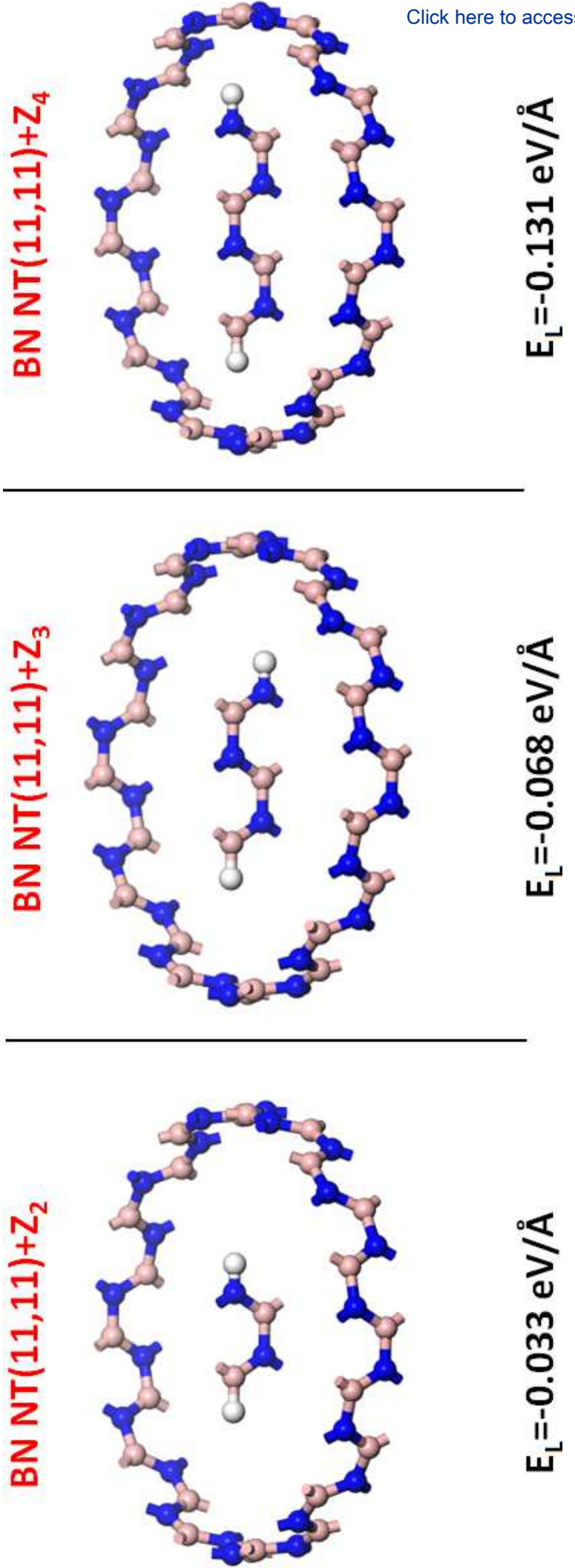

Figure 1 (psd)

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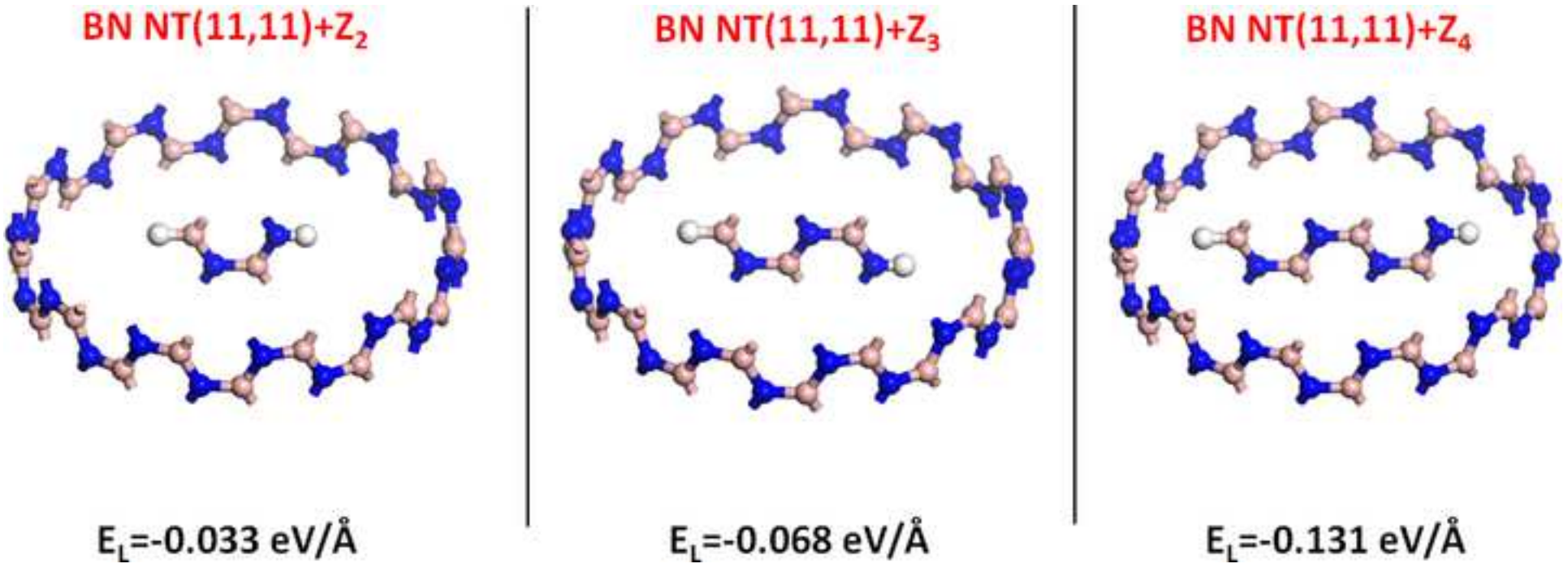

Figure 2 (eps)

