ELECTRONIC STRUCTURES IN C\textsubscript{60}-POLYMERS

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Variations in the band structures of C\textsubscript{60}-polymers are studied, when conjugation conditions and the electron number are changed. We use a semiempirical model with the Su-Schrieffer-Heeger type electron-phonon interactions. In the neutral one-dimensional C\textsubscript{60}-polymer, electronic structures change among direct-gap insulators and the metal, depending on the degree of conjugations. High pressure experiments could observe such pressure-induced metal-insulator transitions. The C\textsubscript{60}-polymer doped with one electron per one molecule is always a metal. The energy difference between the highest-occupied state and the lowest-unoccupied state of the neutral system becomes smaller upon doping owing to the polaron effects. When the C\textsubscript{60}-polymer is doped with two electrons per one C\textsubscript{60}, the system is insulating. When the conjugation in the direction of the polymer chain is smaller, it is a direct-gap insulator. The energy gap becomes indirect when the conjugation is stronger. We also study the antiferromagnetic phase of AC\textsubscript{60} by using a tight-binding model with long-range Coulomb interactions. The antiferromagnetism is well described by the model. The comparison with the photoemission studies shows that the new band around the Fermi energy of the AC\textsubscript{60} phase can be explained by the extremely large intrusion of an energy level into the gap of the neutral system. This indicates that the interaction effects among electrons are important in doped C\textsubscript{60}-polymers.

INTRODUCTION

Recently, it has been found that the linear (one-dimensional) C\textsubscript{60}-polymers are realized in alkali-metal doped C\textsubscript{60} crystals: AC\textsubscript{60} (A = K, Rb, Cs) [1-5], and their solid state properties are intensively investigated. One electron per one C\textsubscript{60} is doped in the polymer chain. It seems that Fermi surfaces exist in high temperatures, but the system shows antiferromagnetic correlations in low temperatures [1]. The C\textsubscript{60}-polymer has lattice structures where C\textsubscript{60} molecules are arrayed in a linear chain. The bonds between C\textsubscript{60} are formed by the \([2+2]\) cycloaddition mechanism. The structures of the one-dimensionals polymers are shown in Fig. 1. There are four membered rings between neighboring C\textsubscript{60} molecules.

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In this article, we shall review studies on variations in the band structures of the one-dimensional C$_{60}$-polymers when the conjugation conditions between molecules are changed. We assume that the bonding states between C$_{60}$ might be changed easily possibly by applying high pressures. As discussed in ref. [2], there are three candidates for the classical bonding structures around the four membered ring. The first case (A) is that there are only weak van der Waals interactions between C$_{60}$, and the classical double bonds remain as in the isolated C$_{60}$ molecule. The second case (B) is that all the bonds of the four membered ring are the single bond, so they are $\sigma$-like in the quantum chemistry. All the four-membered rings in Fig. 1 are formed by the $\sigma$-bonds in this case. Then, the third case (C) is that the bonds which connect the neighboring molecules are the double bonds, and the bonds derived from the isolated C$_{60}$ – the bonds, $\langle 1,2 \rangle$ and $\langle 3,4 \rangle$, of Fig. 1 – are destroyed completely. Here, $\langle i, j \rangle$ indicates the pair of the neighboring i and jth atoms. In this case, the bonds between C$_{60}$ – the bonds, $\langle 1,3 \rangle$ and $\langle 2,4 \rangle$, of Fig. 1 – have the $\sigma$- as well as $\pi$-characters. In other words, the degree of the conjugations between the molecules becomes maximum. We have proposed a model which can deal with changes of conjugation conditions among the above three cases, and have looked at electronic band structures of C$_{60}$-polymers [6,7]. We note that the operator at the lattice sites of the four membered rings is one of the relevant linear combinations of $sp^3$ orbitals, assuming a possibility of local $\sigma$-conjugations at the four membered rings. The similar assumption of the $\sigma$-conjugation has been used in Si-based polymers, for example, in ref. [8]. We, however, use the term “conjugation” for simplicity in this article, because the local $\sigma$-conjugations can be regarded as a part of the global conjugations which are extended over the system.

