EPR studies of a new family of organic conductors: Dimethyl(ethylenedithio)diselenadithiafulvalene compounds (DMET)$_2$X [X = PF$_6$, BF$_4$, Au(CN)$_2$, I$_3$, and AuBr$_2$]

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A new family of organic conductors dimethyl(ethylenedithio)diselenadithiafulvalene compounds (DMET)$_2$X [X = PF$_6$, BF$_4$, Au(CN)$_2$, I$_3$, and AuBr$_2$] has been investigated by means of EPR. In spite of the large variety of conductive behaviors ranging from insulator to metal, the spin susceptibility at higher temperatures was found to be quite similar in magnitude and in temperature dependence for all the salts (except the AuBr$_2$ salt). Below 20–25 K, the first three salts are found to undergo phase transitions; the transitions in the BF$_4$ and Au(CN)$_2$ salts are clearly magnetic, i.e., an antiferromagnetic or spin-density-wave transition. In the PF$_6$ salt, EPR properties suggest a possible spin-Peierls transition. The I$_3$ and AuBr$_2$ salts have no EPR anomaly in the measured temperature region. The difference in the EPR linewidth among these salts is well understood as a result of the variation of dimensionality, in the framework of the Elliott mechanism. The DMET family contains all the electronic features observed in the tetramethyltetrathiafulvalene, tetramethyltetraselenafulvalene and bis(ethylenedithio)-tetrathiafulvalene families.

I. INTRODUCTION

Since the discovery of metallic conductivity in TTF-TCNQ,[1] the experimental study of organic conductors as low-dimensional electronic systems has been extensive.[2] This epoch-making compound exhibited a charge-density-wave (CDW) instability expected in the one-dimensional system, and prompted many experimental and theoretical investigations. This material and analogs contain two kinds of conductive chains, that make interpretation of the electronic properties rather complicated.[3] The organic conductors of the second generation were single-chain compounds, tetramethyltetrathiafulvalene (TMTTF) (Ref. 4) and tetramethyltetraselenafulvalene (TMTSF) (Ref. 5) systems: these form 2:1 compounds, (TMTTF)$_2$X and (TMTSF)$_2$X, with various counter anions X, that allow a systematic study.[4-6] Most of them are characterized by an open Fermi surface of quasi-one dimensional nature.[7] TMTTF salts generally show gradual metal-insulator transition in resistivity around 200–250 K.[4] At lower temperatures they undergo antiferromagnetic or spin-Peierls (nonmagnetic) transitions in the insulating state.[8] On the other hand, typical TMTSF salts are metallic down to the temperature of the order of 10 K, at which a spin-density-wave (SDW) transition occurs.[5,9] The SDW state is easily suppressed by pressure and gives way to the superconducting state below 1 K.[10] The third family, bis(ethylenedithio)-tetrathiafulvalene (BEDT-TTF) salts,[11] has a more two-

dimensional feature, characterized by a cylindrical closed Fermi surface,[12] than the previous two families. The BEDT-TTF family does not exhibit any Fermi-surface instability and contains several ambient-pressure superconductors,[13] including the recently synthesized superconductor (BEDT-TTF)$_2$Cu(NCS)$_2$ (Ref. 14) with the highest transition temperature T_c of 10.4 K among the organic systems to date. Each family possesses distinctive electronic properties: The difference is so large that our knowledge on the organic conductors is still far from systematic understanding.

Recently, Kikuchi et al.[15-19] have synthesized a new family of organic conductors with dimethyl-(ethylenedithio)diselenadithiafulvalene (DMET) as donors. The DMET molecule is a hybrid between TMTSF and BEDT-TTF molecules: one-half of the molecule is that of TMTSF and the other half, BEDT-TTF (see Fig. 1). This molecule forms 2:1 compounds, (DMET)$_2$X with a variety of monovalent anions X^-, as TMTTF, TMTSF, and BEDT-TTF do. One of the most remarkable characters of this family is a rich variety of transport properties ranging from insulator to superconductor.[19] From the resistivity and the structural analysis it has been found that this family can be classified into five groups which are represented by the five salts given in the title, respectively.[19] The temperature dependence of the resistivity for these salts is summarized in Fig. 2. (DMET)$_2$PF$_6$ belongs to group I and is an insulator, at least, below room temperature;[15] (DMET)$_2$BF$_4$ in group
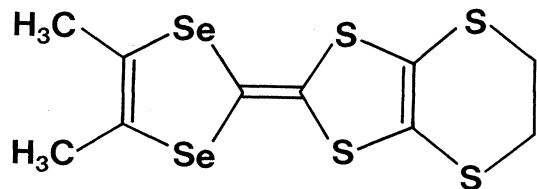
FIG 1. Dimethyl(ethylenedithio)diselenadithiafulvalene molecule.

