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Coexistence of charge order and antiferromagnetism in (TMTTF)₂SbF₆: NMR study

K. Nomura*, M. Yamamoto, N. Matsunaga, S. Hirose, N. Shimohara, T. Satoh, T. Isome, Y. Liu and A. Kawamoto

Department of Physics, Hokkaido University, Sapporo 060-0810, Japan

Abstract

The electronic state of (TMTTF)₂SbF₆ was investigated by the ¹H and ¹³C NMR measurements. The temperature dependence of T_1^{-1} in ¹H NMR shows a sharp peak associated with the antiferromagnetic transition at $T_{AF}=6$ K. The temperature dependence of T_1^{-1} is described by the power law $T^{2.4}$ below T_{AF} . This suggests the nodal gapless spin wave excitation in antiferromagnetic phase. In ¹³C NMR, two sharp peaks at high temperature region, associated with the inner and the outer carbon sites in TMTTF dimer, split into four peaks below 150 K. It indicates that the charge disproportionation occurs. The degree of charge disproportionation $\Delta\rho$ is estimated as $(0.25\pm0.09)e$ from the chemical shift difference. This value of $\Delta\rho$ is consistent with that obtained from the infrared spectroscopy. In the antiferromagnetic state (AFI), the observed line shape is well fitted by eight Lorentzian peaks. This suggests that the charge order with the same degree still remains in the AF state. From the line assignment, the AF staggered spin amplitude is obtained as $0.70\mu_B$ and $0.24\mu_B$ at the charge rich and the poor sites, respectively. These values corresponding to almost $1\mu_B$ per dimer are quite different from $0.11\mu_B$ of another AF (AFII) state in (TMTTF)₂Br with effective higher pressure. As a result, it is understood that the antiferromagnetic staggered spin order is stabilized on the CO state in the AFI phase of (TMTTF)₂SbF₆.

Keywords: Charge order ; Antiferromagnetism ; NMR ; Quasi-one-dimensional conductor

1. Introduction

The family of organic charge transfer salts (TMTCF)₂X, where C=T or S and X=SbF₆, PF₆, ClO₄ etc., show a variety of electronic properties depending on the effective pressure, which is given by both the physical and chemical pressure. The ground state of (TMTSF)₂ClO₄ located at the highest pressure region is the superconducting phase at ambient pressure. With decreasing pressure, its ground state varies to spin-density wave (SDW), antiferromagnetic (AFII), spin-Peierls (sP) and another antiferromagnetic (AFI) phases, successively. These phases are realized in a

common quasi-one dimensional electron band formed on the array of TMTCF molecules against the electron correlation and the strength of the one-dimensional character in the generic *P-T* phase diagram [1]. In the lower pressure region, the charge order (CO) phase appears widely in the high temperature side of the ground state. This CO phase is observed commonly in (TMTTF)₂MF₆ (*M*=F, As, Sb and Ta), although the ground state is the sP for PF₆ or AsF₆ and the AFI for SbF₆ or TaF₆. Accordingly the clarification of the CO phase and its relation with the ground state are very important for the understanding of the properties in these salts with the unified picture.

* Corresponding author. Tel.: +81-11-706-4430; fax: +81-11-706-4926; e-mail: knmr@phys.sci.hokudai.ac.jp.

The CO in this system was clearly demonstrated with the observation of splitting of ^{13}C NMR line [2]. The degree of charge disproportionation $\Delta\rho = \rho_{\text{rich}} - \rho_{\text{poor}}$ was initially estimated from the ratio of the spin-lattice relaxation rate T_1^{-1} between splitted lines. The CO was also observed as the spectral splitting due to the molecular vibration in the optical measurement. The value of $\Delta\rho$ was also obtained from the interval of splitted lines. However, there is inconsistency between the estimated values of $\Delta\rho$ for each salt depending on the measuring method. For example, for the CO phase $(\text{TMTTF})_2\text{AsF}_6$ with the sP ground state $\Delta\rho$ was estimated as 0.50e and 0.33e from the ratio of T_1^{-1} [3,4], while 0.21e and 0.16e were obtained by the infrared spectroscopy of two independent groups [5,6]. In addition, the almost same $\Delta\rho$ in the sP phase was reported in the optical investigation [6], whereas the previous NMR study suggested the disappearance of the charge disproportionation in the sP phase [4]. For $(\text{TMTTF})_2\text{SbF}_6$ with the AF ground state $\Delta\rho$ was estimated as 0.50e and 0.55e by the NMR T_1^{-1} [3,7] and 0.29e by the infrared spectroscopy [8]. In each case the estimated $\Delta\rho$ by T_1^{-1} is much larger than that by the optical measurement. So these discrepancies should be settled.