In refs. [6,7], we have proposed a semiempirical tight-binding model analogous to the Su-Schrieffer-Heeger (SSH) model [9] of conjugated polymers. The first purpose of this article is to review the model and the main results. The electronic structures may depend sensitively upon the conjugation conditions even in the neutral polymer, because the several bonds connecting neighboring molecules are largely distorted, and the mixings among $sp^3$ orbitals will change only by slight change of the bond structures [2]. We have studied effects of the change of the conjugation conditions by introducing a phenomenological parameter in a tight-binding model. The model is an extension of the SSH-type model which has been applied to C$_{60}$ [10,11] and C$_{70}$ [11,12] molecules. The model is solved with the assumption of the adiabatic approximation, and band structures are reported in order to discuss metal-insulator changes and polaron effects by varying conjugations.

We have concluded that the electronic structures change among direct-gap (indirect-gap) insulators and the metal, depending on the degree of conjugations. In the neutral polymer, the electronic structures change from the direct gap insulator with the gap at the $\Gamma$ point, through the metal, to the insulator with the direct gap at the Brillouine zone boundary, as increasing the conjugation between C$_{60}$ molecules. The high pressure experiments may be able to change conjugation conditions between C$_{60}$
molecules, and the electronic structure changes could be observed. The reentrant change from the insulator through a metal into an insulator is specific to the present C\textsubscript{60}-polymer systems and is quite interesting.

We will also review the doping effects in one-dimensional polymers [13]. We discuss the following properties. (1) The C\textsubscript{60}-polymer doped with one electron per one molecule is always a metal. The energy difference between the highest-occupied state and the lowest-unoccupied state of the neutral system becomes smaller upon doping owing to the polaron effects. (2) When the C\textsubscript{60}-polymer is doped with two electrons per one C\textsubscript{60}, the system is insulating. When the conjugation in the direction of the polymer chain is smaller, it is a direct-gap insulator. The energy gap becomes indirect when the conjugation is stronger.

The last purpose of this article is to review a microscopic model for the antiferromagnetism in one-dimensional C\textsubscript{60}-polymers [14] in order to show that the antiferromagnetic ground state can be actually explained by our model with strong electron-electron interactions. We have pointed out the importance of Coulomb interactions, comparing with photoemission experiments of C\textsubscript{60}-polymer systems [15]. The model is one dimensional and has two contributions. The first term is the hopping interactions along the polymer backbone structure, and the second term is the Coulomb interaction among electrons. We use the long-range Ohno potential which we have used in the previous paper [16]. In ref. [17], the relevance of the Hubbard model, where the C\textsubscript{60} molecule is regarded as a site, has been discussed. We will compare the present calculation with the assumption of the Hubbard model.

**MODELS**

We first apply an SSH-type model to C\textsubscript{60}-polymers. In C\textsubscript{60}, all the carbon atoms are equivalent, so it is a good approximation to neglect the mixing between \pi- and \sigma-orbitals. The presence of the bond alternation and the energy level structures of the neutral C\textsubscript{60} molecule can be quantitatively described by the calculations within the adiabatic approximation. In C\textsubscript{70}, the molecular structure becomes longer, meaning that the degrees of the mixing between \pi- and \sigma-characters are different depending on carbon sites. In this respect, the extended SSH model does not take account of the differences of the mixings. However, it has been found [11,12] that qualitative characters of the electronic level structures are reasonably calculated when the extended SSH model is applied to the C\textsubscript{70}. This is a valid approach because the \sigma-orbitals can be simulated by the classical harmonic springs in the first approximation.

In this article, we assume the same idea that the lattice structures and the related molecular orbitals of each C\textsubscript{60} molecule in the C\textsubscript{60}-polymers can be described by the SSH-type model with the hopping interactions for the \pi-orbitals and the classical springs for the \sigma-orbitals. However, the mixings between the \pi- and \sigma-orbitals near
the eight bonds, \( \langle i, j \rangle (i, j = 1 - 4) \), shown in Fig. 1 are largely different from those of regions far from the four bonds. We shall shed light on this special character of bondings between the neighboring \( \text{C}_{60} \). Electronic structures would be largely affected by changes of conjugation conditions (or local \( \sigma \)-conjugations as in Si-based polymers [8]) around the four bonds. We shall introduce a semiempirical parameter \( a \) as shown in the following hamiltonian:

