RESEARCH ACTIVITIES IV
Department of Molecular Assemblies

IV-A Spectroscopic Study of Charge Carriers in Organic Conductors

The low-frequency reflectivity of an organic conductor provides us with a wealth of information on the nature of charge carriers. For instance, the anisotropy of a band structure, bandwidth, effect of electron-electron correlation, and electron-molecular vibration (EMV) coupling parameters can be extracted from the analysis of the reflectivity or optical conductivity curve. We are investigating the polarized reflection spectra of various organic conductors in the spectral region of 50–33000 cm^{-1} and in the temperature range of 6–300 K. Usually the molecular vibrations (local phonons) are screened by strong electronic transition by charge carriers. Therefore, very few molecular vibrations are detected in the reflection spectrum. In this sense, the Raman spectroscopy is a complementary method to reflection spectroscopy for understanding molecular vibrations in a metallic state. Since some molecules have charge-sensitive vibrational modes, the Raman spectroscopic method is a powerful tool to detect the site-charge distribution (oxidation state of molecule). We are investigating the charge ordering (CO) phenomena in organic conductors using the technique of infrared and Raman spectroscopy. In the organic charge-transfer salts, CO is originated from the localization of the charge carriers. Since the charge carriers in organic crystal is located at the boundary between localized and extended (delocalized) states, CO will be widely found through the phase transition. The charge ordering was first found in inorganic narrow-band systems such as copper, manganese, and vanadium oxides. Recently, CO has been found in several organic conductors, and the electronic phase diagrams of typical organic conductors are re-examined taking CO into account. The CO state is drawing attention, since CO is theoretically considered as being related to the unconventional superconducting mechanism. The Raman and infrared spectra change dramatically at the CO phase-transition temperature, since CO is accompanied by a charge disproportionation, forming an inhomogeneous charge distribution. Our goal is the complete understanding of the CO phase transition through the interpretation of the vibrational spectra, and the drawing of a *P-T* phase diagram.

IV-A-1 Raman Study of the Charge Ordering in α-(BEDT-TTF)$_2$I$_3$ at High Pressure

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We have carried out Raman experiments on α-(BEDT-TTF)$_2$I$_3$ to investigate the site-charge distribution among BEDT-TTFs in the paramagnetic semi-metal-like (PM) and diamagnetic insulating (DI) phases at ambient and quasi-hydrostatic pressures. We focused on the charge-sensitive ν_2 and ν_3 modes of BEDT-TTF, which are related to the ring and central C=C stretching modes with A_g symmetry. We found a distinctive spectral change associated with metal-insulator phase transition up to 1.5 GPa (above this pressure the DI phase was suppressed).

In the DI phase, ν_2 splits into four bands labeled by $\mathbf{c_{1a}}$, $\mathbf{c_{1b}}$, $\mathbf{c_2}$, and $\mathbf{c_3}$, which are confirmed by the isotope shift of the central ^{13}C=^{13}C band). The presence of the four bands of ν_2 indicates the presence of the four crystallographically non-equivalent BEDT-TTFs. This observation means that the inversion symmetry characteristic of the PM phase disappears in the DI phase. The magnitude of the observed splitting and the symmetry breaking indicate that charges are disproportionate in so-called horizontal stripes (perpendicular to stacks). Quasi-hydrostatic pressures induce a progressive downshift of $\mathbf{c_{1a}}$ and $\mathbf{c_{1b}}$ accompanied with an upshift of $\mathbf{c_2}$ and $\mathbf{c_3}$, indicating the continuous change of the charge disproportionation (CD) ratio, while the band position is temperature independent under a constant pressure. Assuming the linear dependence of the ν_2 frequency on the ionicity of BEDT-TTF, the CD ratio between the charge-rich and charge-poor sites is evaluated as *ca.* 0.2*e* : 0.8*e* at low pressures, which gradually decreases to *ca.* 0.3*e* : 0.7*e* near 1.5 GPa. The CD ratio decreases and eventually CD is discontinuously suppressed above 1.5 GPa. This is because the transfer integrals (*t*) increase faster than the inter-site (*V*) Coulomb repulsions (on-site (*U*) is pressure independent) as expected. In the PM phase the ν_2 mode is split into two $\mathbf{a_1}$ and $\mathbf{a_2}$ bands (for ^{13}C substituted sample three bands were observed) indicating not uniform charge distribution among BEDT-TTFs. The charge difference evaluated from the splitting between the $\mathbf{a_1}$ and $\mathbf{a_2}$ bands is small, say 0.4*e* : 0.6*e*, which is weekly dependent upon the pressure.

