The superstructure wave vector in the low-temperature electronic state of the organic conductor alpha -(BEDT-TTF)$_2$RbHg(SCN)$_4$ determined from angular magnetoresistance oscillations

LETTER TO THE EDITOR

The superstructure wave vector in the low-temperature electronic state of the organic conductor α-(BEDT-TTF)₂RbHg(SCN)₄ determined from angular magnetoresistance oscillations

M V Kartsovnik†¶, H Ito†, T Ishiguro†, H Mori‡, T Mori§, G Saito† and S Tanaka‡

† Faculty of Science, Kyoto University, Sakyo-ku, Kyoto 606-01, Japan
‡ International Superconductivity Technology Centre, Shinonome, Tokyo 135, Japan
§ Faculty of Engineering, Tokyo Institute of Technology, Tokyo 152, Japan

Abstract. Angular oscillations of magnetoresistance of a layered organic metal α-(BEDT-TTF)₂MHg(SCN)₄ with M=Rb have been studied at temperatures below the electronic phase transition. The derived superstructure is found to be very similar, both qualitatively and quantitatively, to that reported earlier for the salt with M=Tl. A minor series of oscillations is observed, which is the mirror reflection of the main oscillations against the $(a, 2b + c)$-plane. The data obtained for M=K reveal significant quantitative differences from the Rb and Tl compounds.

During the past few years, amongst the most attractive organic metals have been the compounds of the α-(BEDT-TTF)₂MHg(SCN)₄ family, which uniquely combine one-, two- and three-dimensional properties, their Fermi surface (FS) consisting of both slightly warped cylinders and open sheets as predicted by a band-structure calculation [1]. The compounds with M=K, Tl and Rb (hereafter referred to as K-SCN, Tl-SCN and Rb-SCN, respectively) undergo a phase transition at $T_p \simeq 10$ K into a state characterized by anisotropic magnetic susceptibility [2] and a number of striking magnetoresistance (MR) anomalies [3].

Strong periodic oscillations of semiclassical MR have been found in the K-SCN and Tl-SCN salts below T_p in a magnetic field rotating in various planes perpendicular to the highly conducting plane of the BEDT-TTF layers (i.e. the crystallographic ac-plane) [4,5]. A detailed analysis of these angular MR oscillations (AMRO) in Tl-SCN [5,6] has revealed the existence of a specific plane in the electronic system, which does not coincide with any basic plane of the reciprocal lattice. To understand this result, it was proposed that a new periodicity arises in the low-temperature (LT) state and, based on the AMRO positions, the corresponding superstructure vector Q has been suggested [6]. Since the Q_a component was found to be very close to the predicted distance between the open FS sheets, it was naturally supposed that the phase transition leads to a Peierls-like nesting of the one-dimensional band with the formation of a spin- or charge-density wave.

¶ Permanent address: Institute of Solid State Physics, Russian Academy of Science, Chernogolovka, Moscow District 142432, Russia.

Recent studies of AMRO in the K-SCN salt [7–10] have revealed essentially the same behaviour as in Tl-SCN: the AMRO period was found to vary as $\Delta_0/\cos(\theta - \theta_0)$ with the azimuthal angle θ between the field rotation axis and the crystal c-axis. However, considerable discrepancies between the basic parameters, Δ_0 and θ_0, reported by different authors should be pointed out. The value $\Delta_0 = 1.35$ given in [10] is significantly different from those found in the other works, $\Delta_0 = 1.25 \pm 0.02$ [7,9] and 1.27 [8]. Even larger discrepancy is noticed for the angle θ_0, which determines the position of the specific plane with respect to the room-temperature crystallographic axes; while values $\theta_0 = 19 \pm 2°$ [7] and $21\pm3°$ [9] may be regarded as agreeing with each other, the data reported in [8] and [10] are significantly higher, 27° and 30°, respectively. Assuming their results to be identical with those obtained on Tl-SCN, the authors [10] speculate that the LT superstructure is the same in both compounds, originating directly from the relationship between the crystal parameters, which are very similar in these salts. Meanwhile, if the phase transition is associated with the nesting of the one-dimensional FS sheets, one can expect that even a small variation in the sheet corrugation will lead to a considerable modification of the nesting vector Q. In this connection, it is interesting to study the effect of slight changes in the initial electronic structure introduced by different substitutions of the cation M^+ in the complex anion $[MHg(SCN)_4]^-$ on the oscillation parameters and, consequently, the wave vector Q.