Recent paper for the optical measurements [6] in $(\text{TMTTF})_2\text{AsF}_6$ successfully explained these discrepancies, taking into account the commensurate AF fluctuation which play the dominant role in the NMR T_1^{-1} . Our recent ^{13}C NMR measurement for $(\text{TMTTF})_2\text{AsF}_6$ [9] revealed the smaller value $\Delta\rho$ for the CO phase by the analysis of the chemical shift free from the AF fluctuation and the coexistence of the CO with the sP phase at the same time, consistent with the optical measurement [6]. These suggest that the estimation of $\Delta\rho$ by T_1^{-1} is not correct in these systems because of the AF fluctuation.

The AFI phase of $(\text{TMTTF})_2\text{SbF}_6$ was also investigated with the ^{13}C NMR measurement [7]. The four peaks were observed in the AFI phase. It suggested at a glance the disappearance of the charge disproportionation in the AFI phase, although the authors didn't discuss it. In the sP phase it was confirmed from the recent NMR [9] and optical measurement [6] that the CO still remains in the sP phase. So the coexistence of the CO and AF order is naturally expected even in the AFI phase adjacent to the sP phase. The theoretical calculation for the quarter-filled one dimensional electronic band with dimerization by the extended Hubbard Hamiltonian predicted the coexistence of the CO and the AF for an appropriate nearest-neighbour Coulomb repulsion [10]. Accordingly, the clarification of the coexistence of the

CO and AF order in the AFI phase becomes quite important for the total understanding of this quasi-one dimensional system. In addition the difference between the AFI and AFII phase in $(\text{TMTTF})_2\text{Br}$ should be elucidated.

We performed the ^1H and ^{13}C NMR measurement [11] on a single crystal of $(\text{TMTTF})_2\text{SbF}_6$ with single site of the central carbon ^{13}C -enriched TMTTF molecules in order to clarify the electronic properties mentioned above. Here, we present the experimental results and discuss its microscopic structure of the charge order and antiferromagnetic state. We also report the coexistence of the CO and AF in the AFI phase.

2. Experimental

Single crystal of $(\text{TMTTF})_2\text{SbF}_6$ was synthesized by the standard electrochemical method with use of single site of the central carbon ^{13}C -enriched TMTTF molecules. It assures the absence of the additional splitting of ^{13}C NMR line by the Pake doublet. In ^1H NMR the spin-lattice relaxation rate T_1^{-1} was mainly studied in the field of 0.87T. In ^{13}C -NMR, NMR line shape was investigated in the field of 9.4T along the c^* axis, in which the chemical shift is almost the largest and the Knight shift is small. The NMR spectrum was obtained by the Fourier transformation of the spin-echo signal with π - $\pi/2$ pulse sequences. For the antiferromagnetic phase, the total line shape was obtained by the sum of the spectral measurement with NMR frequencies with 25 kHz interval between 100.590 MHz and 100.840 MHz because of the large width exceeding the range covered by one frequency of H_1 .

3. Results and Discussion

In ^1H NMR, the spin lattice relaxation rate T_1^{-1} was investigated. The recovery of magnetization after the saturation comb-pulse is well described by a single exponential curve in all temperatures. The temperature dependence of T_1^{-1} with the static magnetic field along the c^* axis is shown in Fig. 1. The sharp peak observed at 6.25 K is associated with the increasing of low frequency fluctuation due to the antiferromagnetic transition at T_{AF} . We could not observe any anomaly associated with the charge order transition or other ones.