\[
H_{\text{pol}} = a \sum_{l, \sigma} \sum_{\langle i, j \rangle} \left( -t + \alpha y_{l, \langle i, j \rangle} \right) (c_{l, i, \sigma}^\dagger c_{l+1, j, \sigma} + \text{h.c.}) \\
+ (1 - a) \sum_{l, \sigma} \sum_{\langle i, j \rangle} \left( -t + \alpha y_{l, \langle i, j \rangle} \right) (c_{l, i, \sigma}^\dagger c_{l, j, \sigma} + \text{h.c.}) \\
+ \sum_{l, \sigma} \sum_{\langle i, j \rangle = \text{others}} \left( -t + \alpha y_{l, \langle i, j \rangle} \right) (c_{l, i, \sigma}^\dagger c_{l, j, \sigma} + \text{h.c.}) \\
+ \frac{K}{2} \sum_{i} \sum_{\langle i, j \rangle} y_{l, \langle i, j \rangle}^2 \tag{1}
\]

where \( t \) is the hopping integral of the system without the bond alternations in the isolated \( \text{C}_{60} \) molecule; \( \alpha \) is the electron-phonon coupling constant which changes the hopping integral linearly with respect to the bond variable \( y_{l, \langle i, j \rangle} \), where \( l \) means the \( l \)th molecule and \( \langle i, j \rangle \) indicates the pair of the neighboring \( i \) and \( j \)th atoms; the atoms with \( i = 1 - 4 \) of the one-dimensional polymer are shown in Fig. 1; the other \( i \) and \( j \) in the third column of eq. (1) label the nonnumbered atoms in the same molecule; \( c_{l, i, \sigma} \) is an annihilation operator of the \( \pi \)-electron at the \( i \)th site of the \( l \)th molecule with spin \( \sigma \); the sum is taken over the pairs of neighboring atoms; and the last term with the spring constant \( K \) is the harmonic energy of the classical spring simulating the \( \sigma \)-bond effects. Note that the sum with the prime is performed over \( \langle i, j \rangle = \langle 1, 3 \rangle \) and \( \langle 2, 4 \rangle \). The sum with the double prime is performed over \( \langle i, j \rangle = \langle 1, 2 \rangle \) and \( \langle 3, 4 \rangle \).

As stated before, the parameter \( a \) controls the strength of conjugations between neighboring molecules. When \( a = 1 \), the \( \sigma \)-bodings between atoms, 1 and 2, 3 and 4, in Fig. 1 are completely broken and the orbitals would become like \( \pi \)-orbitals. The bond between the atoms 1 and 3 and the equivalent other bonds become double bonds. This is the case (C). As \( a \) becomes smaller, the conjugation between the neighboring molecules decreases, and the \( \text{C}_{60} \) molecules become mutually independent. In other words, the interactions between molecules become smaller in the intermediate \( a \) region. In this case (B), the operator \( c_{l, i, \sigma} \) at the lattice sites of the four membered rings is one of the relevant linear combinations of the \( sp^3 \) orbitals. Here, we assume a possibility of local \( \sigma \)-conjugations at the four membered rings. When \( a = 0 \) [the case (A)], the \( \text{C}_{60} \) molecules are completely isolated each other. The band structures of the \( \text{C}_{60} \)-polymers will change largely depending on the conjugation conditions. The effects are reviewed in the next section.

We further discuss the mechanism of the antiferromagnetism in one-dimensional
C_{60}-polymers doped with one electron per one C_{60}. The model is the following:

\[
H = H_0 + H_{\text{int}},
\]

\[
H_0 = -at \sum_{l,\sigma} \sum_{(i,j)=(1,3),(2,4)} (c_{l,i,\sigma}^\dagger c_{l+1,j,\sigma} + \text{h.c.})
- (1-a)t \sum_{l,\sigma} \sum_{(i,j)=(1,2),(3,4)} (c_{l,i,\sigma}^\dagger c_{l,j,\sigma} + \text{h.c.})
- t \sum_{l,\sigma} \sum_{(i,j)=\text{others}} (c_{l,i,\sigma}^\dagger c_{l,j,\sigma} + \text{h.c.}),
\]

