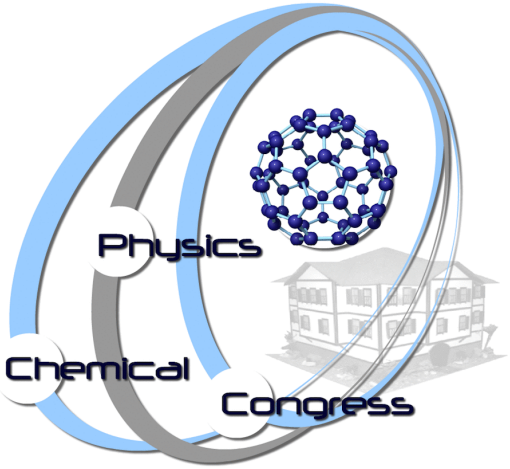
Investigation of Energy Storage Capabilities of Graphene and h-BN Double Layer Electrostatic Supercapacitors



Nazan KARA[1,3], Hatice KÖKTEN[1], Cem ÖZDOĞAN[2]

[1] *Department of Physics, Middle East Technical University, 06800 Ankara, Turkey*
[2] *Department of Engineering Sciences, İzmir Katip Çelebi University, 35620 İzmir, Turkey*
[3] *Grad. School of Natural and Appl. Sciences, Çankaya University, 06530 Ankara, Turkey*

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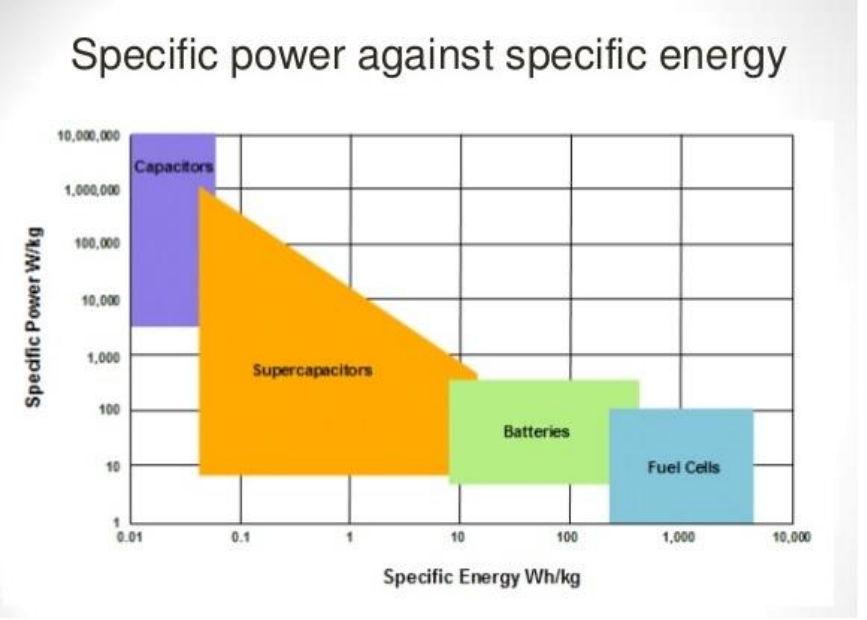
OUTLINE

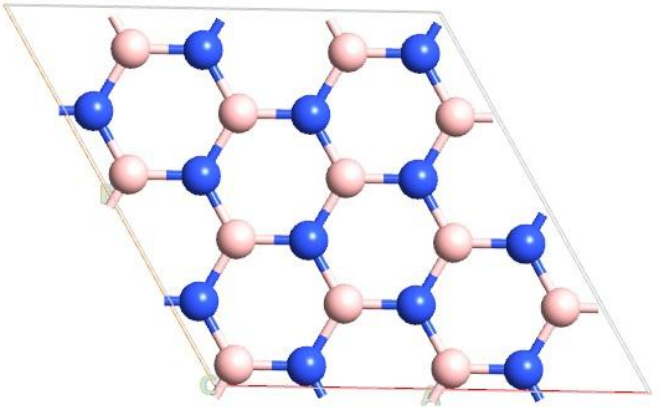
- Introduction
- Models of capacitors
- Methods
- Result and Discusion
  - Graphene Capacitor
  - h-BN Capacitor
  - Silicene Capacitor
- Conclusion