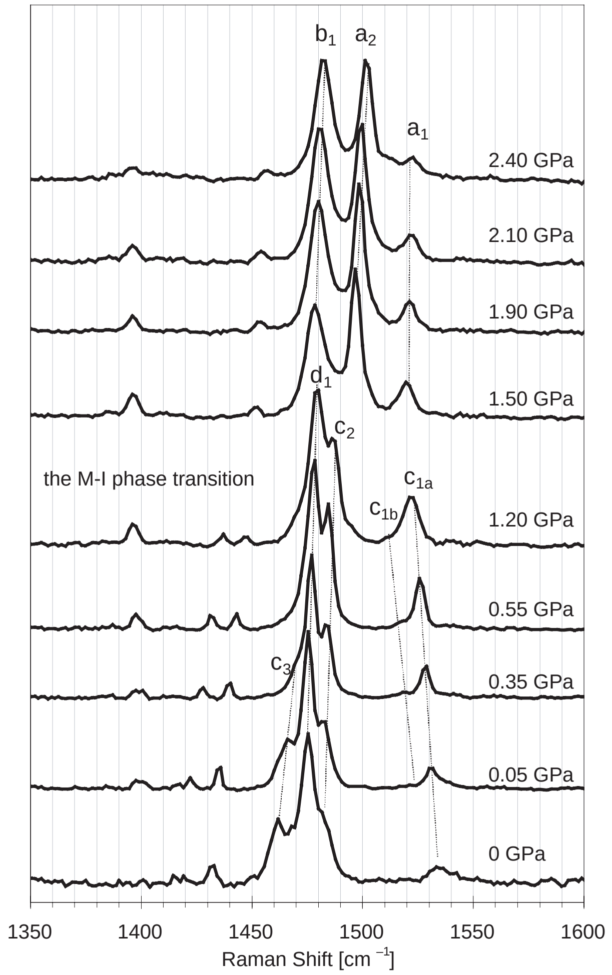

Figure 1. The pressure dependence of the α-(BEDT-TTF)$_2$I$_3$ Raman spectra taken at 20 K with 514 nm laser line polarized along *b*-axis *i.e.* perpendicular to the stacks.

IV-A-2 Raman Study of the Charge Ordering in α'-(BEDT-TTF)$_2$IBr$_2$ at High Pressure

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Raman spectroscopy experiments have been carried out for α'-(BEDT-TTF)$_2$IBr$_2$ at ambient and under several quasi-hydrostatic pressures up to 2.3 GPa. We have focused on the charge-sensitive ν_2 and ν_3 modes of BEDT-TTF', which are related to the rings and central C=C stretching modes with A_g symmetry. We have examined these modes from room temperature to 20 K in order to detect the change in charge-distribution resulting from the structureless phase transition at 250 K at ambient pressure. We first confirmed from the splitting of ν_2 ($\mathbf{a}_1$, $\mathbf{a}_2$) and ν_3 ($\mathbf{b}_1$, $\mathbf{b}_2$) [See the spectrum at ambient pressure in Figure 1] that this phase transition is characterized as the charge-ordering (CO) phase transition. On increasing pressure, the phase-transition temperature (T_{CO}) decreases monotonously. Eventually above 1 GPa, the Raman spectra characteristic of the CO phase, which consist of two ν_2 ($\mathbf{a}_1$, $\mathbf{a}_2$) and two ν_3 ($\mathbf{b}_1$, $\mathbf{b}_2$), are suppressed in the entire temperature range. This suggests that inter-stack transfer integrals between BEDT-TTFs increases faster than the inter-site Coulomb repulsion when the unit cell volume is reduced by pressure. In the semiconducting phase of α'-(BEDT-TTF)$_2$IBr$_2$, the Raman spectra consist of a rather broad band in contrast to the spectrum of metallic β-(BEDT-TTF)$_2$IBr$_2$ or θ-(BEDT-TTF)$_2$I$_3$ salts, in which ν_2 and ν_3 are well separated. We have found that the pressure over 1.8 GPa is required for this compound to reach the uniform charge distribution like metallic salts [See the spectra above 1.8 GPa in Figure 1].