We investigated the angular dependence of T_1^{-1} with the field in the c^*-b' plane in the AF phase. It shows a abrupt change suggesting the spin flop

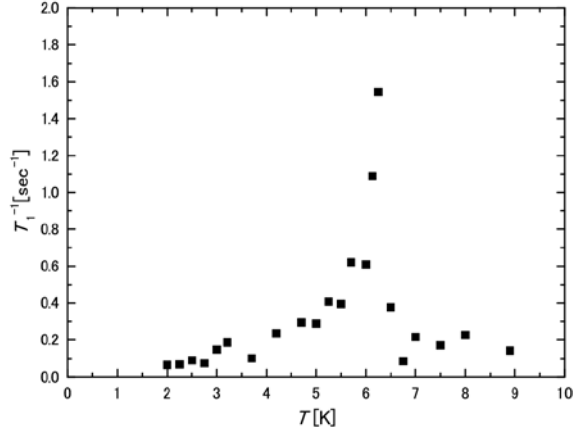


Fig. 1. Temperature dependence of ^1H NMR T_1^{-1} .

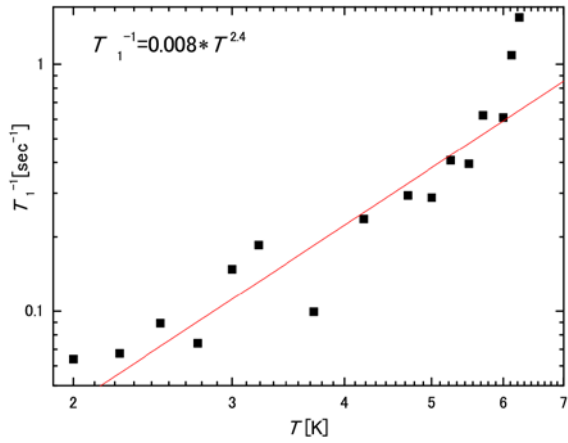


Fig. 2. Temperature dependence of T_1^{-1} below T_{AF} . The solid line represents the power law $\propto T^{2.4}$.

similarly to the incommensurate SDW phase in $(\text{TMTSF})_2\text{PF}_6$ [12]. The observed angular dependence is consistent with the previous report [13], where the spin easy and intermediate axes are the b' and the c^* , respectively, and the spin-flop field $H_{\text{SF}}=0.33$ T. Below T_{AF} , T_1^{-1} shows a sharp decrease with decreasing the temperature. The temperature dependence is described not by the activation type function but by the power law $\propto T^n$ with $n \approx 2.4$ as shown in Fig. 2. This behavior suggests that the gapless spin wave excitation dominate the spin lattice relaxation below T_{AF} . We didn't observe any additional structure in AF phase like the sub-phase transition the incommensurate SDW phase of $(\text{TMTSF})_2\text{X}$ [14]. These suggest that the AFI is a usual anti-ferromagnetic phase.

The ^{13}C NMR line shapes obtained from the FT of spin-echo signal are shown in Fig. 3 for several temperatures above T_{AF} . The magnetic field of 9.4 T is