INTRODUCTION

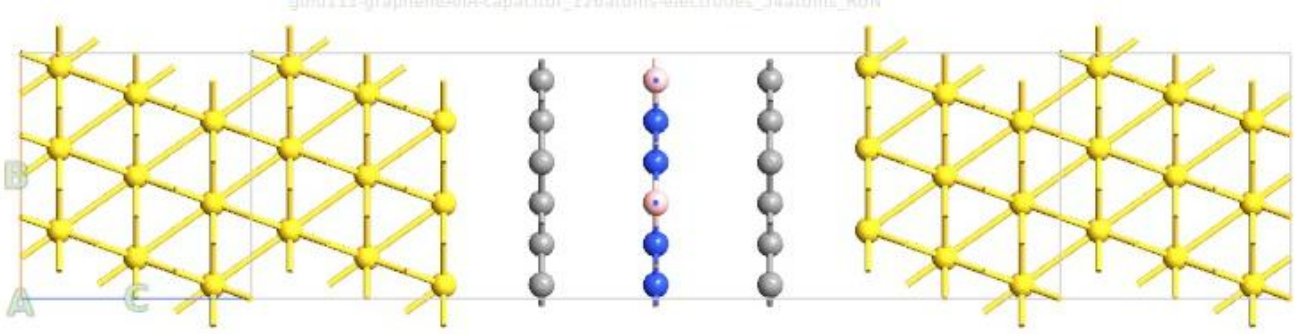
- Quantum capacitance and energy storage capabilities of systems formed in different geometries and shapes are investigated.
- Increasing power density
  - The internal resistance has to be decreased.
  - It can be provided by exchanging of materials.
- Increasing energy density
  - The capacitance has to be increased.
  - Either by increasing the specific capacitance or by optimizing the electrode layers.



MODELS



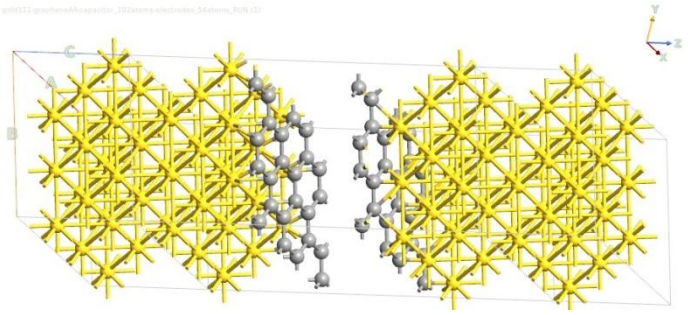
- In the present study; graphene, hexagonal boron nitride (h-BN) and silicene sheets were investigated theoretically as 2-dimesional planar materials in nano size capacitors.
- It is aimed to design nano devices with the best theoretical quantum capacitance value.