II is metallic down to 37 K, at which a metal-insulator transition occurs;[15] $(DMET)_2Au(CN)_2$ in group III exhibits a metal-insulator transition at 25 K at ambient pressure, but a superconducting transition at 1 K when pressurized;[16] $(DMET)_2I_3$ in group IV is an ambient-pressure superconductor with a T_c of 0.5 K, above which the resistivity is metallic;[17] $(DMET)_2AuBr_2$ has at least two different crystal structures, one of which has been classified as group V; this salt shows an anomalous temperature dependence of the resistivity shown in Fig. 2 and is an ambient-pressure superconductor with a T_c of 1.9 K, the highest among the DMET family to date.[20] The difference in the transport properties corresponds to that in the structure. (The structures of groups III and IV with linear anions are essentially the same, however.) The family of DMET salts seems to contain all the features of the transport properties found in the TMTTF, TMTSF, and BEDT-TTF salts. The experimental study of $(DMET)_2X$ will serve for a comprehensive understanding of organic conductors. At the present stage, accumu-
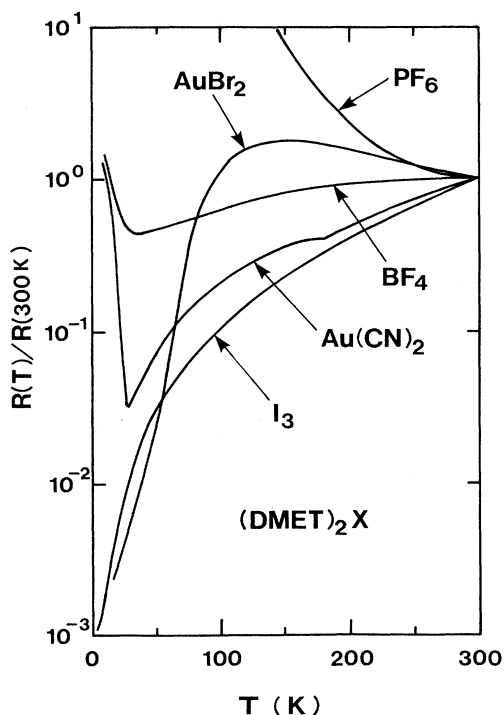
lation of various experimental data for this family is most required.

We have performed electron-paramagnetic-resonance (EPR) measurements to explore the electronic states microscopically for a series of $(DMET)_2X$ [$X=PF_6$, BF_4, $Au(CN)_2$, I_3, and $AuBr_2$]. In this paper, the temperature dependences of the spin susceptibility and the linewidth for each salt are given. The high-temperature susceptibility behavior and the observed low-temperature phase transitions are discussed comparatively. A brief report of the present results was previously made.[21]

II. RESULTS

The preparation method and structural analysis of $(DMET)_2X$ were described in the previous papers.[15-18,22,23] The EPR measurements were performed in the temperature range between 10 and 300 K, using an X-band ESR spectrometer, JES-FE3XG (JEOL). The temperature dependence of the Q factor of the cavity was calibrated with the use of Mn^{2+}-MgO solid solution. The absolute value of the spin susceptibility, χ_{spin}, was determined with reference to the signal of $(TMTSF)_2PF_6$, χ_{spin} of which was well established as 2.3×10^{-4} emu/mol at room temperature by Pedersen et al.[24]