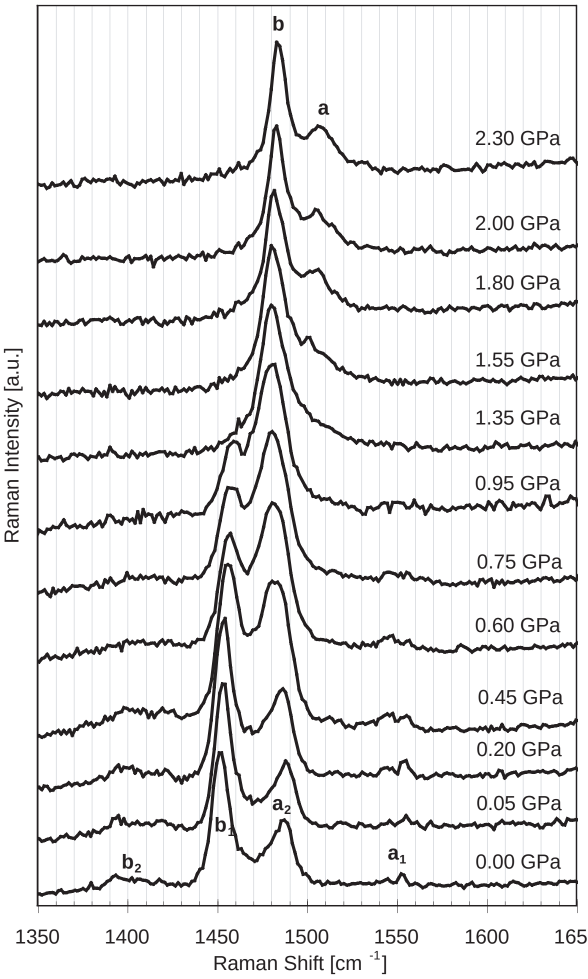

Figure 1. Raman spectra recorded at 100 K under several pressures up to 2.3 GPa, from the (001) crystal face with 514 nm laser line polarized parallel to [1$\bar{1}$0] *i.e.* perpendicular to BEDT-TTF stacks.

IV-A-3 Charge Ordering in θ-(BEDT-TTF)$_2$Tl*M*(SCN)$_4$ (*M* = Co and Zn) Studied by Vibrational Spectroscopy

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[*Synth. Met.* to be published]

Mori *et al.* systematically synthesized the series of θ-(BEDT-TTF)$_2MM'$(SCN)$_4$, [*M* = Cs, Rb, Tl; *M'* = Co, Zn], (abbreviated as θ-*MM'*), and proposed the phase diagram which demonstrated the relation between the metal-insulator transition temperature and the dihedral angle between BEDT-TTF molecules in adjacent stacks.[1)] Recent experimental and theoretical studies including our research group have manifested that the MI transition of θ-Rb*M'* is caused by the charge ordering (CO). θ-Tl*M'* [*M'* = Co, Zn] have the largest dihedral angle among the θ-*MM'* family, which means that theses compounds have the most narrow bandwidth. We examined the Raman and infrared spectra of these two compounds in the temperature range of 50–300 K. Vibrational spectra of θ-TlCo at low temperature suggest the charge ordering with a horizontal stripe accompanying a doubling of the unit cell. θ-TlZn, which has larger dihedral angle, also underwent a charge-ordering phase transition at T_{CO} = 165 K. Different from θ-RbZn and θ-TlCo, however, the fluctuation of CO appeared at 240 K, which is much higher than T_{CO}. Furthermore, the pattern of ν_3 modes is quite different from θ-RbZn, which reflects the structure-sensitive nature of ν_3 modes, which was claimed in our preceding paper.[2)]

References

1) H. Mori, S. Tanaka and T. Mori, *Phys.Rev.B* **57**, 12023 (1998).

2) K. Yamamoto, K. Yakushi, K. Miyagawa and K. Kanoda, *Phys.Rev.B* **65**, 085110 (2002).

