Density Waves in the Organic Metal α-(BEDT-TTF)$_2$KHg(SCN)$_4$

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Abstract

We have investigated possible spin and charge density waves in the organic metal α-(BEDT-TTF)$_2$KHg(SCN)$_4$. This system shows density wave like transition at $T = 8$ K, and the nature of the density wave is not clarified up to now. Using a realistic tight binding model and an inhomogeneous mean-field theory, we obtained several stable density wave states. Especially, spin density wave states (SDW) are stable only for large on-site Coulomb interaction $U \geq 400$ meV. Their spin moments are tiny and inhomogeneous even in the unit cell. Also charge density wave (CDW) appears simultaneously with the SDW.

Key words: organic conductors; BEDT-TTF; density waves

A series of organic conductors α-(BEDT-TTF)$_2$MHg(SCN)$_4$ (M=K, NH$_4$ and Rb) shows a variety of the ordered states at low temperature. Among this series, α-(BEDT-TTF)$_2$KHg(SCN)$_4$ is considered to become density wave state below $T = 8$K[1]. Recent electron spin resonance experiments (ESR) support of this picture [2–4]. But details of the density wave state are not yet known until now.

Therefore we have investigated possible stable density wave states of this material.

We use the band structure calculation of Campos et al.[5]. They derived transfer integrals between highest molecular orbitals (HOMOs) on the neighbor molecules. Then Hamiltonian is,

$$\mathcal{H} = -\sum_{\langle ij\rangle\sigma} t_{ij}\left(C^\dagger_{i\sigma}C_{i\sigma} + \text{h.c.}\right) + U\sum_i \hat{n}_{i\uparrow}\hat{n}_{i\downarrow}, \qquad (1)$$

where the tranfer integrals are defined in Fig. 2 and in Table II of Ref. [5] and U is an on-site Coulomb interaction.

We apply an inhomogeneous mean-field theory [6] to this Hamiltonian. In this theory, we consider a cluster of $L_x \times L_y$ unit cells with the periodic boundary condition. Then we define local mean-field order parameters

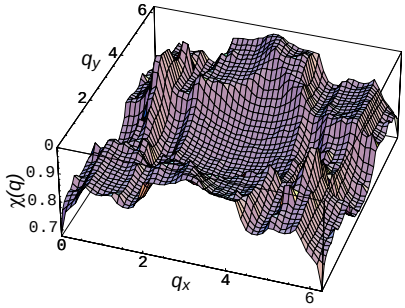

Fig. 1. Static susceptibility $\chi(\boldsymbol{q})$.

for each molecule as, $n_{i\sigma} = \left\langle C^\dagger_{i\sigma}C_{i\sigma}\right\rangle$, where $\langle\cdots\rangle$ is the thermal average using eigenstates following mean-field Hamiltonian,

$$\mathcal{H}_{\text{MF}} = -\sum_{\langle ij\rangle\sigma} t_{ij}\left(C^\dagger_{i\sigma}C_{i\sigma} + \text{h.c.}\right) + U\sum_{i\sigma}\left[n_{i\sigma}C^\dagger_{i-\sigma}C_{i-\sigma}\right] - U\sum_i n_{i\uparrow}n_{i\downarrow}. \qquad (2)$$

We solve these equations self-consistently.

First we have calculated a static susceptibility for the non-interacting system. In Fig.1, we show the wave number dependence of the susceptibility. The non-interacting energy $\epsilon_{\boldsymbol{k}}$ has two kinds of Fermi surfaces.

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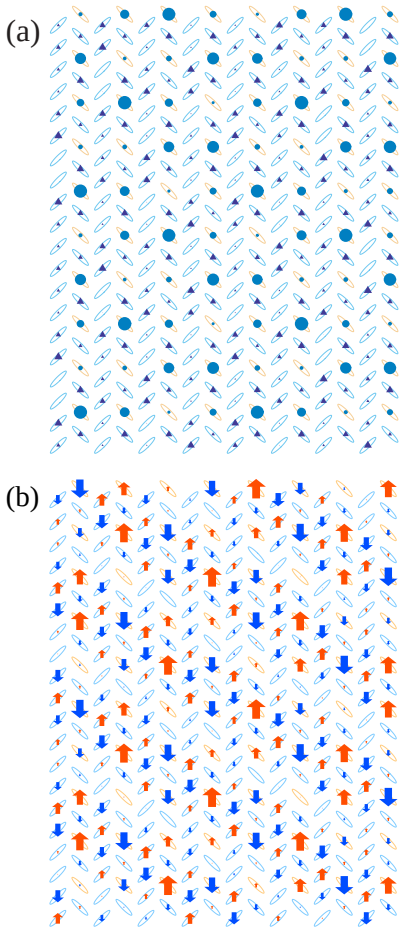

Fig. 2. A stable density wave state for $U = 432$ meV and $T = 6$ K. An oval represents a BEDT-TTF molecule. In (a), a disk (triangle) shows that number of electrons on a molecule is larger (smaller) than the average value 1.5, respectively. Maximum magnitude of the deviation of the electron number from 1.5 is 0.043. In (b), an arrow means the averaged spin on a molecule. Maximum magnitude of the spin is 0.21.

One is one-dimensional open orbit and another is two-dimensional closed orbit. The sharp peaks around $\boldsymbol{k} = (3\pi/4, 7\pi/5)$ and $\boldsymbol{k} = (5\pi/4, 3\pi/5)$ comes from the nesting of the one-dimensional Fermi surfaces.

From this bare susceptibility, we have searched for density waves with the characteristic wave vector $\boldsymbol{Q} = (3\pi/4, 7\pi/5)$. Therefore we have set $L_x = 8$ and $L_y = 10$. And we have started the self-consistent calculation form the initial state that has $\boldsymbol{Q}$. There are four molecules in a unit cell, therefore there are many possible density wave states with same characteristic vector. So, we have chosen several possible initial states and repeated the self-consistent calculation.

In Fig. 2, we show the stable density wave state for $U = 432$ meV and $T = 6$ K. Maximum magnitude of averaged moment is small. This spin density wave state accompany the variation of the charge density, although it is also small.

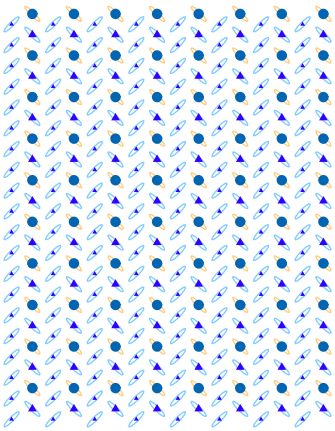

Fig. 3. A stable density wave state for $U = 324$meV and $T = 6$ K. The symbols are same as Fig. 2.

In Fig.3, we show the stable density wave state for $U = 324$ meV and $T = 6$ K. For $U < 400$ meV, magnetic moments disappear and only the inhomegeneity of the electron number remains. But the period of the variation is same as the original lattice. Also, as decreasing temperature, the magnitude of the charge density variation becomes small. Therefore this state may be considered as a normal state.

These results agree with the tiny magnetic moment of the density wave state. But for completeness, we must consider the possibility of the purely charge density wave state by including the electron-phonon interaction into the Hamiltonian. Also, analysis of the ESR experiment is a future problem.

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