A. $(DMET)_2PF_6$

Figure 3 shows the spin susceptibility, χ_{spin}, of $(DMET)_2PF_6$ as a function of temperature. The room-temperature value of χ_{spin} is 2.0×10^{-4} emu/mol. The χ_{spin} is rather insensitive to temperatures between room temperature and 230 K, and then gradually decreases at lower temperatures down to 25 K. Below 25 K, χ_{spin} decreases more rapidly, suggesting some phase transition. Such a temperature dependence of the spin susceptibility has widely been observed in many organic systems.[8,24]
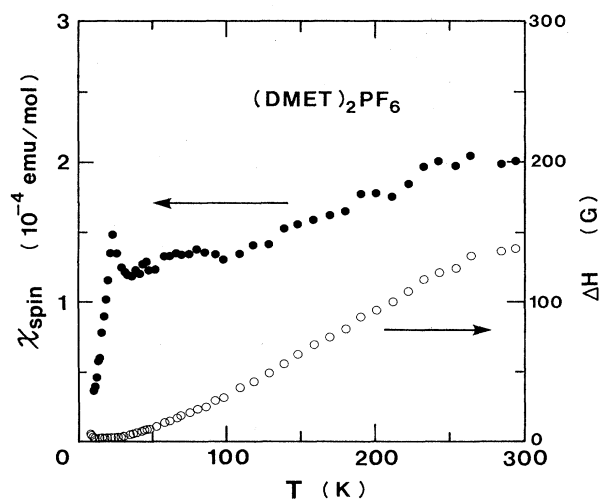

FIG. 2. Temperature dependences of the resistivities along the conductive axis or plane of the crystals for $(DMET)_2X[X=PF_6$, BF_4, $Au(CN)_2$, I_3, and $AuBr_2]$.

FIG. 3. Electron-spin susceptibility and linewidth determined by EPR for $(DMET)_2PF_6$ as a function of temperature. The sample was mounted such that the long axis of the crystal (the stacking axis) was parallel to the microwave field and the crystal face (the ab plane) was parallel to the static field.

As mentioned earlier, the PF_6 salt is insulating in resistivity, so that discussion based on the Pauli paramagnetism is irrelevant to this salt. It is natural to consider the observed temperature dependence from a view of the Bonner-Fischer–type paramagnetism,[25] which applies to a system of antiferromagnetically interacting quasi-one-dimensional localized spins. It is well known that such a system has two different kinds of ground states; one is a spin-Peierls state and the other is an antiferromagnetically ordered state. Either of them occurs depending on the relative strength between the interchain interaction and the spin-lattice coupling.[26] An example in the former case is $(TMTTF)_2PF_6$ (Ref. 27) and in the latter, $(TMTTF)_2SCN$.[28]

In the present salt $(DMET)_2PF_6$ we do not have any direct evidence thus far which distinguishes the two possibilities. From the general point of view of EPR, in the case of the antiferromagnetic transition, the EPR signal disappears suddenly at the transition (the Néel temperature), accompanied by an abrupt line broadening in the transition region. On the contrary, it diminishes more moderately (following the temperature dependence of an activation type) in the case of the spin-Peierls transition. The decrease of χ_{spin} below 25 K in this salt seems to favor a spin-Peierls transition. The linewidth, ΔH, decreases monotonously with decreasing temperature and does not show any anomaly but a slight broadening at the lowest temperature well below the phase transition. Taking into consideration possible sample inhomogeneity, however, we cannot make a definite conclusion. To determine which transition is the case for this salt, studies of NMR and diffuse x-ray scatterings[29] are required.

B. $(DMET)_2BF_4$

Next we present the results for $(DMET)_2BF_4$ in Fig. 4. The resistivity behavior of this salt is metallic down to 37 K, at which a metal-insulator transition occurs.[15] Accordingly, the observed EPR signals become appreciably of a Dysonian type at lower temperatures. The asymmetry parameter, A/B defined in the figure is also shown in this figure, where one can see that A/B becomes maximum at 37 K. So the values of χ_{spin} is a little underestimated in the temperature range between 35 and 100 K. In any case, both the overall temperature dependence and the magnitude of χ_{spin} (2.2×10^{-4} emu/mol at room temperature) are similar to the previous sample, $(DMET)_2PF_6$, in spite of a large difference of the transport properties. This will be discussed later.