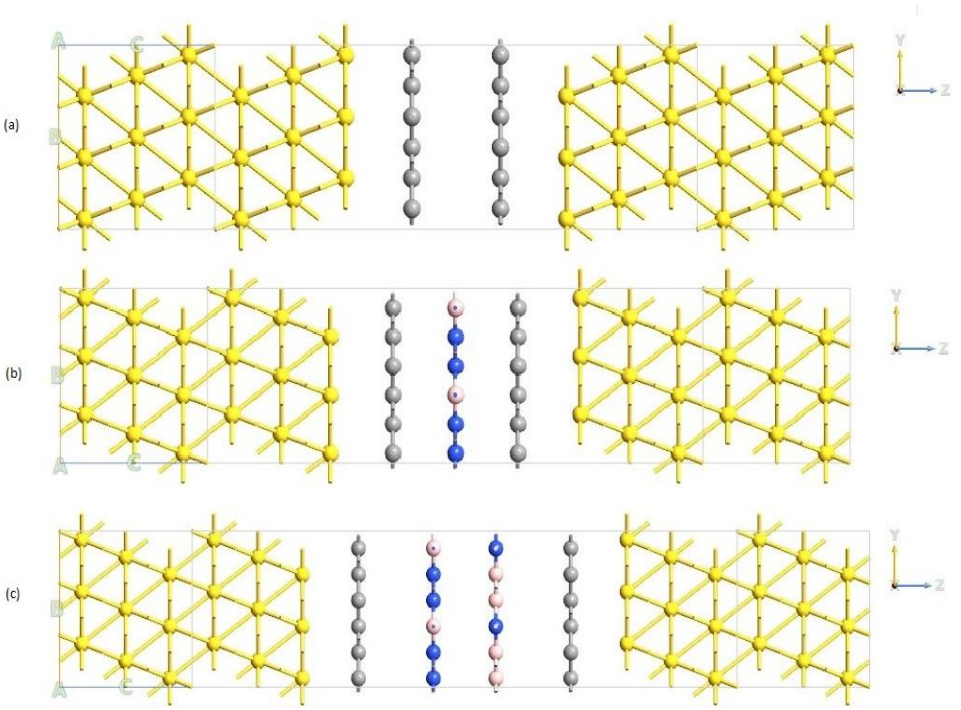


MODELS

Three different capacitor structures were studied:

- Graphene Capacitors
  - Distance Analysis
  - Vacancy Defect Effects
  - Stacking
- h-BN Capacitors
  - Number of h-BN Layers
- Silicene Capacitors





METHOD: Calculation of Capacitance

Electronic transport modelling can be explain in two parts:

1. To obtain self- consistent Hamiltonian matrices for electrodes and central regions.

$$[E - H]\varphi = S'$$

$$\boldsymbol{H} = \begin{pmatrix} H_L + \Sigma_L & V_L & 0 \\ V_L^{\dagger} & H_C & V_R \\ 0 & V_R^{\dagger} & H_R + \Sigma_R \end{pmatrix}$$

- Then Hamiltonian matrices are formed and used to obtain density matrices in the Non-Equilibrium Green Function (NEGF) method in electron transport.
- The potential difference applied to the electrodes.
- Electrons accelerate from low potential to high one. They form a constant equilibrium state current and density in the central region. Density matrix given as;

$$D_{\mu\nu} = \int_{-\infty}^{\infty} d\epsilon \, [\rho_{\mu\nu}^{L}(\epsilon) n_F^{L}(\epsilon - \mu_L) + \rho_{\mu\nu}^{R}(\epsilon) n_F^{R}(\epsilon - \mu_L)]$$

METHOD: Calculation of Capacitance

The Atomistic ToolKit (ATK) Program

- Procedure:
  - Choose initial density
  - Calculate $V_{eff}[n]$
  - Costruct Hamiltonian (**H**) from $V_{eff}[n]$
  - Solve Khon-Sham eigenvalue problem
  - Compute new density n’
  - Control if n=n’
  - Repeat procedure up to self-consistency is achieved.
- Calculations are carried out for zero bias and 0.2V, 0.4V, 0.6V, 0.8V and 1.0 V bias.
- We use k-point sampling (4,4,100) of the Monkhorst type.

METHOD: Calculation of Capacitance

2. Then transmission calculations; to obtain the conduction , I-V characteristics, capacitance etc.
   - The quantum transport quantities, as well as charge distributions, the quantum capacitance of the system can be found by;

$$C_{\alpha\beta} = e\frac{dQ_\alpha}{d\mu_\beta}$$

$$\Delta V = d\mu_\beta / e$$

$$\Delta Q = Q(V + \Delta V) - Q(V)$$

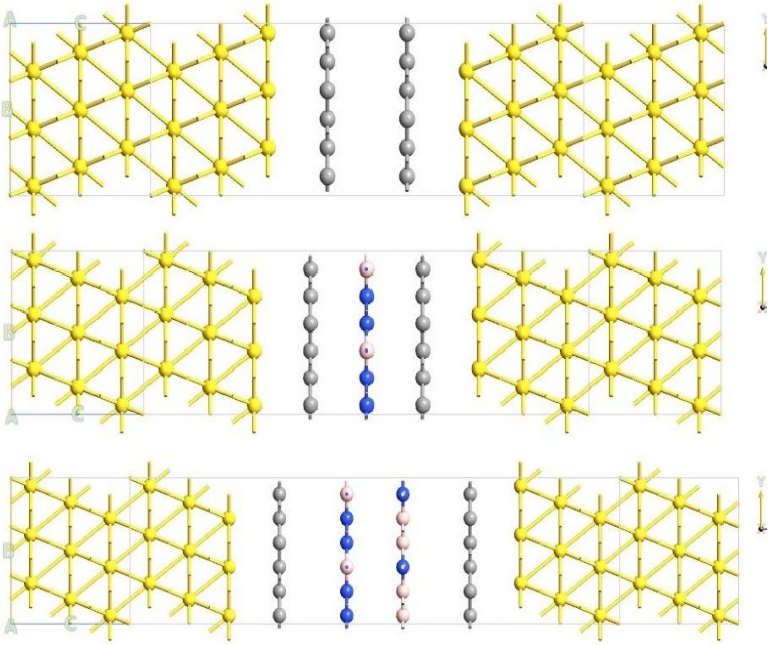
$$E(V) = \frac{1}{2}\int \delta v(r,V)\,\delta n(r,V)dr$$