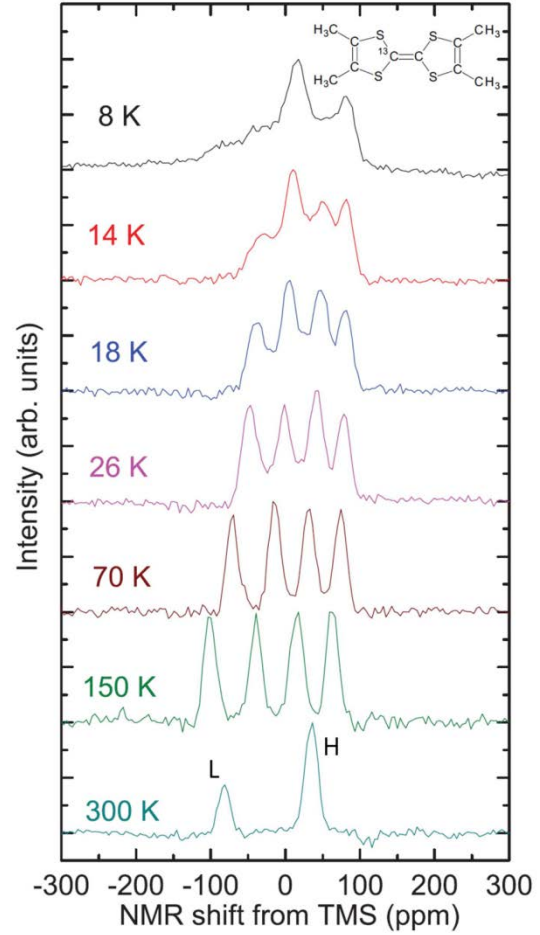


Fig. 3. ^{13}C NMR spectra in the paramagnetic state. The molecular structure of TMTTF of single site of the central $\text{C}=\text{C}$ carbons site substituted with ^{13}C is shown in the top right corner.

applied along the intermediate c^* axis. As shown in the top right corner, we observed the signal from a single site enriched ^{13}C of TMTTF. At room temperature two sharp peaks are observed at -80 ppm and 40 ppm. These are associated with the inner and the outer carbons non-equivalent in the crystal free from the Pake doublet. In order to specify each peak we need detailed angular dependence with the magnetic field direction. However, it doesn't affect the analysis of the CO and AF state. Accordingly, we proceed with the discussion without specifying each peak to the inner or the outer sites. So we call the lower frequency (L) peak and higher frequency (H) one hereafter. Below 150 K these two peaks split into four sharp peaks. This splitting of lines is caused by the charge disproportionation in the CO phase. This behaviour is qualitatively consistent with the previous ^{13}C NMR [7], although each line doesn't correspond in the same order because of the different

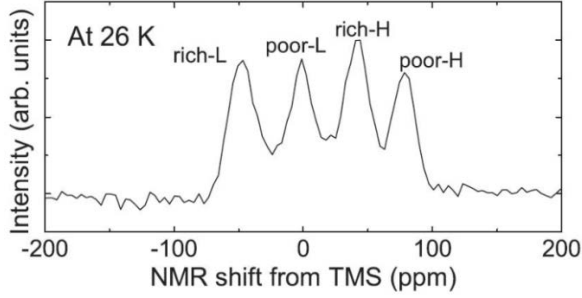


Fig. 4. ^{13}C NMR spectrum at 26 K in the charge order phase. Each peak is assigned to four non-equivalent sites.

magnetic field direction. This indicates the charge disproportionation with the appearance of four non-equivalent sites, i.e. charge rich-inner, rich-outer, poor-inner and poor-outer. With decreasing the temperature the peak positions varies toward the higher frequency, i.e. lower shift. This behaviour is consistent with the decreasing spin susceptibility. However we can see that the intervals of four peaks are not much changed.

For further discussion we need to assign the charge rich site and the poor one. We can deduce the origin of NMR shift as around 200 ppm from the previous study of similar salts [9,15]. So for the spectra at 26 K as shown in Fig. 4, we can naturally assign the peak at -50 ppm, which have the largest shift, as the charge rich site and the peak at 78 ppm as the poor site. With decreasing the temperature, both peaks at around -50 ppm and 43 ppm at 26 K show a slight broadening. We confirmed with measuring the spin-echo amplitude against the pulse interval that this broadening is not the homogeneous one related to the spin-spin relaxation rate T_2^{-1} but the inhomogeneous one. The widths of these two peaks show the same temperature dependence and are assigned as the inner and the outer sites of identical charge molecule, i.e. the charge rich molecule. As a result, four peaks at -50 ppm, 0 ppm, 43 ppm and 78 ppm at 26 K are assigned as the charge-rich-L, the charge-poor-L, the charge-rich-H and the charge-poor-L sites, respectively. We can recognize the broadened peaks of charge-rich site even at 8 K just above T_{AF} . In the previous NMR experiment [7], only two peaks were observed below 20 K. It is understood that the much broadened signal is missing in the previous experiment according to the magnetic field direction in which the Knight shift is larger. The origin of this broadening is not elucidated. We deduce the spatial distribution of the magnetization coupled with large antiferromagnetic fluctuation just above the AF transition, because such a broadening is not observed in the spectra of the

optical measurement for the corresponding AFII phase of $(\text{TMTTF})_2\text{TaF}_6$ [16] and the charge disproportionation itself looks rather uniform in space.