In this paper we present results on the LT AMRO in the Rb-SCN salt, the third member of the α-(BEDT-TTF)$_2$MHg(SCN)$_4$ family exhibiting the phase transition, and compare them with those in Tl-SCN and K-SCN. In order to eliminate the ambiguity in the K-SCN data, we have also measured a sample of this compound, obtaining a good agreement with the results of [7] and [9].

The crystals of the Rb-SCN and K-SCN salts used for the experiment were grown electrochemically [11] and had shapes of thick distorted hexagons or thin square-like platelets with their longest edges usually running along the crystal diagonals. The crystal axis orientations were determined by an x-ray technique. The interlayer (i.e. perpendicular to the highly conducting ac-plane) MR was measured by a standard AC technique in a magnetic field of 11 T, at $T = 1.4$ K, as a function of the angle ϕ between the field direction and the normal, b^*, to the ac-plane. The orientation of the field rotation plane was defined by the angle θ between this plane and the crystal c-axis.

Figure 1 shows the MR angular sweeps obtained on the best of four studied Rb-SCN samples and on a K-SCN sample. The planes of the field rotation correspond to the highest AMRO amplitude, $\theta_0 = 24°$ and 20° for Rb-SCN and K-SCN, respectively. Prominent oscillations are observed for both samples. Figure 2 demonstrates that the dependence of the AMRO period on the azimuthal angle θ obeys the $\propto 1/\cos(\theta - \theta_0)$ law for Rb-SCN. The positions of the MR dips can be described as

$$\tan \phi_n \cos(\theta - \theta_0) = \tan \phi_0 + \Delta_0 n \qquad n = 0, \pm 1, \pm 2, \ldots \qquad (1)$$

with $\theta_0 = 24°$, $\tan \phi_0 = 0.51$ and $\Delta_0 = 1.35$, very similar to the values obtained for the Tl-SCN salt. It should be noted that the oscillations in the Rb-SCN samples are weaker and fade out with increasing ϕ more rapidly than in K-SCN. This is consistent with the low values of MR, $\Delta R/R_0 = 4.7$ at $H = 11$ T, $T = 1.4$ K and the resistance ratio $R_{293\,K}/R_{1.4\,K} = 5.2$ (cf. the corresponding data for the measured K-SCN, $\Delta R/R_0 = 13$ and $R_{293\,K}/R_{1.4\,K} = 40$) indicating the poorer quality of the Rb-SCN samples. The crystal imperfection causes some scattering of the data obtained on different samples. However this scattering does not exceed the error bars indicated in table 1 in which the basic parameters of the AMRO
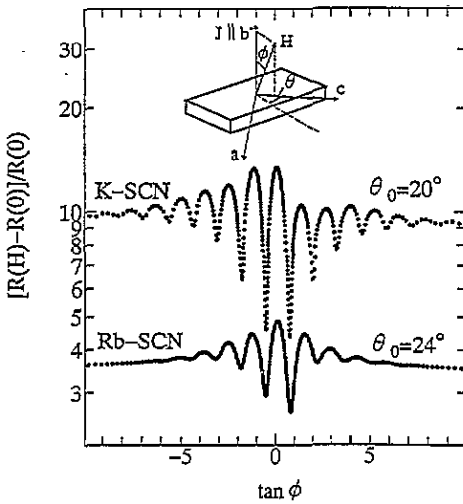
Figure 1. Examples of the AMRO in the Rb-SCN and K-SCN crystals: $H = 11$ T, $T = 1.4$ K; the orientations of the field rotation plane correspond to the highest AMRO amplitudes. The inset illustrates the experimental geometry.
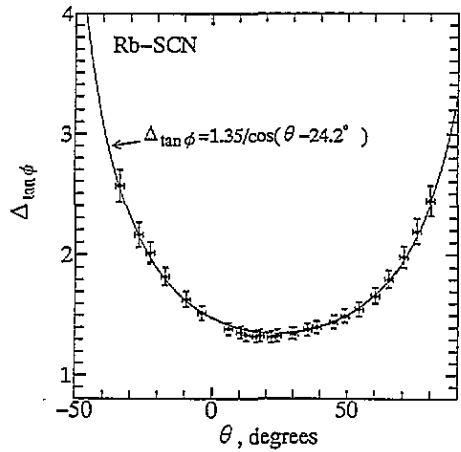
Figure 2. The dependence of the AMRO period $\Delta_{\tan \phi}$ in the Rb-SCN salt on the azimuthal angle θ.