METHOD

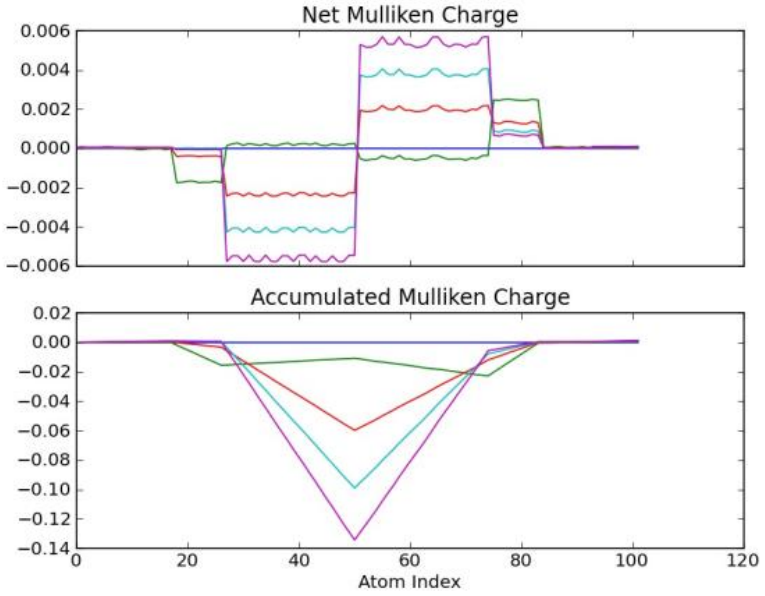
-Spin unpolarized GGA/PBE + NEGF approach is used. (If needed spin polarized calculations will be done.)
-The wave functions are expressed by the Double-Zeta-Polarized (DZP) basic sets, which are local numerical atomic trajectories.
-The optimization for running systems is done by the Fast Relaxation Engine (FIRE) Method.
-150 Ry cut-off energy, Fast Fourier Transformations (FFT) –solutions of Poisson Equations

RESULT: The Graphene Capacitors

- Contact Distances (CD) and Inter-Layer Distances (ILD)
- Number of gold atoms in electrode extensions
- Stacking



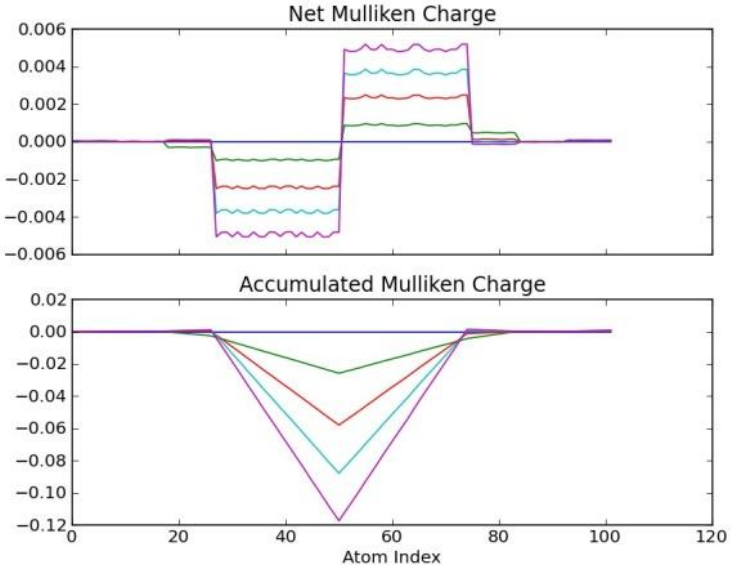
RESULT: The Graphene Capacitors



- CD: 3 Å
- ILD: 3.5 Å

By Mulliken Analysis:

- C = 1.82 x 10^{-20} F



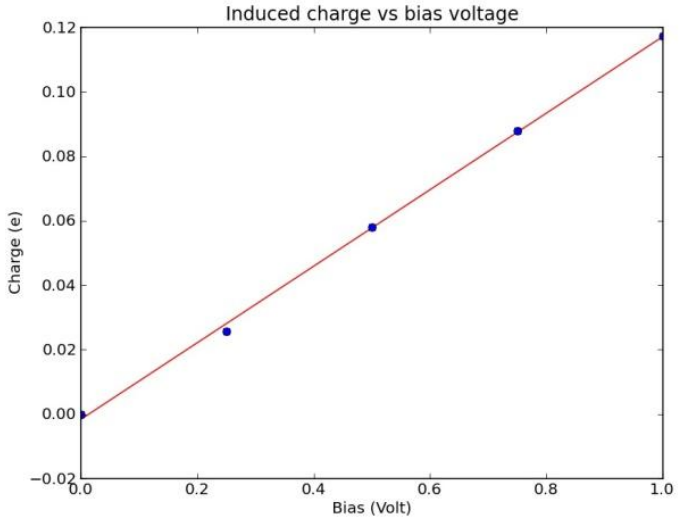
- CD: 3 Å
- ILD: 4.5 Å

By Mulliken Analysis:

- C = 1.90 x 10^{-20} F

RESULT: The Graphene Capacitors

Electrostatic Potential Analysis: CD: 3 Å, ILD: 4.5 Å



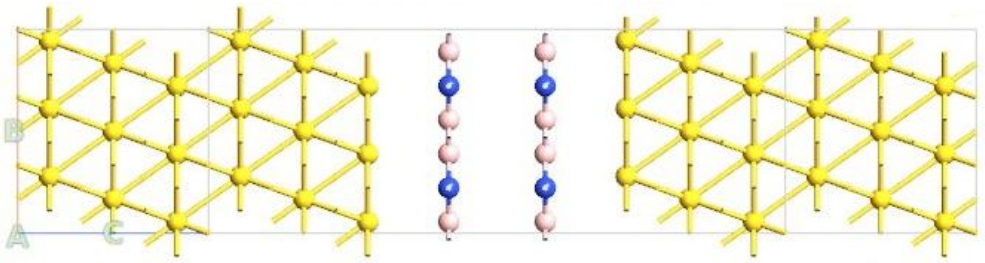
$$E(V) = \frac{1}{2} C V^2$$

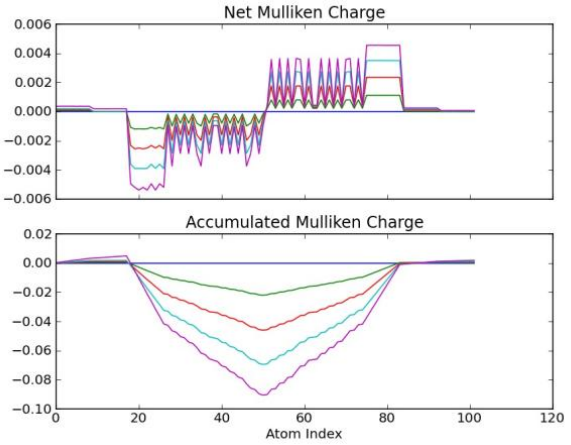
System Name	Capacitance (x10^{-20}F)	
Analysis	Mulliken	Electrostatic Potential
Au/G-G/Au	1.90	1.50

System Name	Specific Capacitance (μF/cm^2)		Gravimetric Capacitance (F/g)	
Analysis	Mulliken	Electrostatic Potential	Mulliken	Electrostatic Potential
Au/G-G/Au	2.93	2.32	39.76	31.41
Au/G-G'/Au	3.37	2.19	45.61	29.66

RESULT: The h-BN Capacitor

- In both directions of the h-BN layers, the induced charge density is shown.
- The charge density between the h-BN layers is higher than the other layers; gold and graphene layer.