At around 20 K (well below the metal-insulator transition in resistivity), the susceptibility vanishes suddenly. Around this temperature, the EPR linewidth, ΔH, increases sharply as is seen in the figure. These characteristics are typical of an antiferromagnetic ordering and indicate that the ground state of this material is an antiferromagnetic state. Note that the magnetic transition takes place at 20 K, much lower than the temperature of the resistance minimum 37 K. The other salts in group II (the ClO_4 and ReO_4 salts) have the resistance minimum at higher temperatures around 100–200 K (Ref. 19) and magnetic transitions are expected well below those tem-
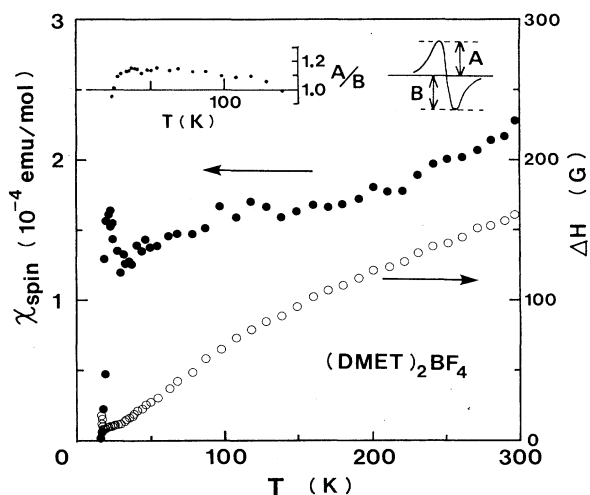

FIG. 4. Electron-spin susceptibility and linewidth determined by EPR for $(DMET)_2BF_4$ as a function of temperature. The inset shows a definition of the asymmetry parameter, A/B, and its temperature dependence. This compound has two different stacking axes (the b and c axes). The sample was mounted such that the crystal face (the bc plane) was parallel to both the static and the microwave fields.

peratures as in the BF_4 salt. This situation is quite similar to the TMTTF family, where $(TMTTF)_2Br$ has the lowest temperatures, 100 K for the resistance minimum and 14 K for the SDW transition.[4,30,31]

The temperature dependence of ΔH as a whole is quite similar to those of the TMTTF and TMTSF families. ΔH is 160 G at room temperature and it decreases linearly with decreasing temperature. It seems to shift to a steeper linearity below 130 K. Pedersen et al.[24] considered that the "characteristic temperature" for the break in slope corresponds to the interchain transverse integral; the transverse electron motion should be coherent below that temperature and be diffusive above that. If it is correct, the dimensionality of $(DMET)_2BF_4$ turns out to be similar to that of $(TMTSF)_2PF_6$.

C. $(DMET)_2Au(CN)_2$

The results for $(DMET)_2Au(CN)_2$ at low temperatures have been published separately.[32] As discussed there, the EPR properties of this salt were more or less sample dependent. Figure 5 shows one example of the data which could be taken up to room temperature. The absolute value of the susceptibility at room temperature is 2.3×10^{-4} emu/mol, which is near the values of the previous two salts. The χ_{spin} seems to make a broad peak at around 230 K, below which it decreases with decreasing temperature in a similar manner to the PF_6 and BF_4 salts. At 25 K, χ_{spin} starts to decrease suddenly, indicating a phase transition. For this specimen, the transition was incomplete, probably due to an inhomogeneity in the sample. In the previous paper, we have confirmed for several crystals that χ_{spin} vanishes completely at low temperatures.[32] Crystals of such "good quality" were so small that EPR signals were available only below 100 K.
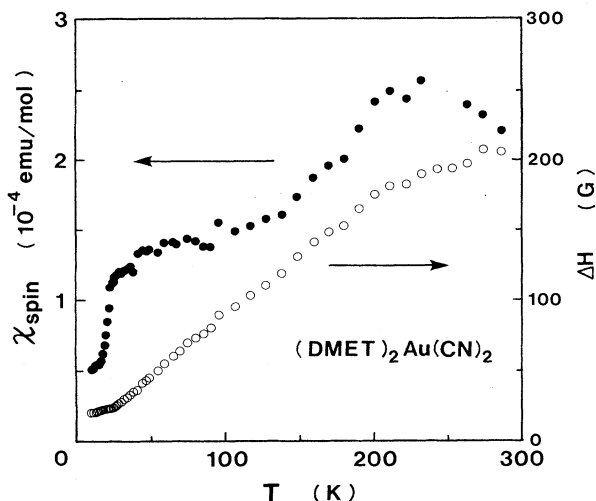
FIG. 5. Electron-spin susceptibility and linewidth determined by EPR for $(DMET)_2Au(CN)_2$ as a function of temperature. The sample was mounted such that the long axis of the crystal (the stacking axis) was perpendicular to the microwave field and the crystal face (the ab plane) was parallel to the static field. The difference of the linewidth in two geometries of the long axis, parallel and perpendicular to the static field, was $\sim 15\%$.

