Fermi Surface and Cyclotron Mass in (DMe-DCNQI)_{2}Cu System


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Abstract

The de Haas - van Alphen measurements of the undeuterated and partially deuterated (DMe-DCNQI)_{2}Cu have been carried out to investigate the Fermi surface. The results are in good agreement with the calculated band structure. There is no significant change in the Fermi surfaces and in the cyclotron masses between all the samples. No sizable mass enhancement of the conduction electrons is observed even for the deuterated samples.

1. INTRODUCTION

The novel molecular conductor containing Cu ions, (DMe-DCNQI)_{2}Cu [1-3], has attracted great interests due to the characteristic physical properties. One of the striking features in the (DMe-DCNQI)_{2}Cu system is the fact that a drastic metal-insulator (M-I) transition is induced by the substitution of the hydrogen with deuterium in the DMe-DCNQI molecules [4,5]. The similar M-I transition has been found for some alloy systems [6-9]. A metal-insulator-metal (M-I-M) reentrant transition is found in the partially deuterated system [5], and in the alloy of the undeuterated and the fully deuterated systems [9]. The M-I and M-I-M transitions induced by the deuteration have been understood in terms of the chemical pressure [4,5] and the P-T phase diagram of the deuterated system has been proposed in Fig. 1 [5].

In order to investigate the Fermi surface (FS), we have carried out the de Haas-van Alphen (dHvA) measurements for the single crystals of the undeuterated and partially deuterated (DMe-DCNQI)_{2}Cu. One undeuterated sample (h) and two selectively deuterated samples (d2 [1,1,0] and d4 [1,1,2]) were measured. Both of the deuterated samples show the M-I-M transition (Fig. 1).

2. RESULTS AND DISCUSSION

Typical dHvA oscillations for the h salt are presented in Fig. 2. The Fourier transform spectra are shown in the insets. The observed frequency is in the range from 350 T to about 15000 T for all the salts. For $\omega=26^\circ$, the three different oscillations ($E_2$, $E_5$ and $E_6$) are seen. Figure 3 presents the angular
Table I. The calculated band masses $m_b$ and the observed cyclotron mass $m_c$ of the dHvA oscillations in the undeuterated (h) and deuterated salts (d2 and d4) of (DMe-DCNQI)$_2$Cu.

| label | direction | $m_b$/m$_0$ | $m_c$/m$_0$ | $d_2$ | $d_4$
|-------|-----------|-------------|------------|-------|-------|
| a     | $\theta=0^\circ$ | 4.0 | 3.5 | 3.9 | 3.5 | 3.4 | 3.4 | 3.4 | 3.4 | 3.5 | 3.5
| $\beta$ | $\gamma$ | 3.6 | 3.9 | 3.4 | 3.4 | 3.4 | 3.4 | 3.4 | 3.4 | 3.4 | 3.4
| $\delta_1$ | $\omega=45^\circ$ | 4.3 | 3.9 | 3.9 | 3.9 | 3.9 | 3.9 | 3.9 | 3.9 | 3.9 | 3.9
| $\delta_2$ | $\omega=25^\circ$ | 6.2 | 6.5 | 6.5 | 6.5 | 6.5 | 6.5 | 6.5 | 6.5 | 6.5
| $\varepsilon_5$ | $\omega=25^\circ$ | 9.5 | 9.5 | 9.5 | 9.5 | 9.5 | 9.5 | 9.5 | 9.5 | 9.5
| $\varepsilon_6$ | $\omega=25^\circ$ | 8.0 | 8.0 | 8.0 | 8.0 | 8.0 | 8.0 | 8.0 | 8.0 | 8.0 | 8.0

*: not calculated  **: not measured

Figure 3. Angular dependences of the low dHvA frequencies of the undeuterated (h) and deuterated (DMe-DCNQI)$_2$Cu salts (d2 and d4). The solid lines show the calculated results.

dependences of the low frequencies ($F < 3000$ T). We find seven different oscillations ($a, \beta, \gamma, \delta_1 - \delta_4$) with low frequencies ($F < 3000$ T). Many high frequency ($F > 3000$ T) oscillations ($e_1, e_2, \ldots$) are observable in the (001) plane.

Taking account of the contribution of the LUMO of the DCNQI molecules and the $d_{xy}$ orbital of the Cu ions, the tight binding band calculation was carried out [2,10]. The band structure consists of the two energy bands; the 1D energy bands having the $p_{xy}$ character, and the 3D energy band arising mainly from the $d_{xy}$ orbital. The solid lines in Fig. 3 show the calculated frequencies corresponding to the extremal cross sectional areas of the 3D FS. The calculated results are in good agreement with the experimental results. Therefore, we conclude that the 3D FS exists, and that the seven low frequency oscillations ($a, \beta, \gamma, \delta_1 - \delta_4$) arise only from the 3D FS.

The high frequency oscillations ($F > 3000$ T) are observed in the (001) plane. On a basis of the calculated band structure, the high frequency oscillations are concluded to come from the large cyclotron orbits on both the 1D and 3D FSs. They are observable only in limited angle ranges, which is ascribed to the characteristic FS structure. The details will be published elsewhere [11]. Any differences are not appreciable in the frequencies between all the salts within the experimental error of about ±5%. Therefore, both the 1D and 3D FSs for all the salts are nearly the same in shape and size.

The observed cyclotron mass $m_c$ and the calculated band mass $m_b$ are listed in Table I. The cyclotron mass $m_c$ is given by $m_c=m_0(1+k_e+c)(1+k_p-c)$, where $k_e$ and $k_p$ are the mass enhancement factors due to electron-electron interaction and electron-phonon interaction, respectively. The mass $m_c$ of each oscillation is found to be the same for all the salts within the experimental error of about ±10%, and $m_c$ is in good agreement with the band mass $m_b$. The agreement leads us to conclude that significant difference in the mass enhancement factors due to the many body effects is absent on both the 1D and 3D FSs in all the salts. Recently, the magnetic susceptibility $\chi_D$ of the d2 salt was carefully measured [12]. The results clearly show that there is no sizable enhancement of the Pauli paramagnetism in the reentrant metallic phase, which is consistent with the dHvA experimental results.

Nishio et al. report that the electronic specific heat coefficient $\gamma$ for the alloy system (DMe$_2$MeBr$_2$-DCNQI)$_2$Cu increases with increasing $x$, and is steeply enhanced near the insulating phase [6]. The similar behavior is observed for the alloy of the undeuterated and fully deuterated systems [13]. The behavior of $\gamma$ is consistent with the enhancement of the $\gamma^2$ dependence in the resistance at low temperatures [5]. Those results suggest that the mass of the conduction electrons is enhanced near the insulating phase. The possibility of the mass enhancement in this system has been predicted theoretically, considering strong electron correlation or strong electron phonon interaction [14,15]. However, even in the deuterated salts, such mass enhancement has not been observed by the dHvA experiments.

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