Table 1. The parameters entering the expression (1) for the AMRO positions and the corresponding superstructure wave vector.

	Rb salt[a]	Tl salt[b]	K salt[c]
Δ_0	1.35 ± 0.03	1.35 ± 0.02	1.26 ± 0.02
θ_0 (°)	24 ± 3	24 ± 2	20 ± 2
$\tan \phi_0$	0.51 ± 0.02	0.48 ± 0.02	0.50 ± 0.02
Q^d	$K_a/6 + K_c/3 + K_b/6$	$K_a/6 + K_c/3 + K_b/6$	$K_a/8 + 3K_c/8 + 3K_b/16$

[a] This work.
[b] [5, 6].
[c] This work and [7].
[d] Factors ζ and η in (2) are assumed to be +1, for simplicity.

in Rb-SCN are presented along with the data on Tl-SCN [6] and K-SCN (measured in this work).

As seen in figure 1, the K-SCN sample exhibits strong AMRO, and the parameters given in table 1 are consistent with those reported in [7] and [9]. Comparing the data in table 1, one can conclude that the parameters in Rb- and Tl-SCN coincide with each other within the experimental error. Hence, assuming that the LT superstructure is commensurate with the original crystal lattice, we propose its wave vector to have the same form [6]

$$Q_{\text{Tl, Rb}} = \zeta K_a/6 + K_c/3 + \left(\eta - \tfrac{1}{2}\right)K_b/3 \qquad \zeta, \ \eta = 1 \text{ or } -1. \qquad (2)$$

On the other hand, despite the difference between the above data and those for K-SCN being apparently small, it exceeds the experimental error, at least for the period Δ_0. These data can be fitted by the superstructure wave vector [7], $Q_K = \zeta K_a/8 + 3K_c/8 + 3(\eta - \tfrac{1}{2})K_b/8$. This form is more complicated than that given above for the Tl- and Rb-salts. In