This low-temperature transition was proved to be an SDW transition by the proton NMR linewidth and the spin-lattice relaxation-time measurements.[32] The transition temperature is quite near the temperature of resistance minimum. These features are generally observed in the $(TMTSF)_2X$ family and, therefore, a close similarity of $(DMET)_2Au$ $(CN)_2$ to this family is evident.

The value of the linewidth at room temperature is 210 G. As temperature is decreased, ΔH decreases linearly and forms a knee at 200 K, below which ΔH shows a steeper linearity. This behavior is similar to $(DMET)_2BF_4$. However, the "characteristic temperature" is higher than $(DMET)_2BF_4$. According to the discussion by Pedersen et $al.$[24] mentioned earlier, it turns out that $(DMET)_2Au(CN)_2$ is of higher dimension than $(DMET)_2BF_4$. The same conclusion may also come from the fact that the magnitude of ΔH in the $Au(CN)_2$ salt is greater than in the BF_4 salt, which is discussed in more detail later. Around the SDW transition at 20 K, ΔH exhibits a deviation from the linear dependence on temperature, a symptom of line broadening due to the antiferromagnetic ordering. The possible reasons why the line broadening was not so clear at the transition for this salt were discussed previously.[32]

D. $(DMET)_2I_3$

The compound $(DMET)_2I_3$ also exhibits a behavior similar to the previous salts, although reliable data were only obtained below 150 K due to small dimensions of the crystals and large linewidth (see Fig. 6). The temperature dependence of χ_{spin} is a weakly increasing function of temperature and the absolute value is 1.6×10^{-4}
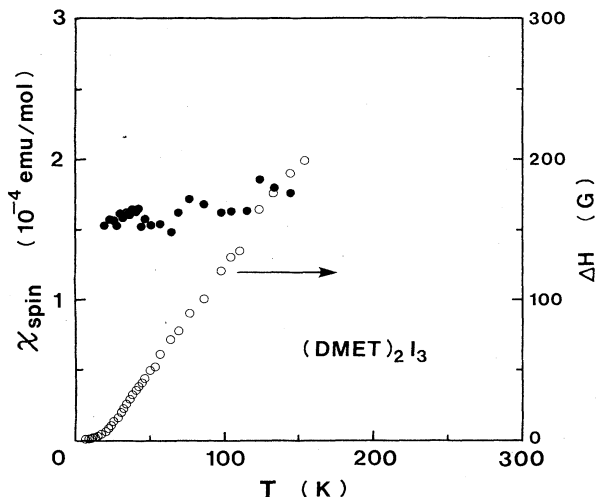

FIG. 6. Electron-spin susceptibility and linewidth determined by EPR for $(DMET)_2I_3$ as a function of temperature. The sample was mounted such that the long axis of the crystal (the stacking axis) was parallel to the microwave field and the crystal face (the ab plane) was parallel to the static field.

emu/mol at 100 K. There exists no χ_{spin} anomaly indicating any type of transition in the measured temperature range. This is consistent with the metallic behavior of resistivity down to 0.5 K.[17] Above 30 K, ΔH exhibits a linear temperature dependence, as in the other salts, but is much larger than them. We show later that the larger linewidth, as well as the absence of Fermi-surface instability, is suggestive of the higher dimensionality; this salt has been found to be an ambient pressure superconductor,[17] like $(TMTSF)_2ClO_4$ and many salts of the BEDT-TTF family.

The linewidth varies with temperature as $T^{2.9}$ in the temperature range between 10 and 30 K. Since this salt is free from a magnetic instability at low temperatures, we expect the power law for the temperature dependence of the linewidth to be related to the transport properties in this system, as is the case for the Elliott mechanism. However, the resistivity of $(DMET)_2I_3$ shows a dependence of $T^{1.8}$ in the wide temperature range.[17] The temperature dependences of the linewidth and the resistivity never agree for all the present salts. This problem will be briefly discussed later.

