Extended Abstracts of the 2015 International Conference on Solid State Devices and Materials, Sapporo, 2015, pp446-447

Radical Spin Interaction of Graphene Flakes Embedded into h-BN Sheet

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Abstract

Based on the first principles total energy calculation within the framework of density functional theory, we investigate the energetics and magnetic properties of graphene flakes with triangular shape embedded into h-BN sheet. Our calculation shows that the spin polarization energy saturates about 100 meV per graphene flake at the separation of 0.8 nm. The spin-spin interaction, *J*, prefers an antiparallel spin coupling to a parallel one with the energy of 25 meV at the flake-flake distance of 0.5 nm.

1. Introduction

Imperfections in graphene occasionally induce unusual electronic states at the Fermi level (E_F) leading to spin polarization around the imperfections. For instance, graphene nanoribbon with zigzag edges has flat dispersion bands at the E_F, showing magnetic ordering at the edge atomic sites [1]: The polarized electron spin is ferromagnetically aligned along each edge and antiparallely coupled between edges. In addition to graphene nanoribbons, graphene flakes or hydrocarbon molecules possess radical spins owing to the imbalance between the numbers of two sublattices. A phenalenyl molecule is one of the smallest examples of such molecules possessing a radical spin arising from a non-bonding molecular orbital. Recently, a transmission electron microscope experiment has demonstrated that the hexagonal boron nitride (h-BN) possesses triangular multi-vacancies [2]. Because of the structural similarity between the graphene and h-BN, these triangular vacancies in h-BN can be filled by graphene nanoflakes with triangular shapes, leading to BNC heterosheets with the spin polarization. In our previous work, we demonstrated that the BNC heterosheet exhibit magnetic ordering depending on the shape of the graphene flakes [3]. Although the several theoretical works investing the fundamental properties of BNC heterosheet, the detailed energetics and spin-spin interaction is not addressed yet. Thus, in this work, we explore energetics and magnetic properties of graphene flakes with the phenalenyl structure embedded into h-BN using density functional theory (DFT) with generalized gradient approximation (GGA) for elucidating the theoretical insight into the radical spin interaction between the flakes.

2. Calculation Methods and Structural Models

All calculations were performed within the framework of DFT. We used GGA with spin degree of freedom for calculating the exchange-correlation energy between electrons. A Vanderbilt ultrasoft pseudopotential was used to describe the electron-ion interaction. The valence wave functions and charge density were expanded in terms of plane waves with cutoff energies of 25 and 255 Ry, respectively. In the present work, we consider the two graphene flakes with phenalenyl shapes embedded into h-BN as shown in Fig. 1 To investigate the spin interaction with respect to the radical spin, we consider various inter flake spacing of 0.25, 0.5, 0.75, 1.00, 1.25, and 1.50 nm.

3. Results

Figure 2 shows calculated total energy of BNC sheet as a function of the distance between phenalenyl flakes in h-BN. We find that the total energy of flakes monotonically increase with increasing the interflake spacing. The spin polarized states are energetically favorable compared with the nonmagnetic states. Furthermore, the total energies of the antiparallel spin states are lower than that of the parallel spin arrangement for the small interflake spacing up to 0.5 nm, while the energies almost degenerate each other for the flake spacing of 0.75 nm or larger indicating that the spin interaction rapidly vanishes with increasing the flake spacing. The increase of the total energy is ascribed to the increase of localization of electrons near E_F, those are distributed onto the graphene flake, with increasing the interflake spacing.

Figure 3 shows the spin density of graphene flakes embedded into h-BN with the flake-flake spacing of 1 nm in the antiparallel and parallel spin arrangements. Spin density reflects the bipartite nature of the flakes. The imbalance between two sublattices in each flake leads to spin moment 1 μ_B on the flakes which are coupled into parallel and antiparallel as their metastable states.

Figure 4 shows the spin-spin interaction, *J*, which is calculated by the energy difference between antiparallel and parallel spin states. The spin interaction possesses the minimum at the interflake spacing of 0.5 nm with the energy of J=25 meV, indicating that the flakes prefer the antiparallel spin arrangement to the parallel spin arrangement. However, the *J* rapidly approaches zero with increase in the interflake spacing. The fact indicates that the radical spin interaction among the graphene flakes embedded into h-BN is the short-range interaction of which interaction range is 0.75 nm or less.

4. Conclusions

Based on the DFT with GGA calculations, we investigated energetics and magnetic properties of graphene flakes embedded into h-BN sheet. We found that polarized electron spin emerges at the graphene flakes due to the sublattice imbalance and the spins are arranged in antiparallel and parallel manners. Our calculations also show the spin interaction J between two flakes has the maximum of 25 meV with antiparallel spin arrangement for the interflake spacing of 0.5 nm. The results indicate that the graphene flakes with triangular shapes embedded into h-BN act as weakly interacting spin dots.

Acknowledgements

MM acknowledges the Grant-in Aid for JSPS Fellows. All calculations are performed on NEC SX-Ace at the Cybermedia Center, Osaka University.