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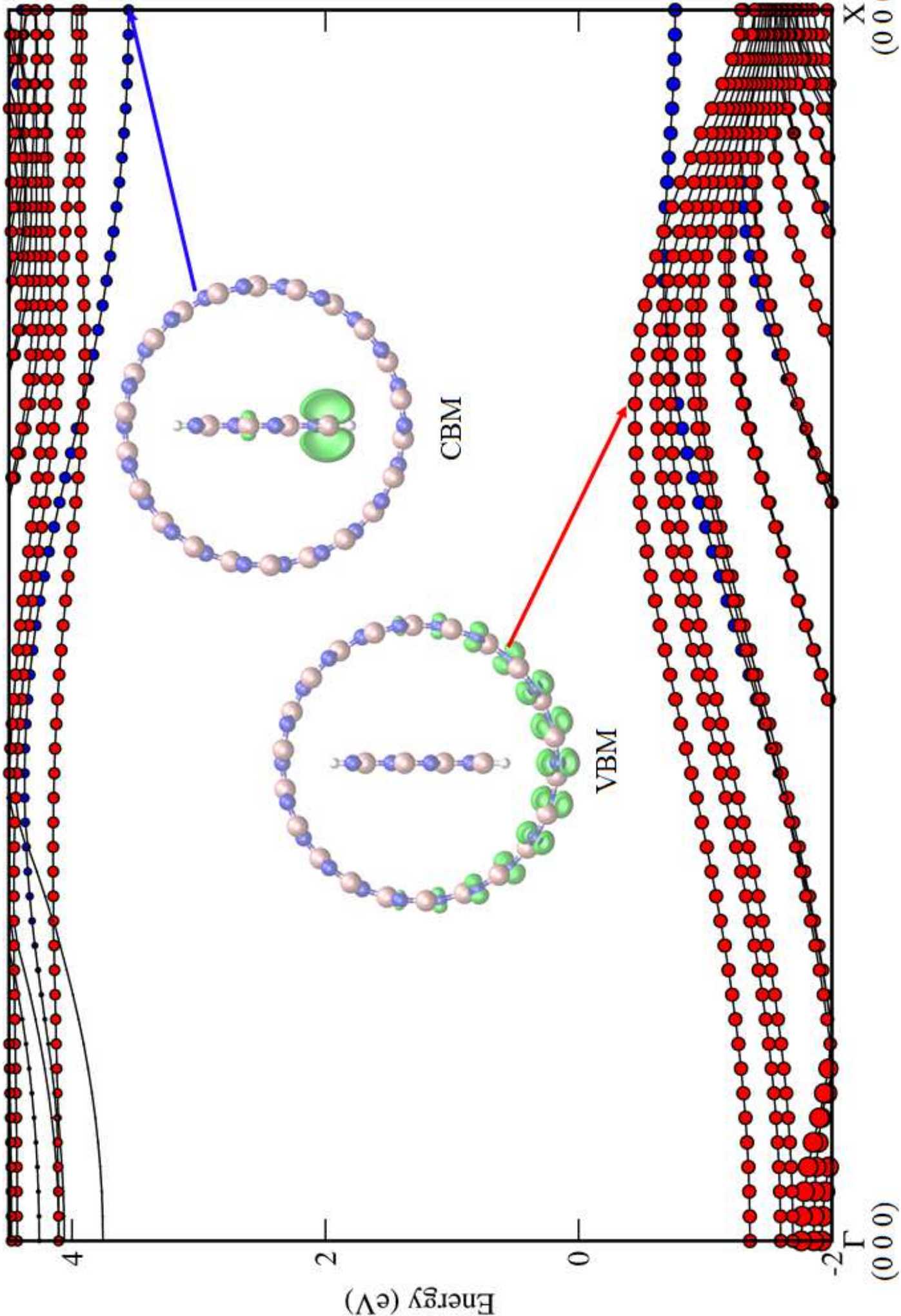

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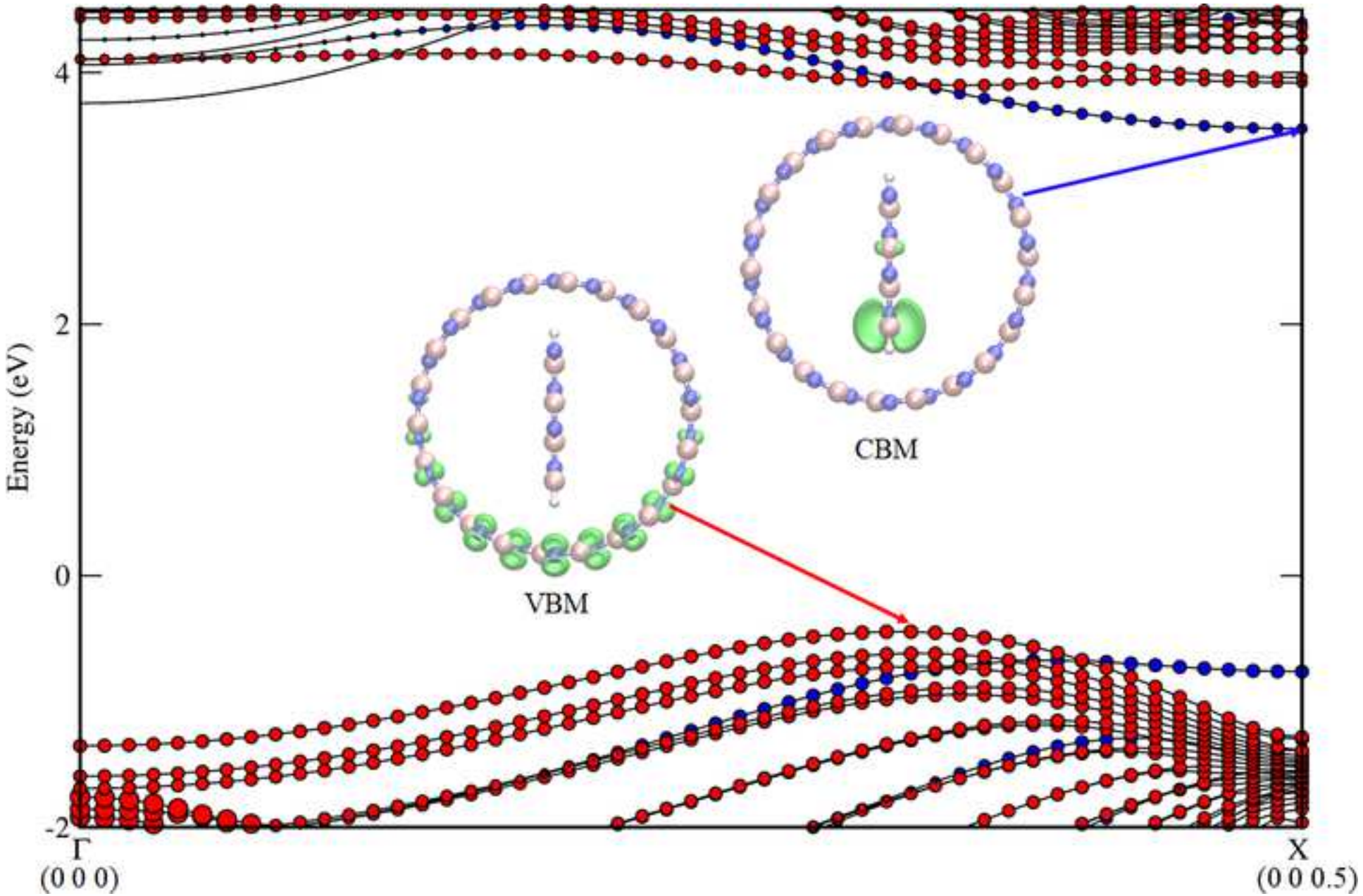