E. $(DMET)_2AuBr_2$

As described earlier, the $AuBr_2$ salt has, at least, two kinds of crystal structures. From the resistivity measurements, we have confirmed that the samples reported here are the ambient-pressure superconductors. The EPR properties of this salt are quite different from the other four salts described before. Above all, the linewidth is extraordinarily large, so that the single-crystal measurements were not possible except below 30 K. We mounted 15 crystals on a sample holder so as to make the crystal faces (the ab plane) parallel to both of the static and the microwave fields. By this method, we have succeeded in observing an EPR signal of $(DMET)_2AuBr_2$ up to 300 K.

The results at low temperatures agree well with those for a single crystal, and thus we consider that they are intrinsic. The result thus obtained are shown in Fig. 7.

The integrated signal intensity, χ_{spin}, was found to be insensitive to temperature within the experimental error, at least above 50 K. The χ_{spin} is $(2.90 \pm 0.25) \times 10^{-4}$ emu/mol between 50 and 300 K, and is larger than any other DMET salt. A decrease of χ_{spin} below 50 K is due to the skin effect witnessed by the appearance of Dysonian line shape, and therefore is not intrinsic.

At room temperature, ΔH is as large as 930 G; this value is the largest in this family. Moreover, it is extraordinarily large among the organic conductors. The temperature dependence of ΔH is also anomalous; with decreasing temperature, ΔH increases gradually and takes a maximum in excess of 1000 G around 150 K, followed by a rapid decrease at lower temperatures. The ΔH at 20 K is still 270 G. This behavior is a strong contrast to those of the usual organic conductors such as the TMTSF, BEDT-TTF salts, and the other salts in the DMET family, where ΔH decreases monotonously with decreasing temperature in the metallic region. Typical values of ΔH for the TMTSF and BEDT-TTF salts at low temperatures are less than 10 G.

Surprisingly, the linewidth anomaly corresponds well to that in the resistivity shown in Fig. 2. The resistivity and the linewidth have a maximum at almost the same temperature, 150 K, although the similarity of the tem-

perature dependences for the two quantities are only qualitative. (Note that the vertical scales in Figs. 2 and 7 are different from each other.) On the other hand, χ_{spin} has no anomaly at that temperature. Therefore, we consider that the present features of the resistivity and the EPR linewidth evidence some anomaly around 150 K, probably, of dynamical origin. Detailed discussions will be published separately.[33]

III. DISCUSSION

From a general view of the present results, one can see that the family of DMET salts contains all the magnetic and transport characteristics of the TMTTF, TMTSF, and BEDT-TTF families, which are distinctive from each other and have been analyzed separately. Now we have a family of organic conductors which may connect the previous families. It is noteworthy that the overall temperature dependence and the magnitude of the susceptibility for $(\text{DMET})_2 X$ [$X = \text{PF}_6$, BF_4, Au(CN)_2, and I_3] are essentially the same, while the resistivity ranges widely from insulating (for the PF_6 salt) to metallic (for the I_3 salt) behaviors. This fact implies that the magnetic behavior is determined by the same mechanism and that it makes no difference, whether the system is metallic or insulating.

The electron-spin susceptibility is a fundamental physical quantity which characterizes electronic systems, and precise measurements have been performed in the earlier organic conductors. To understand the spin susceptibility in the organic systems, two different starting points have been proposed; one is a Pauli paramagnetism for weakly interacting conduction electrons and the other is a Bonner-Fisher–type paramagnetism for antiferromagnetically interacting localized electrons in one-dimensional system. The latter has been applied to highly correlated electronic systems, where the effective exchange interaction J_{eff} is given as $2t^2/U$, with the on-site Coulomb repulsion, U, and the transfer integral, t. Any real system should be in between and a key parameter connecting the two limiting cases is the relative strength of U and the band width, $4t$ (according to the tight-binding model). Since the present salts are based on the same molecule, DMET, it is reasonable to suppose the same U for all the salts. In the above context, the observed similarity of the spin susceptibility suggests that $(\text{DMET})_2 X$ are situated in the same regime of electron-correlation parameters U and $4t$. Therefore, we find difficulty in the naive interpretation that the metallic I_3 salt might show a Pauli paramagnetism relevant to weakly interacting electrons while the insulating PF_6 salt might be a Mott-Hubbard insulator with strong correlations. Now, what is the origin of the large difference in the transport properties? It is a serious problem.

