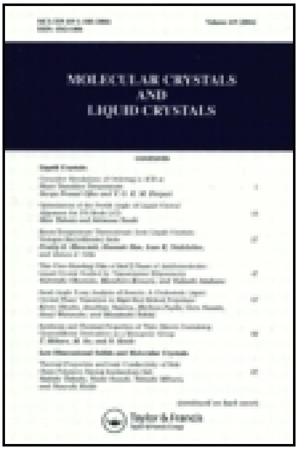

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NMR-Studies of Field-Induced Phase in $(TMTSF)_2ClO_4$

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NMR-STUDIES OF FIELD-INDUCED PHASE IN $(TMTSF)_2ClO_4$

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Abstract ^{77}Se-NMR measurement on a single crystal of $(TMTSF)_2ClO_4$ is revealed to be a sensitive probe of the field-induced magnetic phase transition. A recent trial to determine the magnetic phase boundary below 1 K is described.

The new phase transition induced by strong magnetic field in the organic superconductor, $(TMTSF)_2X$ family, is one of the attracting topics in the studies of low temperature electronic properties of this material. Accumulated experimental evidences up to date have revealed important features of the field-induced phase: i) This phase is of magnetic and semi-metallic nature (very probably a SDW state).[1] ii) Only the applied magnetic field component parallel to the crystalline c*-axis is effective to the transition (suggesting an orbital effect).[2] iii) Succesive phase transition as increasing the field strength are observed.[3]

Recently, several theoretical works on a mechanism of this transition have been presented.[4] They have succeeded to reproduce these characteristic features, at least qualitatively, by considering a SDW transition (nesting of a 2D Fermi surface) interplaying with quantization of electron orbital motions in a strong field. For the sake of getting further insight into the field-induced transition from these points of view, investigations of magnetic properties should be crucial. As a first step of this course, we tried to determine the 'magnetic' phase boundary in a temperature-field (T-H) phase diagram of $(TMTSF)_2ClO_4$.

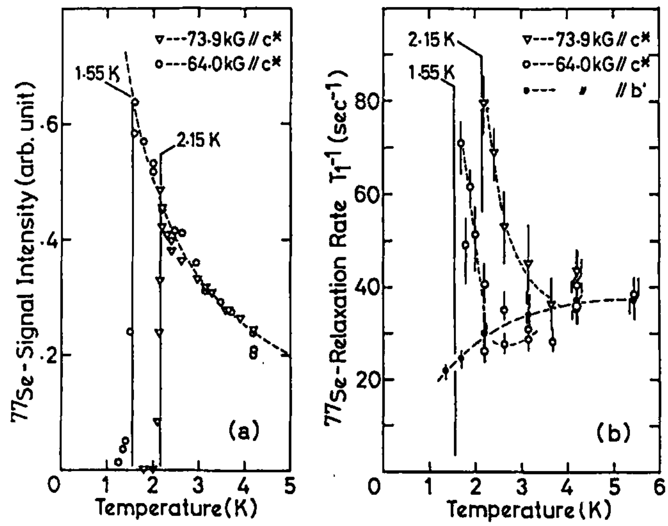

FIGURE 1 Temperature dependence of ^{77}Se-NMR signal intensity (a) and relaxation rate (b), for fields of 7.39 T and 6.40 T parallel to c*-axis.

As demonstrated in the previous work[1], ^{77}Se-NMR measurement on a single crystal is a sensitive probe to detect a magnetic transition. In a magnetic state, couplings, of dipolar and/or of hyperfine origin, between the electronic magnetic moments and nuclear spins produces a strong local field at nuclear sites. Thus, an onset of local field at transition can be easily detected by a drastic change in the NMR lineshape and an anomaly of NMR relaxation rate. Figure 1 shows the results at applied fields of 7.39 T and 6.4 T (parallel to the c*-axis). The transition temperatures were determined as 2.25 K and 1.55 K, respectively. The purpose of the present measurements was to extrapolate this method to the lower temperature side, where Hall effect anomalies were observed.

Experimental apparatus consists of a 6 T-superconducting magnet, a liquid-^{3}He cryostat, a home-made pulsed NMR spectrometer and signal averaging system. We observed echo signal following the usual $\pi/2$-π pulse pairs from ^{77}Se nuclei in a single crystal of $(TMTSF)_2ClO_4$ (Relaxed state) with a size of 0.3 × 0.3 × 3 mm^3. The crystal was carefully cooled down to achieve the so-called Relaxed state. No sign of quenching effect was observed, as far as ^1H and ^{77}Se- NMR properties were concerned.

We measured the temperature dependence of the signal intensity at fields of 4.9 T and 5.9 T, down to 0.53 K (our experimental limit). However, we failed to see any anomaly in the measured temperature region: No magnetic transition was observed!

A surprising feature of this result is obvious when one compares it with the published result of Hall effect measurement.[3] The solid lines in Figure 2 indicate the boundaries where the Hall coefficient has step-like anomalies, between which it is nearly constant in field strength. The broken straight lines indicate the region of the present investigation. (The circles are the previous NMR-results.[1])

If these results are correct, the region II in Figure 2, at least, should be nonmagnetic, so that the above-mentioned theoretical models should be seriously modified. However, we do not claim this here, since several experimental ambiguities, as follows, cannot be ruled out at present:

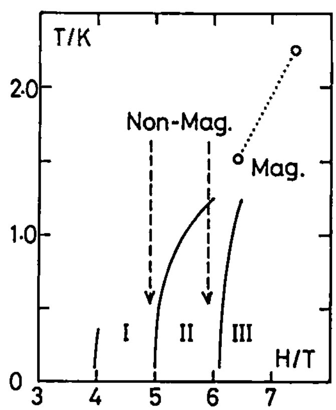

FIGURE 2 T-H phase diagram of $(TMTSF)_2ClO_4$ (Relaxed-state), based on the results of NMR and Hall effect measurements

i) Sample mis-alignment: If the applied external field was not exactly parallel to the c*-axis, the effective field component should be smaller than indicated.

ii) Sample dependence: The measurements were only for one crystal which was not the same that used at higher fields.

iii) Partial quenching effect (?): We have no experimental information about the strong field effect when the cooling rate is not sufficiently slow. (This may not be the case, since a rapid cooling tends to stabilize the SDW state even at low fields.[5])

Further investigations on these possibilities are going on, so that we will postpone concluding until a later publication.

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