As previously discussed [4], in the system with the large antiferromagnetic fluctuation, the spin lattice relaxation rate T_1^{-1} at each site is not only by the local susceptibility but also by the antiferromagnetic fluctuation commensurate with the underlying lattice and the CO pattern. As a result, the ratio of T_1^{-1} at each site doesn't reflect correctly the ratio of charge. On the other hand, the NMR shift is not affected by such a dynamical fluctuation as well as the optical measurement. So we can estimate the charge disproportionation correctly from the NMR shift. The NMR shift δ is given by the sum of the Knight shift K and the chemical shift σ , which are related to the spin susceptibility χ and the orbital part of electronic charge, respectively as,

$$\delta = K + \sigma = \alpha\chi + \sigma. \quad (1)$$

The hyper-fine coupling of ^{13}C α between the local field and the spin polarization in organic salts with π -orbital is given not only by the on-site molecule but also by the neighbouring molecules because of the relatively large dipolar coupling. On the other hand, the chemical shift is determined dominantly by the on-site molecule. Therefore the analysis of the chemical shift is straightforward and precise for the estimation of the degree of the charge disproportionation $\Delta\rho$. In the metallic phase two values of δ corresponds to the inner and outer sites with different α . In the CO state both the local χ and σ are doubled due to the charge disproportionation. We can decouple the difference of chemical shift $\Delta\sigma = |\sigma_{\text{rich}} - \sigma_{\text{poor}}|$ from four peak shifts together with two peak values at the metallic phase without any specified α or χ values. We obtain $\Delta\sigma = |\sigma_{\text{rich}} - \sigma_{\text{poor}}|$ as 25 ± 9 ppm between 20 K and 150 K. It is understood that the temperature dependence of each peak position in this temperature range is not given by the charge disproportionation but the static spin susceptibility. In the spin-Peierls state of $(\text{TMTTF})_2\text{AsF}_6$, the ratio of the charge disproportionation $\Delta\rho$ to the chemical shift difference $\Delta\sigma$ was determined as $\Delta\rho/\Delta\sigma = 0.01e$ [ppm $^{-1}$] [9]. With use of this ratio we obtain $\Delta\rho = (0.25 \pm 9.09)e$ for the CO phase of $(\text{TMTTF})_2\text{SbF}_6$. This value is almost same as that estimated from the infrared spectroscopy [8]. On the other hand, larger values for $\Delta\rho$ were estimated from NMR T_1^{-1} measurements [3,7]. The discrepancy is well explained by considering the contribution of the commensurate AF fluctuation in

spin-lattice relaxation. In addition, the value of $\Delta\rho$ is almost constant below 150 K. The rapid growth of charge disproportionation below T_{CO} may suggest the sign of the first order-like phase transition. It should be noted [16] that the value of $\Delta\rho(0)$ is proportional to the transition temperature T_{CO} , together with those in other salts, in spite of somewhat different temperature dependence.

In the low temperature, the additional line splitting and broadening were observed. Figure 5 show the NMR spectra obtained at 5 K. The magnetic field was also applied along the intermediate c^* axis to avoid the spin flop. Owing to the broad spectra we cannot cover the total line by one H_1 pulse. We obtained the total line by the sum of the spectral measurement with NMR frequencies with 25 kHz interval between 100.590 MHz and 100.840 MHz. The inset in Fig. 5 shows the line obtained by one frequency. There only four peaks are recognized and the broad lines in both sides are nearly missing. This line by one frequency is like that in the previous measurement [7]. The structure of full line is associated with the appearance of staggered moment in the AF phase. In this NMR line at least more than six peaks are recognized and indicate clearly the remaining CO even in the AF state. We try to fit the observed NMR line by several Lorentzian curves. As shown in Fig. 5, the NMR spectra are successfully fitted by the eight Lorentzian curves (represented by dotted lines). In the figure the solid line represents the total sum and well reproduces the observed line. Because of the observed inhomogeneous broadening of line at the charge rich site just above T_{AF} , the broad peaks at around 100.642 MHz and 100.742 MHz can be assigned as the charge