IV-A-4 High-Pressure Raman Study on a 1/3-Filled System (BEDT-TTF)$_3$CuBr$_4$

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While most CT salts showing charge ordering (CO) belong to 1/4-filled system, the semiconducting phase of the 1/3-filled system (BEDT-TTF)$_3$CuBr$_4$ is identified as the CO state by the X-ray diffraction and Raman measurements. In the present study, we investigated the critical behavior of the charge distribution across the phase boundary between the CO and pressure-induced metallic phases. Figure 1 shows the pressure dependence of the Raman spectrum measured at 150 K. The spectrum showed four bands $\mathbf{a_1}$, $\mathbf{a_2}$, $\mathbf{b_1}$, and $\mathbf{b_2}$ ($\mathbf{a_2}$ and $\mathbf{b_1}$ are almost degenerated at ambient pressure). Based on the polarization dependence, we have assigned $\mathbf{a_1}$ and $\mathbf{a_2}$ to ring C=C based stretching-mode ν_2 (a_g). Note that the peak separation of $\mathbf{a_1}$ and $\mathbf{a_2}$ is gradually reduced as the pressure is increased. Then, the two bands are almost merged into one band close to 1 GPa around which the DC conductivity changes into metallic behavior. Since the frequency of ν_2 shifts roughly in proportional to the molecular charge, the approaching of the two bands manifests that the charge disproportionation is continuously suppressed under the application of pressures. It should be important to distinguish whether the pressured-induced phase transformation is a phase transition or crossover phenomena.

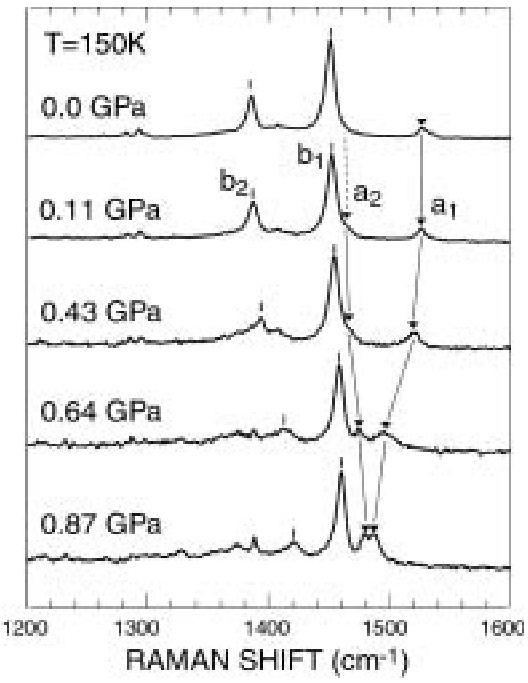

Figure 1. Pressure dependence of the Raman spectrum of (ET)$_3$CuBr$_4$. The exciting (λ_{ex} = 780 nm) and scattered lights are polarized along the *a** and *c* axes.

IV-A-5 Infrared and Raman Studies of the Charge Ordering in the Organic Semiconductor κ-[(Et)$_4$N](ET)$_4$Co(CN)$_6$·3H$_2$O

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[*Macromolecular Symposia* to be published]

Polarized infrared reflectance and Raman spectra of the chare transfer salt κ-[(Et)$_4$N](ET)$_4$Co(CN)$_6$·3H$_2$O were measured as a function of temperature. The salt undergoes a phase transition at *T* = 150 K which is related to a charge ordering inside the conducting ET layers. The charge ordering has a considerable influence on vibrational as well as electronic spectra. New vibrational bands related to ET$^+$ cations are seen below 150 K. Moreover, formation of a new energy gap (charge gap) in electronic excitation spectrum is observed.