\[
H_{\text{int}} = U \sum_l (c_{l,i,\uparrow}^\dagger c_{l,i,\uparrow} - \frac{n_{\text{el}}}{2})(c_{l,i,\downarrow}^\dagger c_{l,i,\downarrow} - \frac{n_{\text{el}}}{2})
+ \sum_{l,l',i,j} W(r_{l,l',i,j}) \left( \sum_{\sigma} c_{l,i,\sigma}^\dagger c_{l,i,\sigma} - n_{\text{el}} \right) \left( \sum_{\tau} c_{l',j,\tau}^\dagger c_{l',j,\tau} - n_{\text{el}} \right).
\]

In eq. (2), the first term is the tight binding part of the C_{60}-polymer, and the second term is the Coulomb interaction potential among electrons. The eq. (3) is equivalent to eq. (1), if \( \alpha = 0 \). The electron-phonon interactions are not taken into account, because they are not necessary in order to show the presence of the antiferromagnetism. The numbers in eq. (3) indicate the carbon atoms displayed in Fig. 1. Equation (4) is the Coulomb interactions among electrons. Here, \( n_{\text{el}} \) is the number of electrons per carbon site; \( r_{l,l',i,j} \) is the distance between the \( i \)th site of the \( l \)th C_{60} and \( j \)th site of the \( l' \)th C_{60}; and

\[
W(r) = \frac{1}{\sqrt{(1/U)^2 + (r/r_0 V)^2}}
\]

is the Ohno potential. The quantity \( W(0) = U \) is the strength of the onsite interaction; \( V \) means the strength of the long range part; and \( r_0 = 1.433 \text{Å} \) is the mean bond length of the single C_{60} molecule.

**DOPING EFFECTS AND ELECTRONIC STATES IN ONE-DIMENSIONAL C_{60}-POLYMERS**

We use the parameters, \( t = 2.1 \text{eV} \), \( \alpha = 6.0 \text{eV/Å} \), and \( K = 52.5 \text{eV/Å}^2 \), which give the energy gap 1.904eV and the difference between the short and bond lengths 0.04557Å for an isolated C_{60} molecule.

Figure 2 shows the excess electron distribution for the three conjugation conditions, \( a = 0.5, 0.8, \) and 1.0. The labels of sites, A-I, are shown in Fig. 1. Due to the reduced symmetry of the polymer chain, mutually symmetry equivalent sites have the same electron density. The each label represents the site with the different electron density. The white bars are for the case of the excess electron number per
$C_{60}$, $N_{\text{ex}} = 1$, and the black bars are for $N_{\text{ex}} = 2$. The excess electron density at the sites A is the largest for all the displayed cases. The bond alternation patterns are largely distorted near these sites, so the electron density change is the largest too. In Fig. (a), the densities at sites D, F, and H, are relatively larger. In Figs. (b) and (c), the densities are larger at sites D and I. In this way, the sites, where excess electrons prone to accumulate and thus the bond alternation patterns are highly distorted, are spatially localized in the molecular surface. This is one of the polaron effects, which we have discussed in ref. [11]. Here, we do not show bond alternation patterns for simplicity. We only note that the distortion of the bond alternation is larger where the change of the electron density is larger. The polaronic distortion pattern is different from that in the isolated $C_{60}$, and this is owing to the difference in the symmetry group. Tanaka et al [17] have drawn a schematic figure where the electron density change is the largest at the molecule center, but the present result does not agree with this feature. Numerical calculations are necessary in order to derive the actual distributions.

Next, we discuss band structures of electrons in detail. Figures 3-5 display the band structures for the conjugation parameters, $a = 0.5$, 0.8, and 1.0, respectively. Figures (a), (b), and (c) are for $N_{\text{ex}} = 0$, 1, and 2, respectively. In each figure, the unit cell is taken as unity, so the first Brillouine zone extends from $-\pi$ to $\pi$. Due to the inversion symmetry, only the wavenumber region, $0 \leq k \leq \pi$, is shown in the figures.

