I-R Dimensional Crossovers and Randomness Effects in Quasi-One-Dimensional Organic Conductors

Electronic states in pure one dimension and those in two or three dimensions are very different from each other. In many organic conductors and copper oxides, applying external or chemical pressure controls dimensionality. The induced dimensional crossovers are classified by what kind of transverse coherence is restored by increasing dimensionality, i.e., one-particle coherence or two-particle coherence. Which coherence is restored depends on the asymptotic property of the corresponding one-dimensional system. The Tomonaga-Luttinger liquid is known to be unstable against interchain hopping since the transverse one-particle coherence is easily restored. In other one-dimensional phases, however, the transverse one-particle process is strongly suppressed by electron correlation, and then the transverse two-particle coherence is relatively easily restored. In the quasi-one-dimensional organic conductor, $(TMTTF)_2X$, the charge gap suppresses the transverse one-particle coherence and the two-particle crossover is accompanied by the antiferromagnetic transition. The details are investigated by the perturbative renormalization-group method from high to low temperatures and by the density-matrix renormalization-group method for the ground states. We also study the effects of random potential scattering on the competition among the Mott insulator, the Anderson localization, and the Fermi liquid phases, all of which are realized in $(DI\text{-}DCNQI)_2Ag_{1-x}Cu_x$ and $(DMe\text{-}DCNQI)_2Li_{1-x}Cu_x$.

I-R-1 Spin-Density-Wave Phase Transitions in Quasi-One-Dimensional Dimerized Quarter-Filled Organic Conductors

KISHINE, Jun-ichiro; YONEMITSU, Kenji

[*J. Phys. Soc. Jpn.* **68**, 2790 (1999)]

We have studied spin density wave (SDW) phase transitions in dimerized quarter-filled Hubbard chains weakly coupled via interchain one-particle hopping, t_{b0}. It is shown that there exists a critical value of t_{b0}, t_b^*, between the incoherent metal regime ($t_{b0} < t_b^*$) and the Fermi liquid regime ($t_{b0} > t_b^*$) in the metallic phase above the SDW transition temperature. By using the 2-loop perturbative renormalization-group approach together with the random-phase-approximation, we propose a SDW phase diagram covering both of the regimes. The SDW phase transition from the incoherent metal phase for $t_{b0} < t_b^*$ is caused by growth of the intrachain electron-electron umklapp scattering toward low temperatures, which is regarded as pre-formation of the Mott gap. We discuss relevance of the present result to the SDW phase transitions in the quasi-one-dimensional dimerized quarter-filled organic conductors, $(TMTTF)_2X$ and $(TMTSF)_2X$.

I-R-2 Phase Transitions from Incoherent and from Coherent Metal Phases in Quasi-One-Dimensional Organic Conductors

KISHINE, Jun-ichiro; YONEMITSU, Kenji

[*J. Low Temp. Phys.* in press]

We study effects of a preformed Mott gap and dimensionality on interchain one-particle coherence and spin-density-wave phase transitions in weakly coupled, dimerized quarter-filled Hubbard chains. A phase diagram is given, based on the two-loop perturbative renormalization-group (RG) approach together with the random phase approximation. Feedback effects of interchain processes on the umklapp process are examined by the $1 + \varepsilon$ expansion. We discuss relevance of the present result to the SDW phase transitions in the quasi-one-dimensional dimerized quarter-filled organic conductors, $(TMTTF)_2X$ and $(TMTSF)_2X$.

I-R-3 Quantum Phase Transition and Collapse of Mott Gap in $d = 1 + \varepsilon$ Dimensional Half-Filled Hubbard Systems

KISHINE, Jun-ichiro

[*Recent Prog. Many-Body Theories* **3** submitted]

A correlation-induced charge gap (Mott gap) plays the key role on physical properties of low-dimensional strongly correlated electron systems such as organic conductors or high-T_c cuprate superconductors. In this paper, we study dimensionality effects on the gap in $d = 1 + \varepsilon$ continuous dimension, where $0 < \varepsilon \ll 1$. By solving the one-loop renormalization-group equations, we found a quantum phase transition at a critical dimension, d_c, which depends on the strength of the Hubbard repulsion, U. For $d < d_c$, the Mott gap opens at the fixed point, $g_{3;\infty} = -G_\infty = \infty$, while for $d > d_c$, the Mott gap collapses at the fixed point, $g_{3;\infty} = const.$, $G_\infty = 0$. This result indicates that the Mott gap decreases with raising dimensionality.