IV-A-6 Charge-Ordering and Magnetic Phase Transitions in θ-(BDT-TTP)$_2$Cu(NCS)$_2$

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[*Phys. Rev. B* to be published]

The charge ordering (CO) in organic charge-transfer (CT) salts draws much attention, since the theoretical studies suggest the relation between the charge fluctuation and the pairing mechanism in superconductivity (SC). The experimental study on charge ordering of organic CT salts has been conducted most intensively in θ-type ET salts. The θ-ET salt is a quarter-filled quasi-two-dimensional (Q2D) system without a dimerized ET unit. In non-dimerized θ-ET salts, the charge localization is accompanied by charge disproportionation (CD) and generates charge-rich and charge-poor sites such as ($0.5+\delta$, $0.5-\delta$). We have discovered such CO phase transition at T_{CO} ~ 250 K in non-dimerized Q2D system, θ-(BDT-TTP)$_2$Cu(NCS)$_2$.$^{1)}$ The CO pattern in a variety of ET-based CT salts was theoretically considered. In the framework of mean-field theory, Seo examined the three types of CO patterns, the horizontal, diagonal, and vertical stripes. In this context, the magnetic property gives some information about the CO pattern. The magnetic susceptibility of 30 K < T < 300 K follows the Curie-Weiss law with Curie constant of 0.154 emuK/mol and Weiss temperature of –29 K. We roughly estimate the exchange interaction between the localized charges as $|J|$ = 8 K. The single-crystal ESR experiment shows a magnetic phase transition to a spin-singlet state at 5 K. We examined the selection rule of the polarized Raman spectrum. Both the Raman spectra and magnetic properties suggest the vertical stripe along the b-axis. In addition to the pattern of CO, we discussed the following three subjects. (1) Through the analysis of the ν_2 modes, we presented the parameter CD ratio δ as 0.4. (2) The linewidth of ν_2 suggested the charge fluctuation above T_{CO}. (3) The structural fluctuation for the doubling of the unit cell was found at 200 K.

Reference

1) J. Ouyang, K. Yakushi, Y. Misaki and K. Tanaka, *Phys. Rev. B* **63**, 054301 (2001).

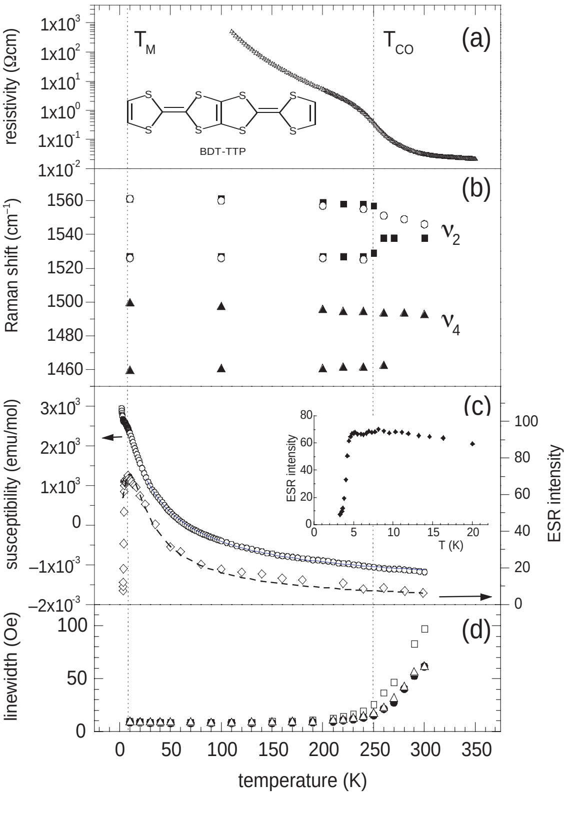

Figure 1. (a) Temperature dependence of resistivity. (b) Temperature dependence of the frequency of ν_2 (solid square) in the ($c,c+b$) spectrum, ν_2 (open circle) in ($a,a+b$), and ν_4 (solid triangle) in ($a,a+b$). (c) Magnetic susceptibility and ESR intensity. The solid and broken lines are respectively the best-fit curves of the Curie-Weiss model and the Bonner-Fisher model with J = –7.9 K. The low-temperature region of ESR intensity is expanded in the inset. (d) ESR linewidth. Open squares, open triangles, and solid circles respectively correspond to the signals of $H||a^*$, $H||c$, and $H||b$. T_{CO} and T_M represent the charge-ordering and magnetic phase transition temperatures, respectively.