Figures 3 (a-c) show the band structures of the polymer for $a = 0.5$ and with $N_{\text{ex}} = 0$, 1, and 2, respectively. In Fig. 3(a), the highest fully occupied band is named as “HOMO”, and the lowest empty band as “LUMO”. There is an energy gap about 0.8 eV at the zone center. The system is a direct gap insulator. When doped with one electron per $C_{60}$, the system is a metal as shown by the presence of the Fermi surface in Fig. 3(b). The system is an insulator again when $N_{\text{ex}} = 2$, as shown in Fig. 3(c). Here, the energy gap is at the boundary of the Brillouine zone, i.e., at $k = \pi$.

As increasing the parameter $a$, the overlap of the HOMO band and LUMO band appears in the neutral system. This is shown for $a = 0.8$ in Fig. 4(a). There are Fermi surfaces, so the system changes into a metal. If $a$ increases further, the positions of the previous HOMO band and LUMO band are reversed as shown for $a = 1.0$ in Fig. 5(a). The system becomes a direct gap insulator again. The energy gap is at $k = \pi$.

When $N_{\text{ex}} = 1$, the system is always a metal when $a$ varies. The representative cases, $a = 0.8$ and 1.0, are displayed in the Figs. 4(b) and 5(b). The number of the Fermi surface is two or four, depending upon the parameter $a$. However, the metallic property is obtained for all the $a$ we take. We also find that the HOMO band and LUMO band of the neutral system shift into the energy gap upon doping. The positions of the other energy bands do not change so largely. This is due to the polaronic distortion of the lattice, which we have discussed in the calculation of an
isolated molecule in ref. [11].

When $N_{ex} = 2$, the system turned out to be always an insulator. For smaller $a$, for example, $a = 0.5$ and 0.8, the energy gap appears at $k = \pi$. For larger $a$, for example, $a = 1.0$ [Fig. 5(c)], the energy gap becomes an indirect gap. The polaronic distortion becomes larger as the doping concentration increases. Thus, the intrusions of the HOMO and LUMO bands of the neutral system become larger, too.

The above variations of the energy gap are summarized for the cases $N_{ex} = 0$ and 2, where a finite energy gap appears for a certain $a$ value. The results are shown in Figs. 6(a) and (b). The white (black) squares indicate that the system is a direct gap insulator where there is a energy gap at $k = 0$ ($\pi$). The squares with the plus mean that the system is an indirect gap insulator. The crosses are for metals. In the neutral system $N_{ex} = 0$ [Fig. 6(a)], the energy gap decreases almost linearly for smaller $a$. The system changes into a metal as $a$ increases, and finally an energy gap appears again. For $N_{ex} = 2$ shown in Fig. 6(b), the system is a direct gap insulator with the energy gap at $k = \pi$ up to $a \sim 0.9$. The system turns into an indirect gap insulator near $a = 1.0$.

**MAGNETISM IN ONE-DIMENSIONAL C$_{60}$-POLYMERS**

The model eq. (2) is solved with the assumptions of the unrestricted Hartree-Fock approximation. We assume that one electron is doped per one C$_{60}$ molecule and $V = U/2$. The results are shown by changing the parameters, $a$ and $U$.

The magnitude of the ordered spin per one C$_{60}$ is calculated, and is shown in Fig. 7 as a function of $a$ and $U$. If the magnitude is zero, there is not an antiferromagnetic order. The magnitude is finite when the magnetic order appears. As $U$ becomes stronger, the magnetization appears and becomes larger. This is a natural consequence. When $a$ is smaller, the itinerancy of electrons in the chain direction decreases, and thus the antiferromagnetism appears more easily. This feature is realized in the present calculation: the critical value of $U$, where a finite magnetization begins to appear, decreases as $a$ becomes smaller.