An important suggestion is given by the earlier discussions to get a systematic understanding of the transport and magnetic properties in the TMTTF and TMTSF families.[34] It has been found that these two families have quite similar magnetic properties in spite of large difference in the resistivity.[8,24] These salts, as well as the DMET salts, are the so-called 2:1 compounds and have a quasi-one-dimensional quarter-filled band. In the salts
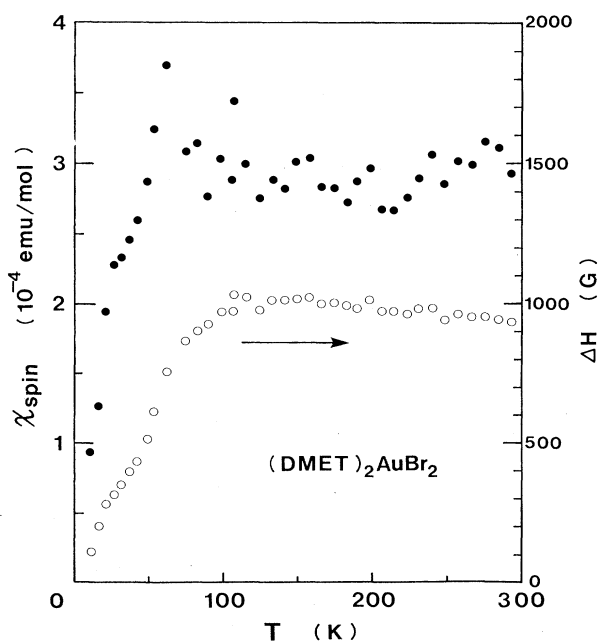

FIG. 7. Electron-spin susceptibility and linewidth determined by EPR for $(\text{DMET})_2\text{AuBr}_2$ as a function of temperature. The samples were mounted such that the crystal faces (the *ab* plane) were parallel to both of the static and the microwave fields. A decrease of the susceptibility below 50 K is due to the skin effect clearly witnessed by the appearance of the Dysonian line shape and therefore is not intrinsic.

with this structure, a slight dimerization of molecules along the stacking axis due to anion potential produces the so-called $4k_F$ potential for charge carriers; the $4k_F$ electron-electron Umklapp scatterings become relevant. For a sizable dimerization, the electron band behaves effectively half-filled and the $4k_F$ Umklapp scatterings lead to a correlation gap in the charge degree of freedom; this situation is equivalent to a Mott-Hubbard insulator in the limit of strong Coulomb repulsion. Emery et al.[34] suggested that the temperature of the resistance minimum generally observed in TMTTF salts should be proportional to the correlation gap. Thus, depending on the dimerization, the resistivity at low temperatures behaves either metallic or nonmetallic. Note that the excitations in the spin degree of freedom are free from the correlation gap; the spin susceptibility should have no anomaly around the temperature of the resistance minimum. This picture applies to a pure one-dimensional system. With increasing the interchain coupling, the Umklapp scattering becomes less effective for the Fermi-surface instabilities so that the system finally behaves metallic; the TMTSF family is believed to be situated in this limit. The variety of the electronic properties of $(TMTTF)_2X$ and $(TMTSF)_2X$ are thus understood by using the $4k_F$ potential (or the degree of dimerization) as a parameter, which is the explanation for these families, so far widely accepted.

We expect a quite similar situation in the present DMET family. Since the asymmetric DMET molecules are stacked alternately, it is quite reasonable to expect the effect of "dimerization" on the electronic properties. In this picture, the PF_6 salt is considered as a correlation-induced insulator, with a correlation gap larger than room temperature (the largest among these salts); the BF_4 and $Au(CN)_2$ salts have smaller gaps in this order. The I_3 salt is of the highest dimension and behaves metallic throughout the entire temperature range. Each salt seems to have its analog in the TMTTF and TMTSF families. The $AuBr_2$ salt is no longer one dimensional but is two dimensional, analogous to the BEDT-TTF family. The validity of these discussions should be checked once we know more about the band structure and its relation to the crystallographic structure; the five groups of the DMET family are distinctive in structure.