Figure 3 (eps)

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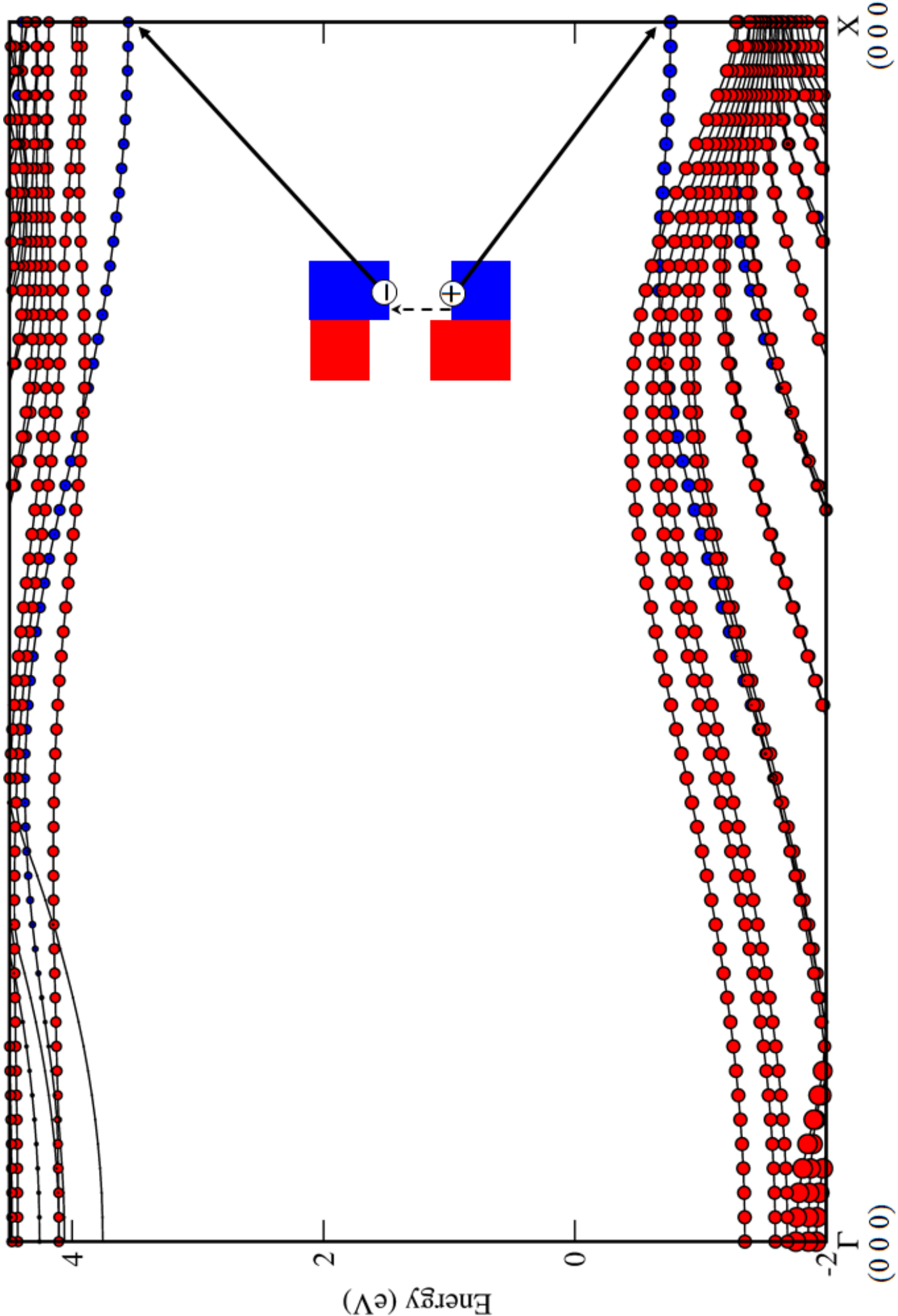