IV-A-7 Spectroscopic Studies of Charge-Ordering System in Organic Conductors

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[*Macromolecular Symposia* to be published]

We reviewed our infrared and Raman studies of charge-ordering phase transitions in organic charge-transfer salts. In molecular conductors, the molecular orbital barely overlaps with those of neighbor molecules. Therefore, the transfer integral (t), which contributes to the delocalization of charge, is usually smaller than the on-site (U) and off-site (V) Coulomb interactions that contribute to the localization the charges. Strong correlation effect originates from the comparative magnitude among t, U, and V, and thus many molecular conductors are located at the boundary between a metal (delocalized state) and insulator (localized state). When the charge is localized, the charge often induces a charge disproportionation (CD) and eventually generates an inhomogeneous charge distribution. This localized state is called as a charge-ordered (CO) state, since the localized charges often form a new periodic structure. It is well known that the frequency of some C=C stretching modes, for example, in BEDT-TTF show downshift depending upon the degree of oxidation of the molecule. Using the C=C stretching mode as a probe of the oxidation state, we investigate the CO phase transition. We first introduce θ-(BEDT-TTF)$_2$$MM$'(SCN)$_4$ (M = Rb, Cs, Tl; M' = Zn, Co) as typical examples of a charge-ordering system. We apply the same spectroscopic technique to α'-(BEDT-TTF)$_2$IBr$_2$, θ-(BDT-TTF)$_2$Cu(NCS)$_2$, and (TTM-TTP)I$_3$, which show the phase transitions from low-resistivity state to high-resistivity state.

IV-A-8 Charge and Molecular Arrangement in (DI-DCNQI)$_2$Ag Studied by Vibrational Spectra

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The quasi-one-dimensional (Q1D) radical anion salt (DI-DCNQI)$_2$Ag (DI-Ag) (Figure 1a) shows antiferromagnetic (AF) ordering below 5.5 K, in contrast to the ordinary Q1D systems that fall down to a spin-singlet state. NMR study proposed that the charge localization (CL) in DI-Ag is the first example of the Wigner crystallization in organic charge-transfer salts. While this proposal has stimulated much interest in the charge-ordering (CO) phenomenon in various strongly correlated systems, no direct evidence has been reported so far for the emergence of CO in the present material. We have suggested *via* infrared reflectance study that the molecular arrangement is dimerized (short-range order) from room temperature, although the proposed model is based on a uniform molecular stacking. To investigate the charge distribution as well as the molecular arrangement, we measured the temperature dependence of the Raman spectrum.

Figure 1b shows the temperature dependence of the Raman spectrum. Two lines **a$_1$** and **b$_1$** found at room temperature are attributed to a_g type of C=C and C=N stretching modes, respectively. On lowering temperature, the spectrum exhibited additional bands **a$_2$** and **b$_2$** attributable to the vibronic bands of **a$_1$** and **b$_1$**. We confirmed that the infrared signals of the C=C and C=N modes appeared at different frequencies from these Raman bands. The present result indicates that the dimerized molecular stack (4k_F bond-order wave (BOW)) transforms into a tetramerized one (2k_F BOW) with an effective inversion center at low temperatures. Hence, the present result suggests that the AF-CL phase should be the 2k_F BOW state, rather than the 4k_F CDW state (Wigner crystal).

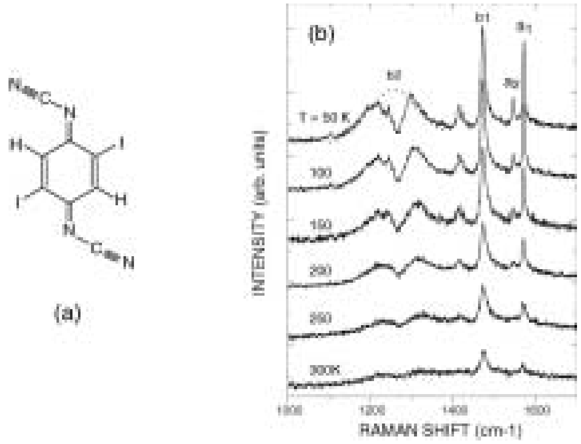

Figure 1. (a) Molecular structure of DI-DCNQI. (b) Temperature dependence of the Raman spectrum measured at ambient pressure. (λ_{ex} = 633 nm) Dotted line shows guide for eyes.