The large variety of the linewidth among the salts is a strong contrast to the global similarity of the χ_{spin} discussed above. Usually, the main contribution to the EPR linewidth is believed to come from the so-called Elliott mechanism;[35] orbital scatterings determine the lifetime of the Zeeman states through the spin-orbit coupling. This contribution to the linewidth is written as $\Delta H \sim a(\Delta g)^2/\tau_{tr}$, where Δg is the g shift from the free-electron value and is a measure of the spin-orbit coupling, τ_{tr} is the orbital scattering time and a is a numerical factor. In the low-dimensional system, however, this mechanism is known to be seriously modified; in a pure one-dimensional system where only forward and backward scatterings are allowed, spin-flip transitions accompanied by the backward scatterings are strictly forbidden because of the requirement of time reversal symmetry. (Instead, the motional narrowing of the dipolar linewidth becomes incomplete because of the so-called long-time tail of the spin-correlation function in a one-dimensional system. The dominant contributon to the linewidth may thus come from the dipolar coupling, which is completely neglected in the usual three-dimensional systems.)[36-39]

In this context, there are two aspects which seem relevant to the difference in the EPR linewidth in the organic conductors, i.e., the spin-orbit coupling and the dimensionality. The former has been accepted as an explanation of the large difference of the linewidth between the TMTTF and TMTSF salts, since the selenium atom has much stronger spin-orbit coupling than sulfur (the spin-orbit coupling is proportional to Z^4, with atomic number Z). It should also be relevant to the difference between the BEDT-TTF and the present DMET salts. However, the large variety among the DMET family cannot be attributed to it, since they are based on the same molecule.

Qualitatively, the difference in the dimensionality seems to explain the variety of the linewidth in the DMET family, consistent with the discussion on the resistivity mentioned earlier. The higher dimensionality in electronic states should release the above-mentioned restriction of the Elliott mechanism, leading to larger linewidth. Actually, the properties of the PF_6 salt described above suggest that this salt is the most one-dimensional among the five salts, and the crystal structure of the $(DMET)_2AuBr_2$ shows two-dimensionality which has also been supported by the preliminary measurements of resistivity anisotropy.

However, a simple Elliott model is not precise enough to discuss quantitatively the temperature dependence of EPR linewidth. While this model claims a perfect proportionality between the electric resistivity and the EPR linewidth, it has never been realized in any real organic conductors, including the present $(DMET)_2X$. (Precise discussions on one-dimensional TTF-TCNQ and analogs appeared earlier.)[4] This may imply that some sophisticated mechanism incorporating other contributions to the linewidth such as dipolar broadening is involved. Further measurements on the linewidth anisotropy, which are going on, are fruitful for this problem. In addition, according to the picture described before to explain the variety of resistivity, the temperature dependence of the resistivity is not simply determined by that of the scattering rate, τ_{tr}, particularly at temperatures lower than the correlation gap. This may be the key to understanding the difference between the temperature dependences of the linewidth and the resistivity in the PF_6 salt. The qualitative similarity between the temperature dependences of the linewidth and the resistivity for the other salts, including the $AuBr_2$ salt, strongly suggests that the essential features are related to the Elliott mechanism.

The low-temperature ground state will depend on some interchain coupling parameters, rather than the intrachain properties. The ground states of the $(DMET)_2X$ at ambient pressure have been found to be the antiferromagnetic-SDW state for the BF_4 and $Au(CN)_2$ salts, and the superconducting state for the I_3 (Ref. 17 and $AuBr_2$ (Ref. 18) salts. In the PF_6 salt, the present EPR results seem to favor a spin-Peierls transition, but

we cannot rule out the possibility that a magnetic transition has simply been smeared out by sample inhomogeneity. It is interesting to note that the low-temperature transitions occur at nearly the same temperature, 20–25 K, in the DMET family. Quantitative discussion will be possible in the near future when the results of other experiments and the calculation of band parameters will be available. The DMET compounds are fascinating materials for a comprehensive study of the organic conductors.

The strong electron-electron correlation should also affect the superconductivity. It is interesting to investigate the superconductivity in $(DMET)_2I_3$, $(DMET)_2Au(CN)_2$, and $(DMET)_2AuBr_2$ from this point of view. The previous NMR studies of $(TMTSF)_2ClO_4$ have given evidence of anisotropic electron pairing in the superconductivity.[41,42] It has been suggested that the strong on-site Coulomb repulsion may prevent a local attractive force and then intermolecular attraction resutls in the anisotropic pairing. If this is true, similar situations are also likely in the DMET salts.

ACKNOWLEDGMENTS

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