fact, it may be only an approximation for an actually incommensurate Q_K. In particular, such incommensurability may be a reason why the LT instability is weaker in K-SCN than in Rb- and Tl-SCN, i.e. the transition temperature and fields are lower for the former compound.
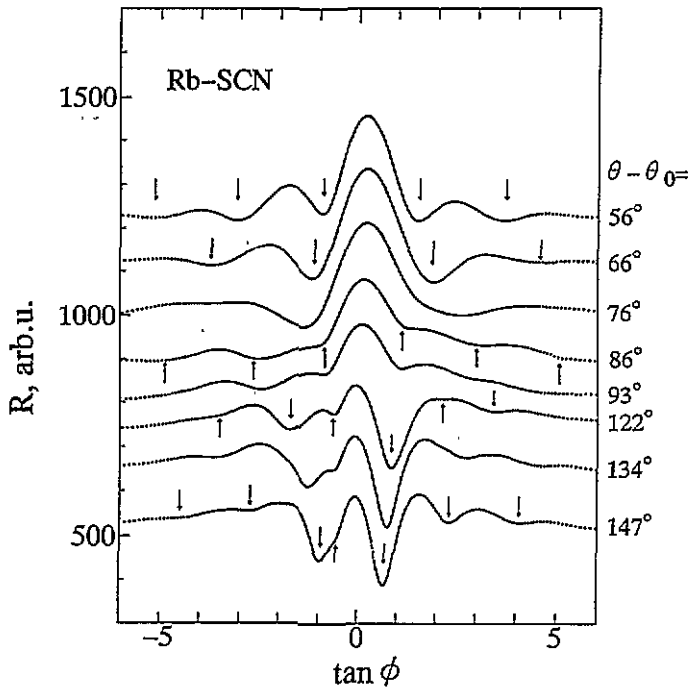

Figure 3. Angular traces of the resistance R of the Rb-SCN salt at several orientations of the field rotation plane defined by the azimuthal angle θ; $\theta_0 = 24°$ corresponds to the plane of the highest AMRO amplitude. The downward and upward arrows point to the main and additional AMRO series (see the text).

Now, turning back to the Rb-SCN salt, we note that besides the main AMRO, there exists an additional series of angular oscillations, most pronounced for the field rotations in the range $(\theta - \theta_0) \simeq 80\text{–}140°$. This is illustrated in figure 3, in which the $R(\tan \phi)$ sweeps at various θ are plotted (the resistance data are offset for clarity). The downward and upward arrows point to the resistance minima corresponding to the main and additional oscillation series, respectively. The dips corresponding to the main AMRO become weaker, their positions diverging in ϕ, on the azimuthal angle $(\theta - \theta_0)$ approaching 90°. At $(\theta - \theta_0) \simeq 70°$ the two series become comparable and it is impossible to separate them from one another. At further increasing θ the second series becomes more pronounced and is the only one observed at $(\theta - \theta_0) = 90 \pm 10°$. In order to clarify the origin of the additional oscillations, we have plotted the positions of *all* the MR dips (in $\tan \phi$) in polar coordinates. The result, presented in figure 4, clearly evidences the coexistence of two AMRO series, so the positions of the second series can be regarded as an exact mirror reflection of the main AMRO against the K_a-axis. This coexistence can be understood by taking into account the specifics of the crystal structure [11]: (i) the angle between the a- and c-axes is nearly 90° ($\beta - 90.57°$),

and (ii) the direction of $(2b + c)$ is almost perpendicular to the ac-plane. In this situation, the reflection against the $(a, 2b + c)$-plane retains the crystal lattice almost (but *not exactly*) unchanged; in particular, the reflection of the c-axis is tilted from the $-c$-direction by less than $1.5°$, taking the room-temperature crystal parameters [11]. Therefore, the crystals can be easily twinned with respect to this plane. The superstructure in the twin domains will be the reflection of that in the original structure against the $(a, 2b + c)$-plane. This will result in the second series of AMRO arising, reflecting the main series with respect to the $K_a K_b$-plane, in agreement with the experiment. Obviously, this corresponds to the apparent change of the sign of the factor ζ in the expression (2) for Q. On the other hand, due to the above-mentioned specifics of the crystal structures, both directions of Q, with $\zeta = 1$ and -1, may be almost equivalent with respect to the open FS nesting conditions. Hence, one cannot exclude the possibility of coexistence of domains with different ζ even in the twinned crystal.
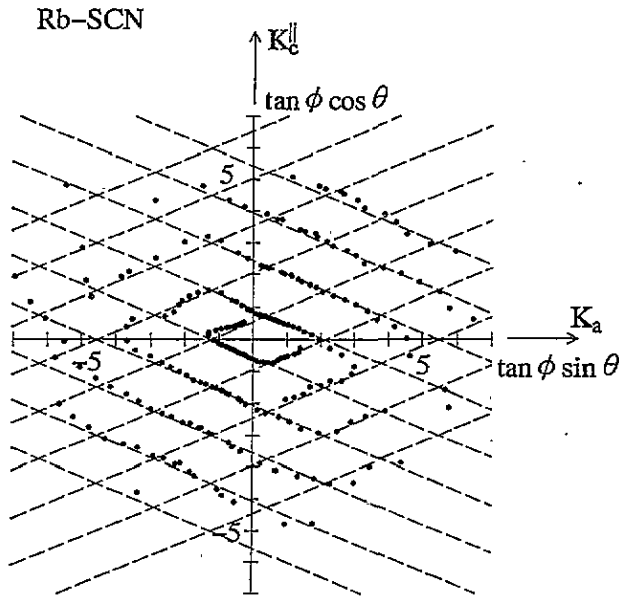