IV-A-9 Charge Distribution and Molecular Arrangement in the Pressure-Induced Metallic Phase of (DI-DCNQI)$_2$Ag

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[*Synth. Met.* submitted]

In contrast to the NMR study which suggests the Wigner-crystal-like charge-ordered state (CO), our previous infrared and Raman study has suggested the important role of lattice modulation rather than the Coulomb interaction for the charge localization (CL) in DI-Ag. It has been confirmed in this compound that the localized charge is released to be metallic at moderate pressures (*ca.* 10 kbar). So the high-pressure experiment will provides crucial data to elucidate the localized state. In the present study, we measured high-pressure Raman spectrum using a sapphire-anvil cell to investigate the pressure dependence of the charge-sensitive C=C and C=N stretching modes.

Figure 1 shows the Raman spectrum observed at 100 K under various pressures. The split phonon bands, **(a$_1$**, **a$_2$**) and **(b$_1$**, **b$_2$**), are respectively assigned to the stretching modes of C=C and C=N. Under the application of pressure, **a$_2$** and **b$_2$** were readily suppressed and almost completely disappeared at *ca.* 1 GPa. Note that **a$_2$** and **b$_2$** do not merge with their parent peaks **a$_1$** and **b$_1$** when they are suppressed. This result indicates that no detectable change occurs in the charge distributions between the localized and metallic states. This result supports our previous conclusion: The localized state of DI-Ag is not regarded as the Wigner crystal but the 2k_F BOW state coexisting with SDW.

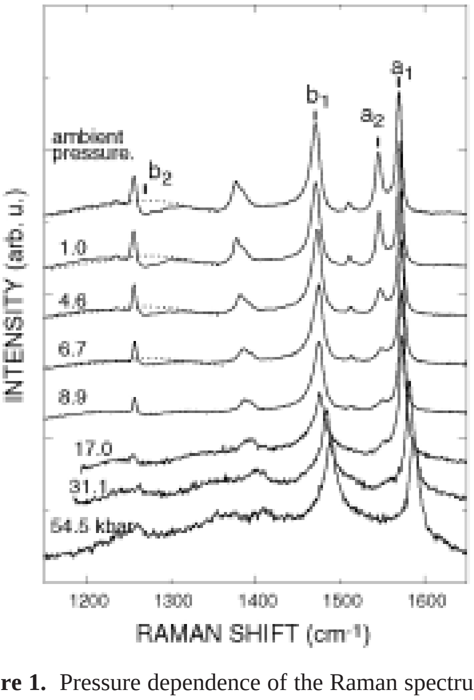

Figure 1. Pressure dependence of the Raman spectrum of DI-Ag measured at 100 K. (λ_{ex} = 780 nm)

IV-A-10 Study of the Phase Transitions of (DI-DCNQI)$_2M$ (M = Ag, Li, Cu) through the Analysis of the Temperature Dependent Vibronic and Vibrational Infrared Absorptions

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[*J. Solid State Chem.* in press]

The temperature dependent phase transitions of 2,5-DI-DCNQI (diiodo-dicyanoquinon-diimmine) title compounds are analyzed by looking at the BOW (bond order wave) and at the CDW (charge density wave) which characterizes the transitions. The analysis is done by studying temperature dependent powder infrared spectra which allow one to have a separate evaluation of both types of contribution to the phase transition of these molecular metals. A model for the spectroscopic infrared features related to the transitions is reported and used for the analysis. It is found that all the title compounds develop a BOW structure at low temperature with characteristics which recall an instability toward a tetramerized $2k_F$ periodicity particularly for the Ag and Li compounds. An appreciable CDW amplitude contribution to the transitions is not found in the spectra.