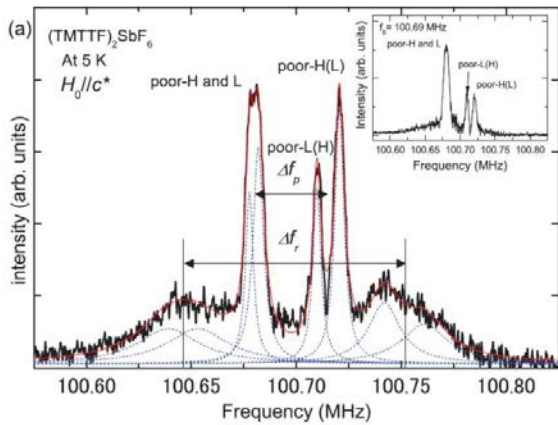


Fig. 5. ^{13}C NMR spectrum at 5 K in the AFII phase. The total line was obtained by the sum of the spectral measurement with NMR frequencies with 25 kHz interval between 100.590 MHz and 100.840 MHz. The total line is well fitted by the eight Lorentzian curves represented by dotted lines. The inset shows the line obtained by one frequency.

rich site. The sharp four peaks at 100.678 MHz, 100.682 MHz, 100.7110 MHz and 100.721 MHz correspond to the charge poor site. As a result, broad peaks at both sides are decomposed to pair of two peaks at (100.639 MHz and 100.653 MHz) and (100.742 MHz and 100.761 MHz). These assigned spectral peaks mean that each peak at the CO phase split into two peaks due to the appearance of the staggered spin moment in the AF phase.

We can estimate the spin amplitude from the interval of peaks splitted by the staggered spin moment. The averaged interval for the inner and outer sites for the charge poor site and the charge rich site, Δf_p and Δf_r , are obtained as 35 kHz and 105 kHz, respectively. Therefore, the ratio of staggered spin moment at the charge rich site to the charge poor site is about 3. This ratio is roughly equal to the observed ratio of T_1^{-1} in the CO phase in the previous measurement [7]. If we assume that the commensurate AF fluctuation with the same ratio exists above T_{AF} , it assures our interpretation for the wrong estimation of the charge disproportionation $\Delta\rho$ by the ratio of T_1^{-1} .

To obtain the absolute value of spin amplitude at each site, we need to know the hyperfine coupling constant. For the metallic phase of $(\text{TMTTF})_2\text{Br}$ the hyperfine coupling constant per a dimer of TMTTF against the magnetic field along the c^* axis was obtained as $A_{b,c^*}^H = 0.03$ [kOe/ μ_B] and $A_{b,c^*}^L = 0.04$ [kOe/ μ_B] for H and L sites. Considering the relatively larger hyperfine coupling for L site, two inside peaks of the poor sites (100.681 MHz and 100.710 MHz) and two inside peaks of the rich site (100.653 MHz and 100.742 MHz) are assigned as the H-site and two outside peaks of the poor sites (100.678 MHz and 100.721 MHz) and two outside peaks of the rich site (100.639 MHz and 100.761 MHz) are assigned as the L-site, respectively. As a result, we can obtain each interval of peaks as $\Delta f_p^H = 0.029$ MHz, $\Delta f_r^H = 0.089$ MHz, $\Delta f_p^L = 0.043$ MHz and $\Delta f_r^L = 0.122$ MHz. With use of the above hyperfine coupling constants, the magnetic moment at H-site is obtained as $0.69\mu_B$ and $0.23\mu_B$ at charge rich and poor sites. Correspondingly those for L-site are $0.71\mu_B$ and $0.25\mu_B$, respectively. We obtain $0.70\mu_B$ and $0.24\mu_B$ as averaged values. The spin moment is totally $0.94\mu_B$ for a dimer and is roughly equal to a full moment.