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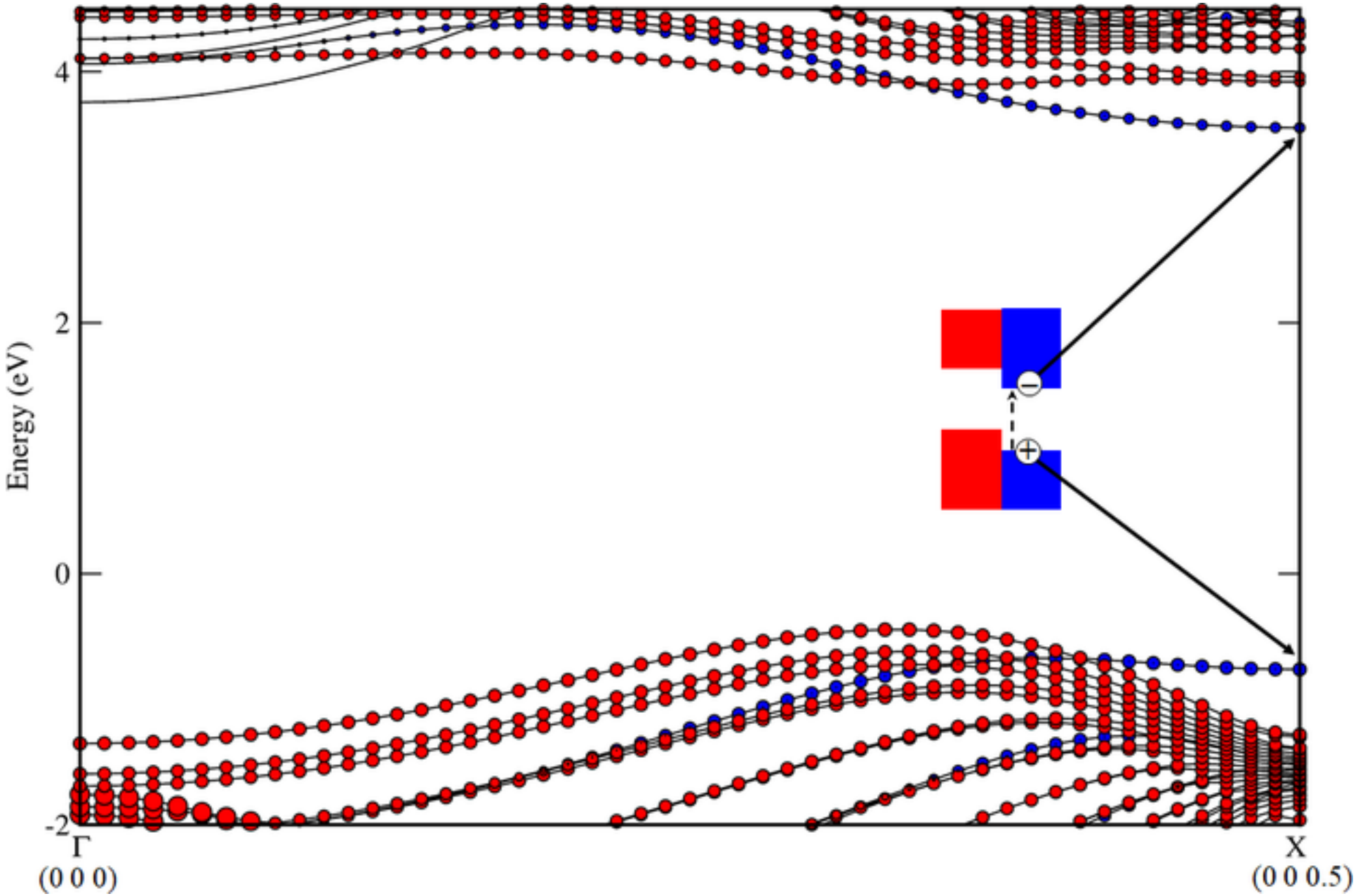

Figure 4 (eps)

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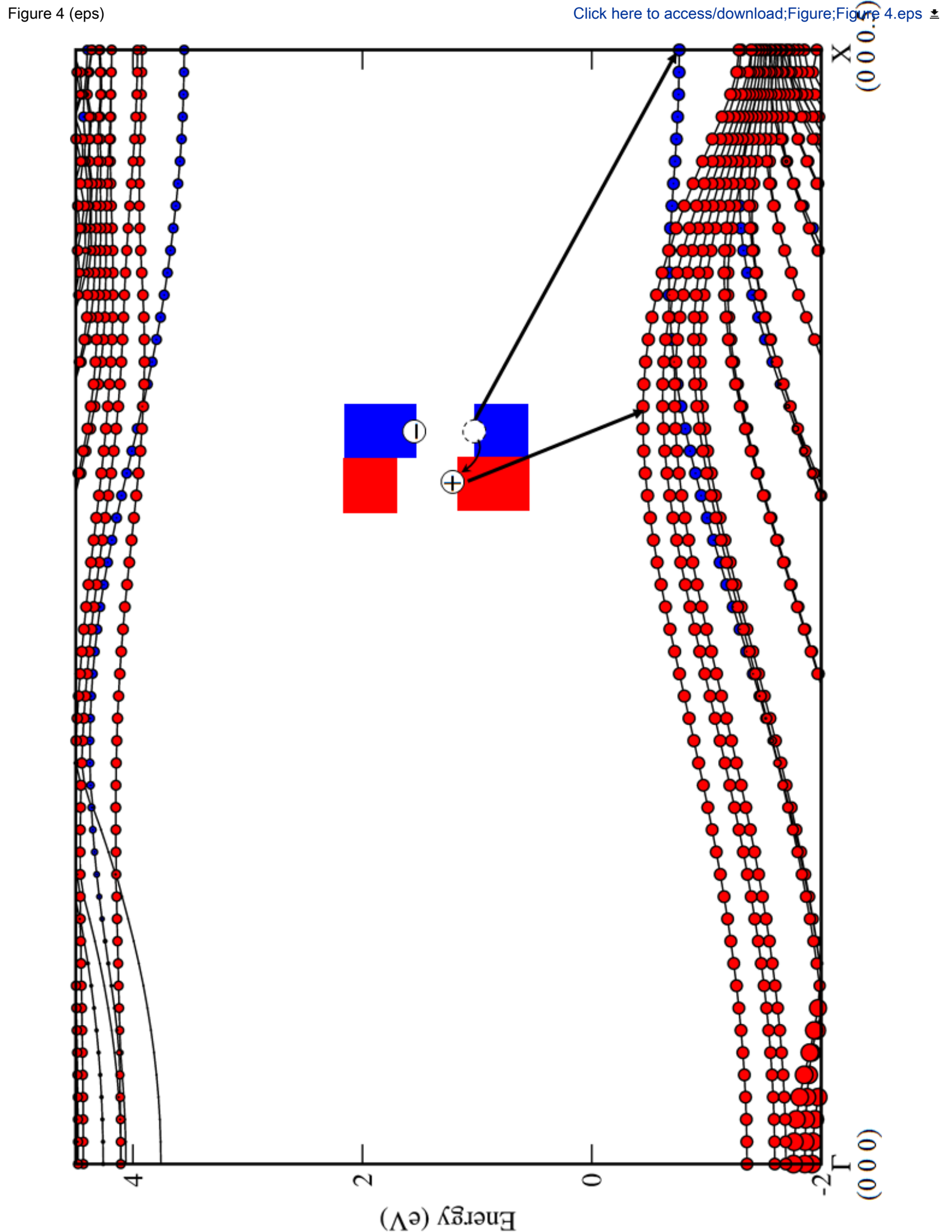