IV-A-11 Development of a High-Pressure Cell for Raman Measurement Using Sapphire Anvil

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[*J. Spectrosco. Soc. Jpn.* **51**, 72 (2002)]

Diamond is most widely used as anvils for a high-pressure cell. However, the strong Raman band at 1332 cm^{-1} of diamond disturbs our Raman study of charge ordering in organic conductors. We can avoid this serious problem by using sapphire anvils, which has been often adopted for the anvil of high-pressure cell. Because sapphire is more brittle than diamond, sapphire anvils are sometimes broken before the pressure is elevated to several GPa. Although a soft gasket can relieve the strain on the anvil surface, the aperture in such gaskets is easily distorted. To prevent the distortion of aperture and the strain on sapphire surface at the same time, we have prepared the following coated gasket. The body of the gasket was made of stainless steel (Inconel 600) plate. The hard plate was covered with copper by means of electroplating. We performed high-pressure Raman measurement using the copper coated Inconel gasket (0.3 mm thick, ϕ 0.5 mm aperture) with sapphire anvils (1.5 mm height, ϕ 1.2 mm culet). Figure 1 of IV-A-9 shows the resultant Raman spectrum measured from the charge-transfer salts DI-Ag. It demonstrates that the coated gasket enables us to generate pressures over *ca.* 5 GPa without any special modifications for the pressure cell that is designed originally for diamond anvils.

IV-A-12 Optical Study of Two-Dimensional Organic Metal $(EO\text{-}TTP)_2AsF_6$ (EO-TTP=2-(4,5-ethylenedioxy-1,3-dithiol-2-ylidene)-5-(1,3-dithiol-2-ylidene)-1,3,4,6-tetrathiapentalene)

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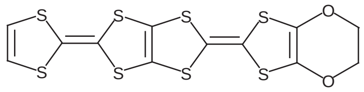
([1]IMS and Yoffe Inst. Phys.; [2]Kyoto Univ.)

[*J. Solid State Chem.* in press]

The charge-transfer salts of organic electron donor molecules involving TTP skeleton provide an enormous number of stable metals down to low temperature. EO-TTP belongs to the metallic BDT-TTP family with an asymmetric structure, in which ethylenedioxy group is substituted at one of the five member rings. Polarized reflectance spectra were measured on the conductive (010) plane of metallic $(EO\text{-}TTP)_2AsF_6$ single crystal. At room temperature, well-defined plasma edges appeared in both directions parallel (E||*a*) and perpendicular (E⊥*a*) to the molecular stack. It was found from the Drude analysis that the effective mass ratio of $(EO\text{-}TTP)_2AsF_6$ was 4 times larger than that of $(TMTSF)_2\text{-}PF_6$ and half of $\beta\text{-}(BEDT\text{-}TTF)_2I_3$ which has a closed Fermi surface. The intra- and inter-stack transfer integrals were estimated from the plasma frequencies in the framework of tight-binding model. Based on these transfer integrals, we suggested that the Fermi surface was open in the k_c direction. Using a generalized Drude model, we obtained the frequency dependence of relaxation rate, which conformed to $\gamma(\omega) = \gamma_0 + b\omega^2$ in the frequency range of 1800–5500 cm^{-1}. This result suggests the view that $(EO\text{-}TTP)_2AsF_6$ has exceptionally weak electron-electron and electron-phonon interaction among organic conductors.



EO-TTP

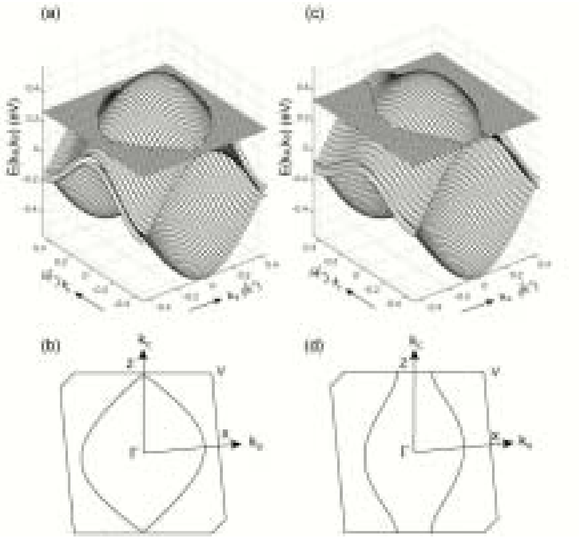


Figure 1. Energy dispersion $E(k_a,k_c)$ in a reciprocal cell and Fermi surface in a Wigner-Seitz cell: (a) $E(k_a,k_c)$ and (b) Fermi surface calculated using the theoretically calculated transfer integrals; (c) $E(k_a,k_c)$ and (d) Fermi surface calculated using the experimentally obtained transfer integrals.