The calculated results are summarized as a phase diagram in Fig. 8. The metallic phase region is named as M. And the antiferromagnetic phase is named as AF. The antiferromagnetism appears in the small $a$ region when $U$ is taken constant. The phase boundary between two phases is an almost linear line. In ref. [17], the applicability of the one-dimensional Hubbard model is discussed. When this discussion is adopted to the present model eq. (2), the model can be mapped to the following hamiltonian:

$$H = -t_{\text{eff}} \sum_{\langle l,l' \rangle,\sigma} (a_{l,\sigma}^{\dagger} a_{l',\sigma} + \text{h.c.}) + U_{\text{eff}} \sum_{l} n_{l,\uparrow} n_{l,\downarrow},$$

(6)
where $t_{\text{eff}}$ is an effective hopping integral between neighboring $C_{60}$ molecules; $U_{\text{eff}}$ is the on-ball Coulomb repulsion; $a_{l,\sigma}$ is an annihilation operator of an effective orbital of the doped electron at the $l$th molecule; and $n_{l,\sigma} = a_{l,\sigma}^\dagger a_{l,\sigma}$. In the one-dimensional Hubbard model, it is known that the antiferromagnetism appears even when $U_{\text{eff}}$ is small but positive. But, there is a parameter region where a magnetic order is not present in Fig. 8. Thus, we find that the mapping onto the Hubbard model is not always relevant to all the parameter set. The mapping could be used for the case with small $a$ and large $U$. In ref. [17], the antiferromagnetism has been discussed in connection with the polaron in the doped $C_{60}$ [11]. In the present section, electron-phonon interactions are not considered, and only electron-electron interactions are taken into account. Therefore, the antiferromagnetism is not directly related with the polaron formation due to the Jahn-Teller effects [11]. Rather, we could regard the antiferromagnetism as one of the spin density wave states in the itinerant electron systems. It is of course that the doped extra electrons tend to have large amplitudes at certain sites on the surface of the $C_{60}$ molecules. However, the main origin of the localization would be the strong electron-electron interactions rather than the Jahn-Teller effects.

It is useful to look at how the band structures change upon the formation of the antiferromagnetic order. The dispersions of the bands of the system before the electron doping and without the magnetic order are shown for $(a, U, V) = (0.3, 2.4t, 1.2t)$ in Fig. 9. Here, the electron number is 60 per one $C_{60}$, and the unit cell consists of one $C_{60}$. The band structures of the antiferromagnetic system are shown for the same $(a, U, V)$ in Fig. 10. The unit cell becomes doubled, so the first Brillouine zone is half of that of Fig. 9. The energy bands in Fig. 10 become narrower. The band named “HOMO” in Fig. 9 changes into the HOMO-1 band and the HOMO-2 band in Fig. 10. There is a small energy gap between the HOMO-1 band and HOMO-2 band at the boundary of the first Brillouine zone in Fig. 10. In the same way, the band named “LUMO” of Fig. 9 is the origins of the “HOMO” and “LUMO” of Fig. 10. There is a large energy gap at the zone boundary as well. These energy gaps are opened by the formation of the antiferromagnetism.

It should be noted that the energy band intrudes into the original energy gap of the neutral system extremely largely. The HOMO band of the system with the antiferromagnetism even locates near the center of the original energy gap. This intrusion is much larger than that in the polaron formation due to the Jahn-Teller interactions in the doped $C_{60}$ [11], and cannot be obtained from models with electron-phonon interactions only. Actually, the new band has been observed in the photoemission studies of the orthorhombic phase of Rb$C_{60}$ [15]. Such the large changes in band structures might be the evidence of strong electron-electron interactions in the $C_{60}$-polymers rather than the results of the Jahn-Teller distortion due to the electron-phonon interactions. The positions of the HOMO band and HOMO-1 band of Fig. 9 do not shift so much upon the formation of the antiferromagnetism. This is also consistent with the photoemission studies [15].
In the present calculations, we have not considered possible interchain interactions which might effect on the dispersion of bands perpendicular to the polymer direction [18,19]. In the tight-binding type models as used in this article, there might be many candidates for the interchain interaction forms. Even though they have not been taken into account in the present calculations, we might expect that the nature of one-dimensinal chains with strong electron-electron interactions is still effective on the antiferromagnetism in the real AC$_{60}$ systems because of the low dimensionality of the polymer-chain which might be favorable for the orderings to occur due to interactions among itinerant electrons.

SUMMARY

In the neutral one-dimensional C$_{60}$-polymers, electronic structures change among direct-gap insulators and the metal, depending on the degree of conjugations. The high pressure experiments may be able to change conjugation conditions in the chain direction, and the electronic structure changes could be observed.