From the line assignment discussed above, we also obtain the similar amount of line splitting by the charge disproportionation in the AF phase as in the CO phase. Although the precise estimation is hard because of large hyperfine field due to the AF staggered moment, we conclude that the charge disproportionation remains even in the AF phase. It is

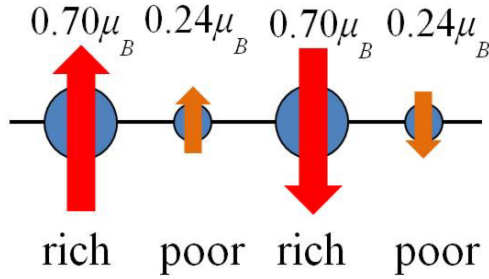


Fig. 6. Staggered spin structure in the AFII phase. The AF order is stabilized on the CO pattern.

consistent with the recent optical measurement in the AFI phase of $(\text{TMTSF})_2\text{TaF}_6$ [16], where the charge disproportionation $\Delta\rho$ doesn't change from the CO phase to the AFI phase. Accordingly, it is understood that the antiferromagnetic order is stabilized on the CO pattern, although the spin is not rigorously proportional to the charge at each site. The spatial spin structure is expressed like Fig. 6.

For the occurrence of the CO state, the quarter-filled one dimensional electronic band with dimerization was investigated with the extended Hubbard Hamiltonian with the nearest-neighbour Coulomb interaction V theoretically [10]. It predicts the coexistence of the CO and AF state with the value of V exceeding the threshold V_c . Both the charge disproportionation $\Delta\rho=0.25$ and the corresponding ratio of spin moment 3:1 are calculated with $V/t_2=0.8$ and other appropriate parameters. The observed AF state stabilized on the CO state is well understood in such a mechanism. The antiferromagnetic order doesn't compete with the charge order.

The determined spin structure ($(\uparrow\uparrow-\uparrow\uparrow)-(\downarrow\downarrow-\downarrow\downarrow)$) (where $\uparrow\uparrow$ and $\downarrow\downarrow$ denote the staggered moment of the charge rich site, $\uparrow$ and $\downarrow$ denote the staggered moment of charge poor site and $[-]$ denotes a dimer of TMTTF molecules) along the stacking chain, shown in Fig. 6, is very different from the spin structure in the AFII in $(\text{TMTTF})_2\text{Br}$. In the latter the spin structure ($\uparrow-0-\downarrow-0$) with a shrunk spin $0.11\mu_B$. The AFII phase is rather considered as the commensurate SDW appearing adjacent to the incommensurate SDW as discussed initially. The AFI phase should be recognized as a standard antiferromagnetic phase stabilized on the charge ordered state.

4. Conclusion

We have investigated the electronic state of $(\text{TMTSF})_2\text{SbF}_6$ with the ^1H and ^{13}C NMR. In the ^1H NMR the spin-lattice relaxation rate T_1^{-1} was mainly

investigated. The temperature dependence of T_1^{-1} well characterizes the AF transition and the power law behaviour T^n with $n\approx 2.4$ observed below T_{AF} reflects the gapless spin wave excitation. In the ^{13}C NMR, the NMR line shape was investigated in both the CO and the AFI phase. Two sharp peaks associated with non-equivalent inner and outer carbon sites in TMTTF dimer, observed at high temperature region, split into four peaks below 150 K. It indicates that the charge disproportionation between dimerized molecules occurs. The degree of charge disproportionation $\Delta\rho$ is estimated as $(0.25\pm 0.09)e$ from the chemical shift difference. This value of $\Delta\rho$ is consistent with that obtained one from the infrared spectroscopy. The wrong estimation of $\Delta\rho$ from the previous T_1^{-1} studies was discussed with the commensurate AF fluctuation in the CO phase. In the AF (AFI) state, the additional peak broadening and splitting were observed. Total line shape is well fitted by eight Lorentzian peaks. This means that the charge order with same degree still remains in the AF state. After the line assignment, we obtain the AF spin amplitude as $0.70\mu_B$ and $0.24\mu_B$ at the charge rich and the poor sites, respectively. These values correspond to almost $1\mu_B$ per dimer which is very different from $0.11\mu_B$ of another AF (AFII) state in $(\text{TMTTF})_2\text{Br}$ with effective higher pressure. It is found that the antiferromagnetic staggered spin order is stabilized on the charge disproportionation remaining even in the AFI phase.

Acknowledgments

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