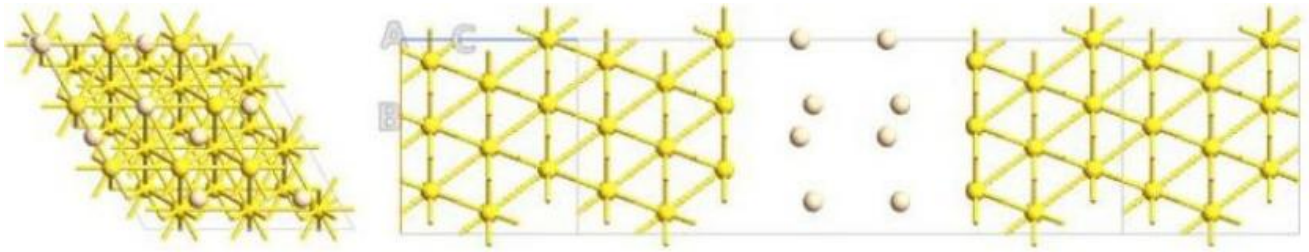


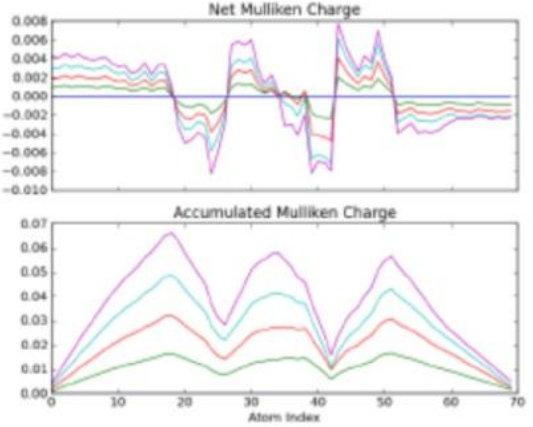


System Name	Specific Capacitance (μF/cm²)		Gravimetric Capacitance (F/g)	
Analysis	Mulliken	Electrostatic Potential	Mulliken	Electrostatic Potential
Au/h-BN/h-BN/Au	2.26	1.29	29.64	16.87
Au/h-BN/h-BN'/Au	2.13	1.26	27.88	16.57

RESULT: The Silicene Capacitors

- The quantum capacitance value obtained was negative and -0.92×10^{-20} F.
- It has been understood that the silicene-based capacitor is not a suitable model system.

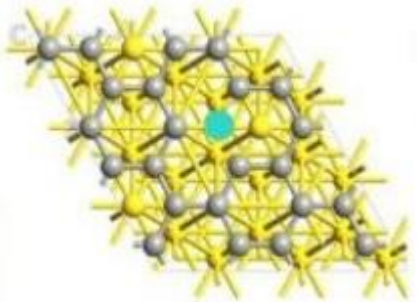
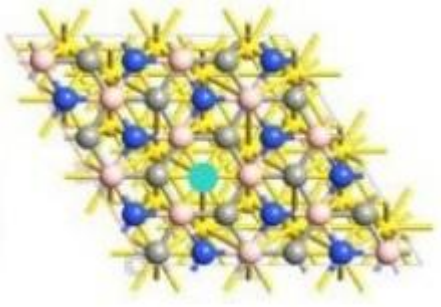
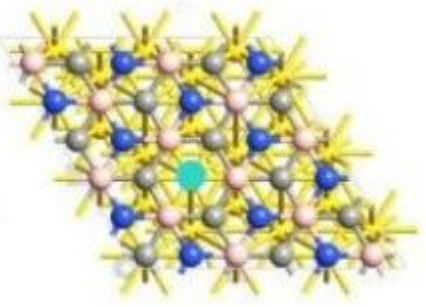




System Name	Specific Capacitance (μF/cm²)		Gravimetric Capacitance (F/g)	
Analysis	Mulliken	Electrostatic Potential	Mulliken	Electrostatic Potential
Au/Si/Si/Au	-1.42	0.67	-24.70	11.65

RESULT: Vacancy Defects

- Vacancy defects were studied by removing one atom for the studied systems.
- With a rough generalization;
  - Vacancy of carbon causes decreasing the capacitance value.
  - Vacancy of boron completely disrupts the structure of the capacitor.
  - Vacancy of nitrogen causes increasing the capacitance value.

FUTURE WORKS

- Different combinations of the structures studied so far will be used as capacitor material.
- Carbon nanotubes will use as materials in between
- The vacancy effects are planned to investigated more deeply.

ACKNOWLEDGMENTS

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