The one-dimensional C$_{60}$-polymer doped with one electron per one molecule is always a metal. The energy difference between the highest-occupied state and the lowest-unoccupied state of the neutral system becomes smaller upon doping owing to the polaron effects. When the one-dimensional C$_{60}$-polymer is doped with two electrons per one C$_{60}$, the system is insulating. When the conjugation in the direction of the polymer chain is smaller, it is a direct-gap insulator. The energy gap becomes indirect when the conjugation is stronger.

We have studied the antiferromagnetic phase of the C$_{60}$-polymers by microscopic calculations. The antiferromagnetism is well described by the present model. The formation of the magnetic order is closely related with the strong interactions rather than the Jahn-Teller mechanisms. We have compared with the photoemission studies to find that the new band around the Fermi energy of the AC$_{60}$ phase can be explained by the fact that a energy level intrudes extremely largely into the gap of the neutral system by the electron doping. This is another indication that the interaction effects among electrons play a crucial role in doped C$_{60}$-polymers.

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Figure captions

Fig. 1. The crystal structure of the one-dimensional C\textsubscript{60}-polymer. The labels, A-I, indicate carbon atoms whose charge densities are not equivalent due to the symmetry. The conjugations along four bonds, which connect carbon atoms with labels, 1-4, are controled by the parameter \( a \) in eq. (1).

Fig. 2. Excess electron distribution for the three conjugation conditions, (a) \( a = 0.5 \), (b) 0.8, and (c) 1.0. The white bars are for the case \( N_{\text{ex}} = 1 \) and the black bars are for \( N_{\text{ex}} = 2 \).

Fig. 3. Band structures of the C\textsubscript{60}-polymer of the case \( a = 0.5 \). The excess electron number per one C\textsubscript{60} is (a) 0, (b) 1, and (c) 2, respectively. In (a), the highest fully occupied band is named as “HOMO”, and the lowest empty band as “LUMO”. The lattice constant of the unit cell is taken as unity.

Fig. 4. Band structures of the C\textsubscript{60}-polymer of the case \( a = 0.8 \). The excess electron number per one C\textsubscript{60} is (a) 0, (b) 1, and (c) 2, respectively. The lattice constant of the unit cell is taken as unity.

Fig. 5. Band structures of the C\textsubscript{60}-polymer of the case \( a = 1.0 \). The excess electron number per one C\textsubscript{60} is (a) 0, (b) 1, and (c) 2, respectively. In (a), the highest fully occupied band is named as “HOMO”, and the lowest empty band as “LUMO”. The lattice constant of the unit cell is taken as unity.

Fig. 6. The variations of the energy gap plotted against \( a \). The cases \( N_{\text{ex}} = 0 \) and 2 are shown in (a) and (b), respectively. The white (black) squares indicate that the system is a direct gap insulator where there is a energy gap at \( k = 0 \) (\( \pi \)). The squares with the plus symbol mean that the system is an indirect gap insulator. The crosses are for metallic cases.

Fig. 7. The magnitude of the magnetization per one C\textsubscript{60} shown by changing \( a \) and \( U \). We use \( V = U/2 \) here. The closed and open squares are for \( a = 0.1 \) and 0.2, respectively. The closed and open circles are for \( a = 0.3 \) and 0.4.

Fig. 8. The phase diagram shown against \( a \) and \( U \). The region with “M” is the metallic phase, and the region with “AF” is the antiferromagnetic phase.

Fig. 9. The band structures of the neutral C\textsubscript{60}-polymer of the case \((a, U, V) = (0.3, 2.4t, 1.2t)\). The unit cell consists of one C\textsubscript{60}. The length of the unit cell is taken as unity. The highest fully occupied band is named as “HOMO”, and the lowest empty band as “LUMO”.

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Fig. 10. The band structures of the C$_{60}$-polymer doped with one electron per one C$_{60}$ of the case $(a, U, V) = (0.3, 2.4t, 1.2t)$. The unit cell consists of two C$_{60}$ molecules. The length of the unit cell is taken as unity. The highest fully occupied band is named as “HOMO”, and the lowest empty band as “LUMO”.