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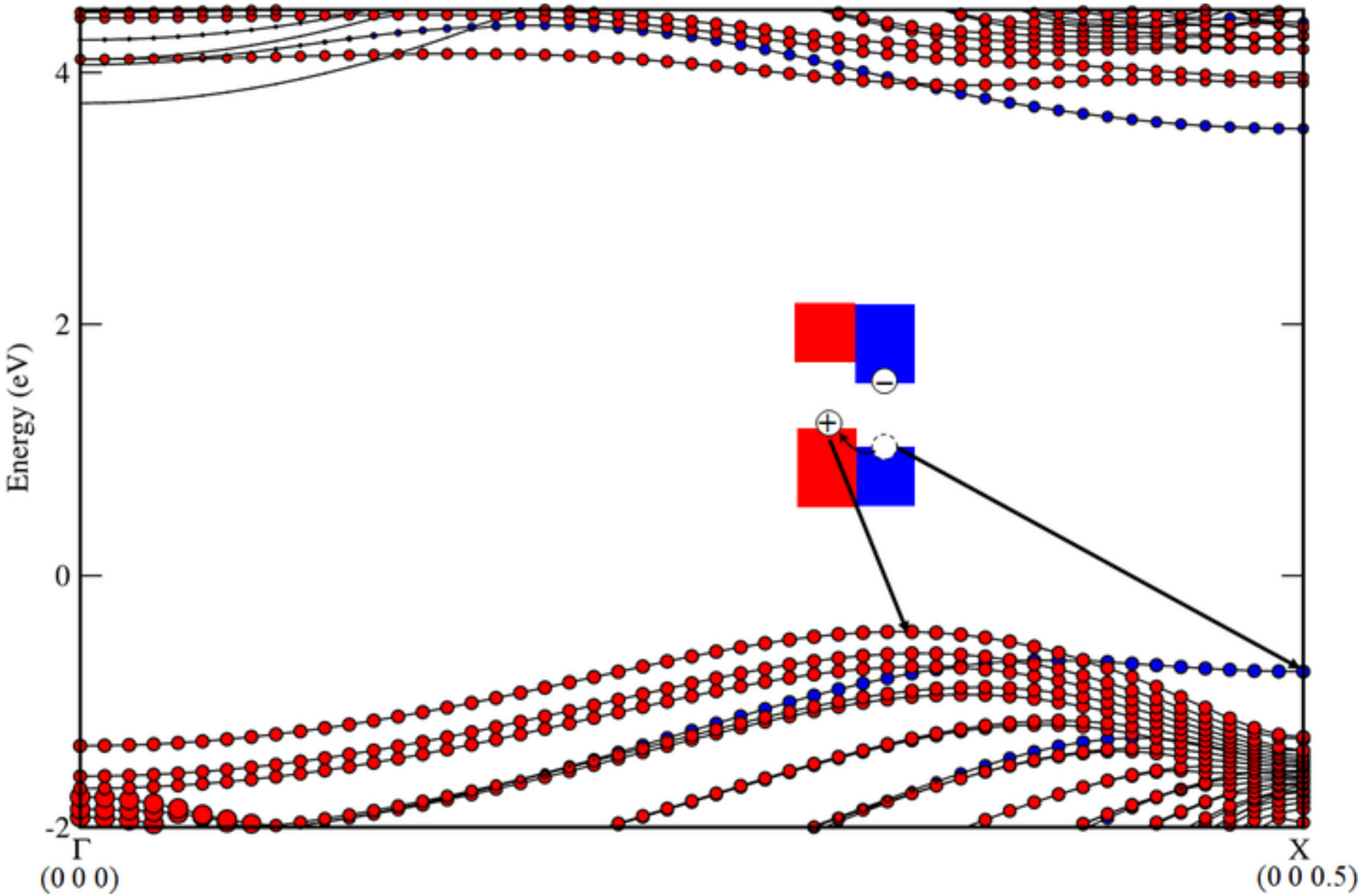

Figure 5 (eps)