References

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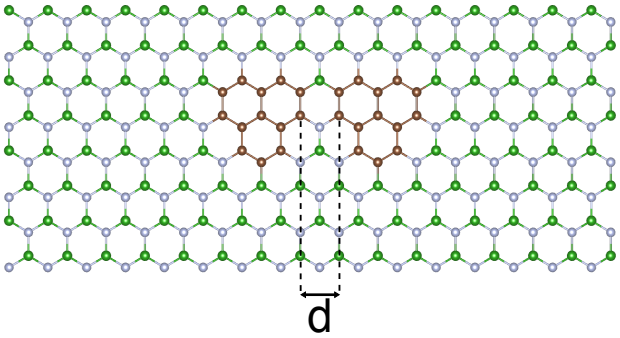

Fig. 1 Structural model of graphene flakes embedded into a h-BN sheet. Green, white, and brown circles denote B, N, and C atoms, respectively.

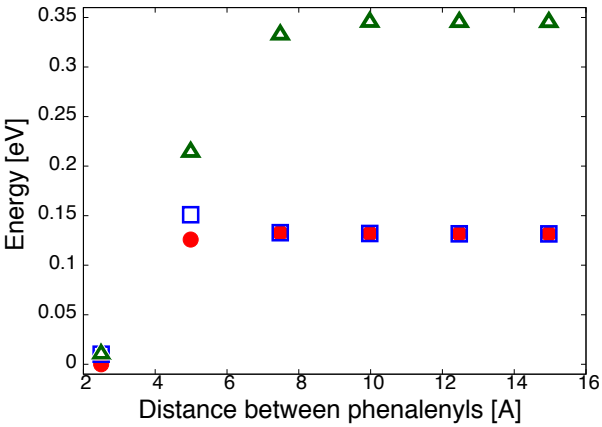

Fig. 2 Calculated total energy of graphene flakes embedded into h-BN sheet as a function of distance between flakes. Energies are measured from the flakes with d = 0.25 nm in antiparallel state. Circles, squares, and triangles denote the total energies of the antiparallel spin arrangement, parallel spin arrangement, and nonmagnetic states, respectively.

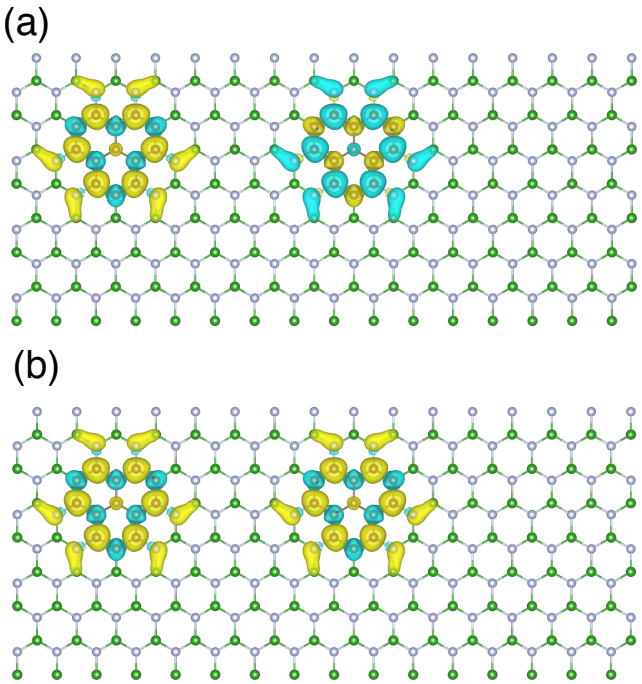

Fig. 3 Ispsurfaces of spin densities of graphene flakes embedded into h-BN with the flake-flake spacing of 1 nm in (a) an antiparallel and (b) parallel spin arrangements. Colors denote the sign of spin components. Green, white, and brown circles denote B, N, and C atoms, respectively.

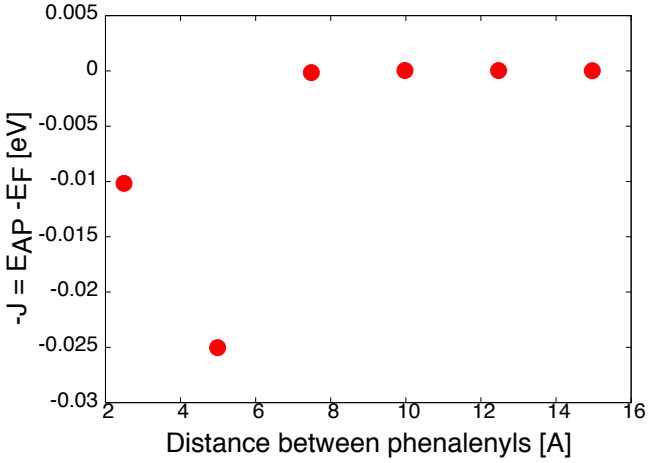

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Fig. 4 The spin-spin interaction J between two graphene flakes as a function of the interflake spacing.