Figure 4. The positions of the MR dips for the Rb-SCN salt in tan ϕ plotted in polar coordinates; $\cos \theta = 1$ corresponds to the field rotation in the cb^*-plane. Arrows show the K_a and $K_c^{\parallel}$ directions of the reciprocal lattice, $K_c^{\parallel}$ being the projection of the K_c-direction on the ac-plane. All the dip positions can be fitted by either the condition (1) with the parameters from table 1 or its mirror reflection with respect to the K_a-axis (dashed lines).

A similar, although relatively weaker, second set of AMRO has been observed in Tl-SCN crystals [6]. As for the K-SCN salt, we have also found some additional features in the $R(\phi)$ curves on rotating the field in the planes corresponding to $(\theta - \theta_0) \simeq 60$–$120°$. However, none of these features can be interpreted in the same way as in the case of Rb- and Tl-SCN. This is demonstrated in figure 5, which shows the polar plot for the positions of MR dips in K-SCN. Despite the fact that the main dips (dots) can be perfectly fitted by the expression (1) with the parameters given in table 1, the additional dips (triangles) can by no means be regarded as their mirror reflections. This result is in agreement with the data reported in [8] and implies another mechanism for the minor oscillations. One, but
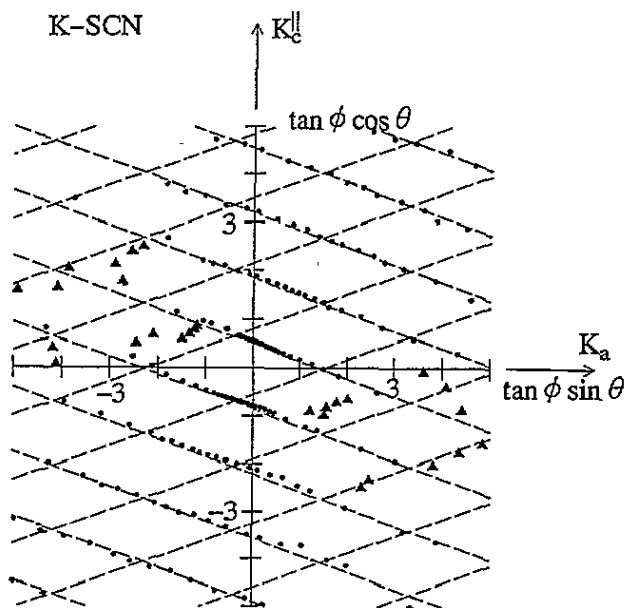
Figure 5. A plot similar to that in figure 4 for the K-SCN salt. While the main AMROS are perfectly described by (1), the additional MR dips (triangles) cannot be regarded as their reflections against the K_a-axis.

not the only, possibility is to involve the 'two-dimensional' AMRO observed in this material above T_p [7]. This would be consistent with the above-mentioned relative weakness of the LT instability in K-SCN.

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