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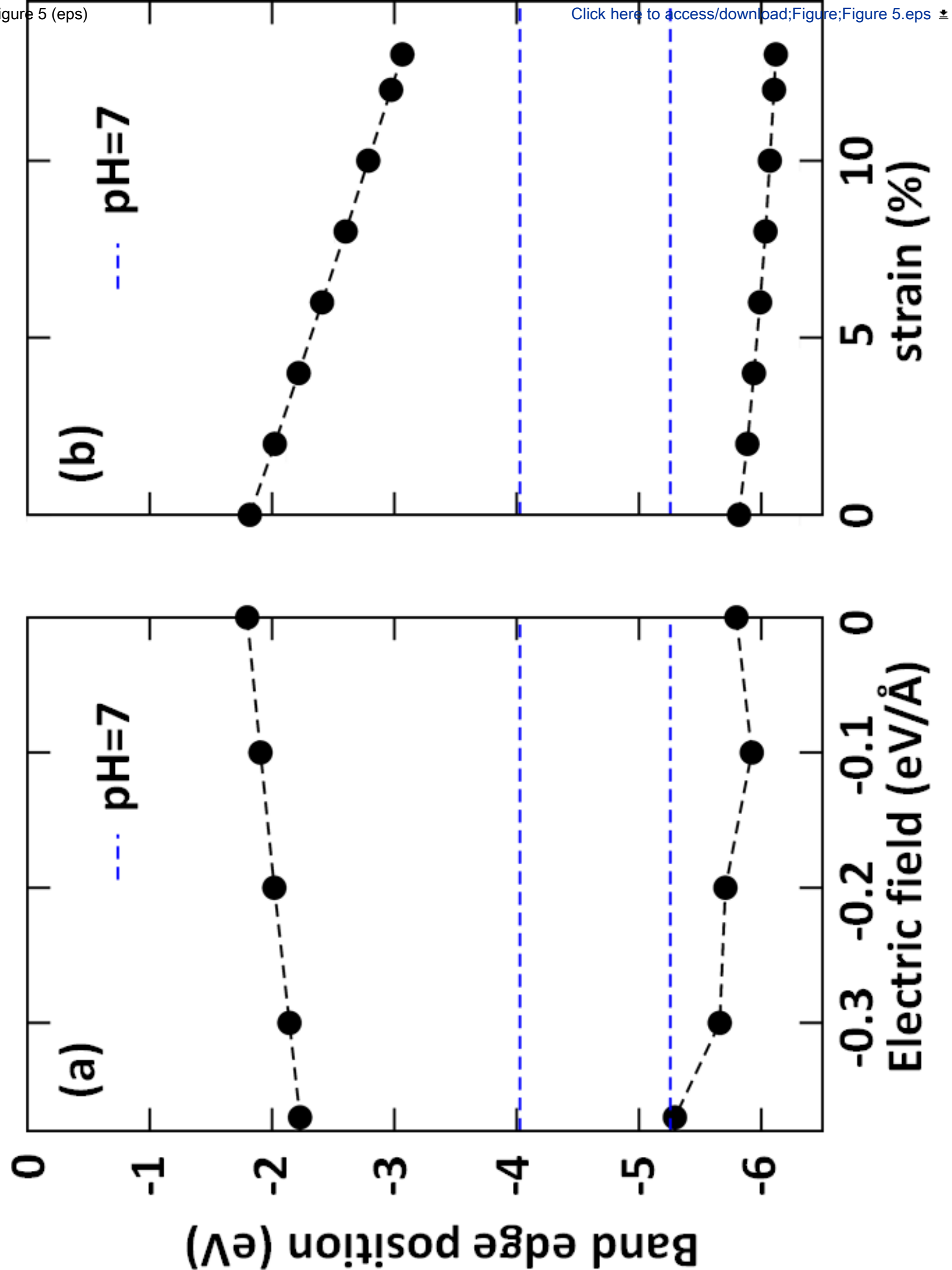

Figure 5 (psd)

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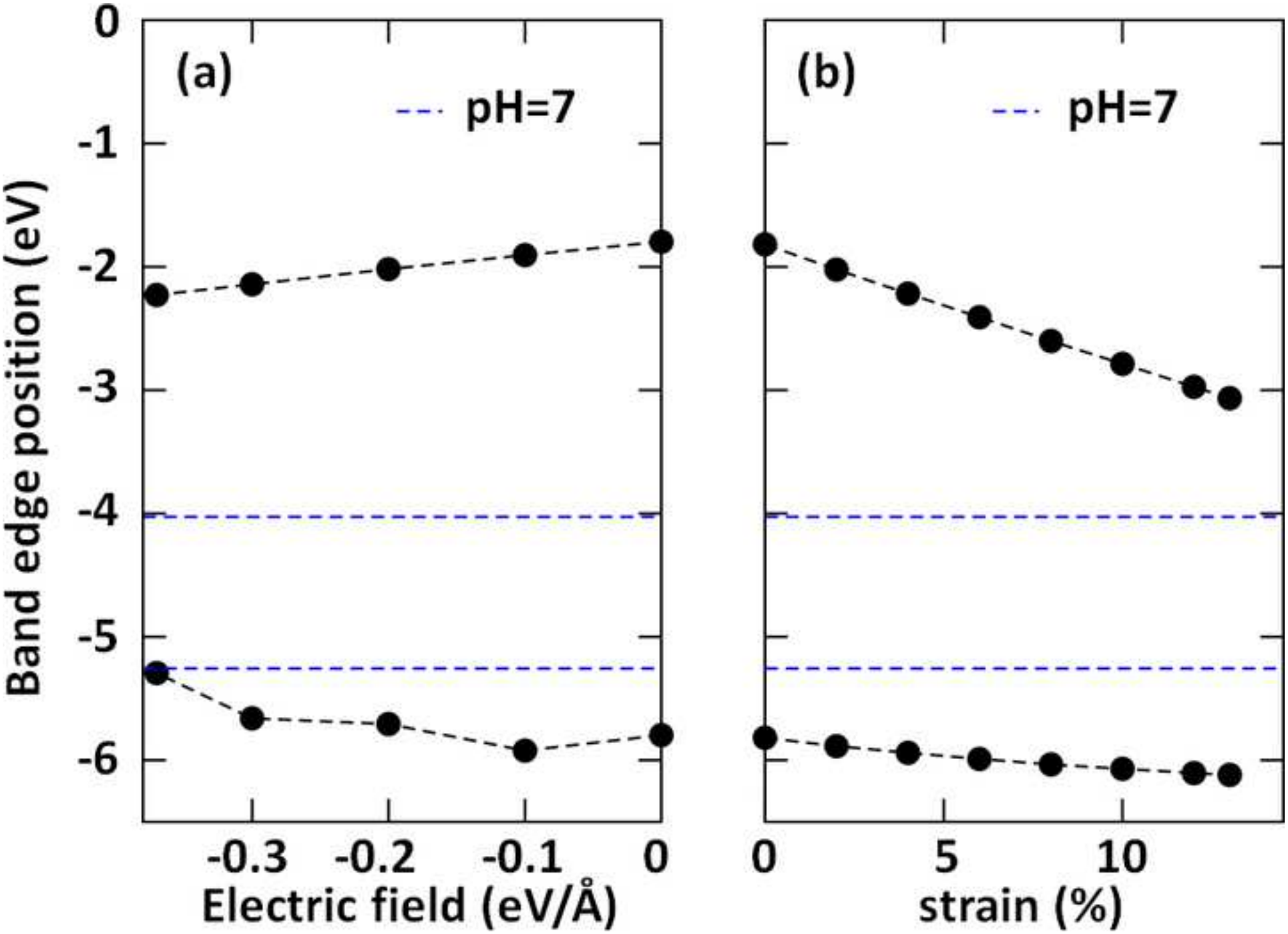

Name of Material/Equipment	Company	Catalog Number	Comments/Description
Nanotube Modeler	Developed by Dr. Steffen Weber	NanotubeModeler1.8	http://www.jcrystal.com/products/wincnt/NanotubeModeler.exe
P4VASP	Orest Dubay	p4vasp 0.3.30	Open source, available at www.p4vasp.at
v2xsf	Developed by Dr. Jens Kunstmann	v2xsf	http://theory.chm.tu-dresden.de/~jk/software.html
VASP software	Computational Materials Physics, Dept. of Physics, University of Vienna	vasp.5.4.1	https://www.vasp.at
VMD software	Theoretical and Computational Biophysics Group, University of Illinois at Urbana-Champaign	vmd1.9.3	https://www.ks.uiuc.edu/Research/vmd
xcrysden	Dept. of Physical and Organic Chemistry, Jozef Stefan Institute	XCrySDen1.5.60	http://www.xcrysden.org/
Xmakemol	Developed by M. P. Hodges	xmakemol5.16	https://www.nongnu.org/xmakemol/XmakemolDownloads.html
Xmgrace software	Grace Development Team under the coordination of Evgeny Stambulchik	xmgrace5.1.25	http://plasma-gate.weizmann.ac.il/Grace/

Dear Editor,

We would like to thanks for all the valuable and constructive comments. We have revised the video and manuscript in accordance with the comments. We have tracked the changes to identify all of the manuscript edits with blue color. Kindly find below our pointwise response to each comment.

Editorial comments:

1. *Please note that the editor has formatted the manuscript to match the journal's style. Please retain the same. The updated manuscript is attached and please use this version to incorporate the changes that are requested.*

Answer: Thanks for the reminding. We have updated the manuscript based on this version to incorporate the changes as requested.

2. *The Protocol should be made up almost entirely of discrete steps without large paragraphs of text between sections. Please simplify the Protocol so that individual steps contain only 2-3 actions per step and a maximum of 4 sentences per step. Use sub-steps as necessary.*

Answer: Thanks for this important comment. We have used more sub-steps and rephrased the large paragraphs to individual steps.

3. *Please revise the Protocol to contain only action items that direct the reader to do something (e.g., "Do this," "Ensure that," etc.). The actions should be described in the imperative tense in complete sentences wherever*

possible. Any text that cannot be written in the imperative tense may be added as a "NOTE."

Answer: Thanks for this important comment. We have described the actions in the imperative tense and moved the text not written in the imperative tense to the "NOTE".

4. *Please add more details to your protocol steps. There should be enough detail in each step to supplement the actions seen in the video so that viewers can easily replicate the protocol. Please ensure you answer the "how" question, i.e., how is the step performed? Alternatively, add references to published material specifying how to perform the protocol action. Please see specific comments marked in the attached manuscript.*

Answer: Thanks for the reminding. We have added more details to address the specific comments.

5. *Please upload Supplemental Figure 1 to your Editorial Manager account as a .png, .tiff, .pdf, .svg, .eps, .psd, or .ai file and include a short title and a short description at the end of the Representative Results in the manuscript text.*

Answer: Thanks for this important comment. We have upload Supplemental Figure 1 which named "Supplemental Figure 1.psd" and "Supplemental Figure 1.eps". We have moved the short title and the short description for Supplemental Figure 1 from the end of "SUPPLEMENTARY FILES.docx" to the end of the Representative

Results in the manuscript text.

Changes to be made by the author(s) regarding the video:

1. *Please update the video according to the revised manuscript.*

Answer: We have updated the video according to the revised manuscript.

2. *The homogeneity between the written protocol and the video narration must be increased. All information in the video should be in the written protocol, but not all information in written protocol needs to be in the video. For example:*

0:22-1:08, 4:03-5:09, 5:24-6:00, 8:58-9:25: Details in these parts of the video are not present in the written protocol.

10:02-10:39, 11:00-11:30: These two figures and their descriptions should be added to the manuscript.

Answer: We have added more details in the manuscript to increase the homogeneity between the written protocol and the narration in the video. There are three figures during 10:02-10:39, 11:00-11:30. We have added these three figures to the manuscript as Figure 1, 3 and 4.

3. *Please do not number Representative Results and Conclusions sections.*

Answer: We have deleted the corresponding numbers.

4. *Please upload a revised high-resolution video here:*

https://www.dropbox.com/request/hhckgB9RKIhsf5xwu4sM

Answer: We have uploaded the revised video with high-resolution through

the given website.

Sincerely yours,

KunPeng Dou

On behalf of the authors

Robust staggered band alignment in one-dimensional van der Waals heterostructures: binary compound nanoribbons in nanotubes

M. Gong, G. Zhang, H. H. Hu, L. Kou, K. P. Dou and X. Shi, *J. Mater. Chem. C*, 2019, **7**, 3829
DOI: 10.1039/C9TC00766K

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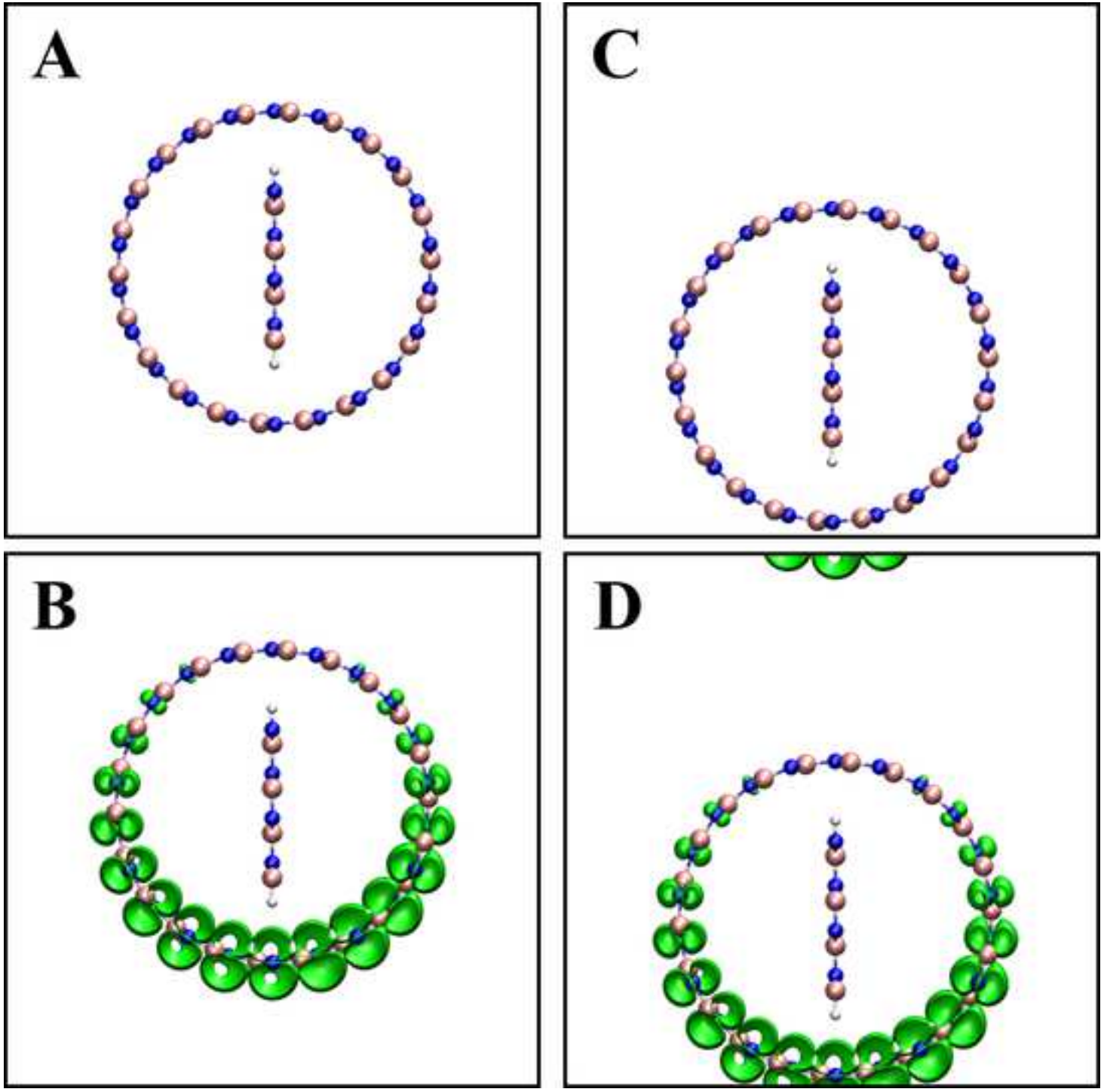

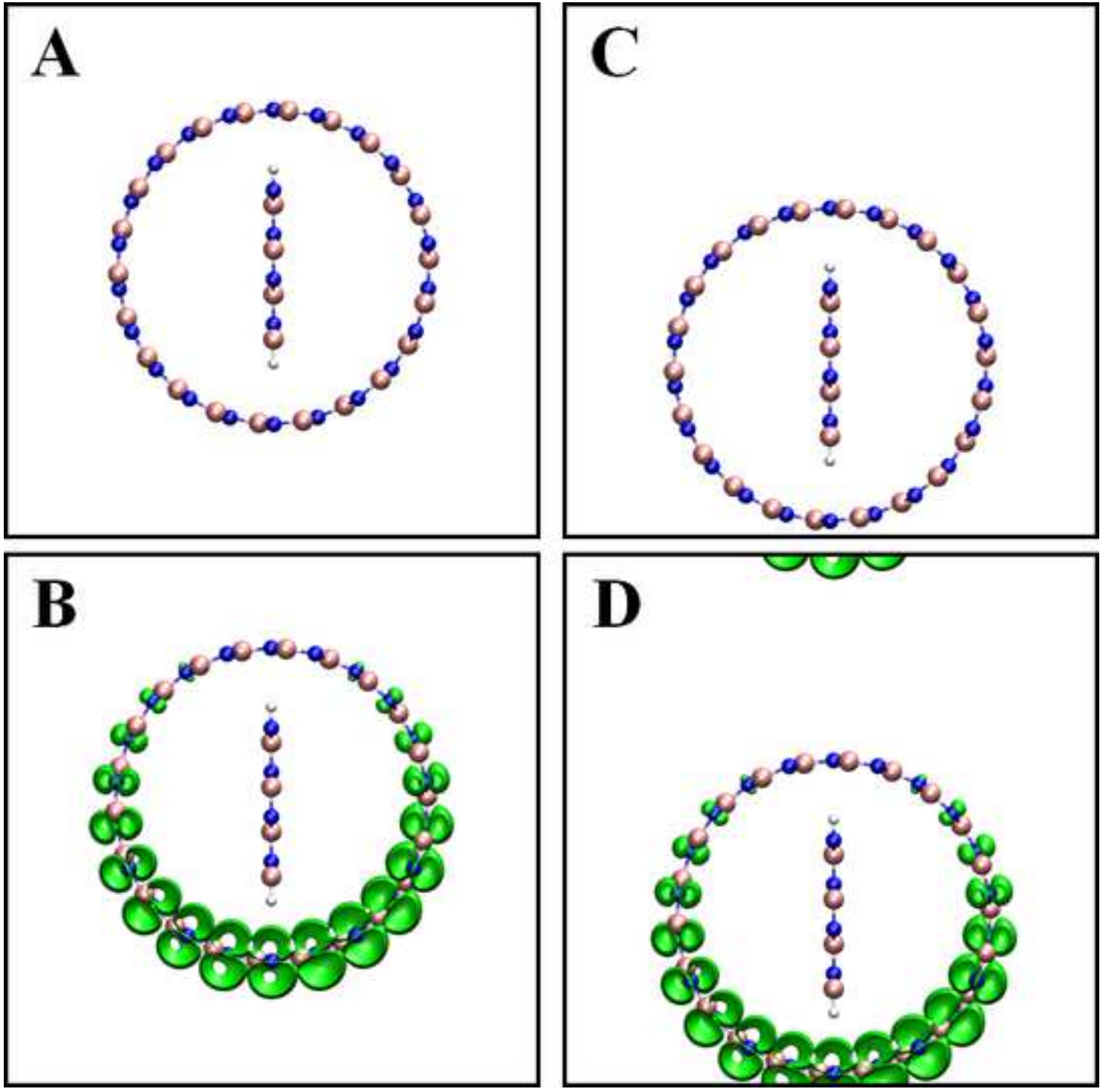

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