I-R-4 Charge Gap and Interchain Correlation in Quasi-One-Dimensional Dimerized Organic Conductors

YONEMITSU, Kenji

[*Mol. Cryst. Liq. Cryst.* in press]

We study the relation between the charge gap and the interchain one-body correlation function in quasi-one-dimensional dimerized organic conductors at quarter filling, by applying the density matrix renormalization group method to a three-chain extended Hubbard model. The charge gap increases with the degree of dimerization in the intrachain hopping integrals. When the charge gap is larger than the interchain hopping integral, the interchain hopping correlation is strongly suppressed, as observed in the $(TMTTF)_2X$ salts.

I-R-5 Magnetic and Pairing Correlation Functions and Interchain Coherence in Quasi-One-Dimensional Dimerized Organic Conductors

YONEMITSU, Kenji

[*J. Low Temp. Phys.* in press]

We study how magnetic and pairing correlation evolves with an increasing interchain hopping integral and decreasing dimerization of intrachain hopping integrals, by applying the density matrix renormalization group (DMRG) method to a three-chain extended Hubbard model at quarter filling for quasi-one-dimensional organic conductors, $(TMTTF)_2X$ and $(TMTSF)_2X$. Magnetic correlation changes from weakly coupled chains of large-amplitude spin density waves to an interchain-coherence-developed spin density wave. Pairing correlation increases, though it still decays exponentially owing to a charge gap for parameters considered here.

I-R-6 Effects of Dimerization on Spin, Charge and Hopping Correlation Functions in Quasi-One-Dimensional Organic Conductors

YONEMITSU, Kenji

[*Physica B* in press]

In order to see the effects of dimerization, interchain hopping, and nearest-neighbor repulsion on various one- and two-body correlation functions in quasi-one-dimensional organic conductors, $(TMTTF)_2X$ and $(TMTSF)_2X$, we apply the density matrix renormalization group method to a three-chain extended Hubbard model at quarter filling. The hopping correlation function shows that the interchain one-particle coherence is strongly suppressed when a charge gap due to the dimerization is larger than the interchain hopping integral. Meanwhile, the spin-spin and charge-charge correlation functions show that the interchain particle-hole coherence is rather insensitive. The charge-charge correlation function is more sensitive to the nearest-neighbor repulsion than the charge gap.

I-R-7 Interplay of Correlation, Randomness and Dimensionality Effects in Weakly-Coupled Half-Filled Random Hubbard Chains

KISHINE, Jun-ichiro; YONEMITSU, Kenji

[*Mol. Cryst. Liq. Cryst.* in press]

We study interplay of electronic correlation, randomness and dimensionality effects in half-filled random Hubbard chains weakly coupled via an interchain one-particle hopping. Based on the two-loop renormalization-group approach, phase diagrams are given in terms of temperature *vs.* strengths of the intra-chain electron-electron umklapp scattering, the random scattering and the interchain one-particle hopping. For strong umklapp scattering and weak interchain hopping, the antiferromagnetic phase is replaced by the Anderson localization phase with increasing random scattering. For weak umklapp scattering and strong random scattering, the Anderson localization phase is replaced by the Fermi liquid phase with increasing interchain hopping. For strong umklapp scattering and weak random scattering, the antiferromagnetic phase is replaced by the Fermi liquid phase with increasing interchain hopping.

I-R-8 Dimensionality Effects in Half-Filled Random Hubbard Chains

KISHINE, Jun-ichiro; YONEMITSU, Kenji

[*Physica B* in press]

We discuss interplay of randomness, electron correlation and dimensionality effects in half-filled random Hubbard chains. Low-temperature phases are given based on the two-loop renormalization group (RG) analysis. Feedback effects of interchain processes on the umklapp process are examined by the $1 + \varepsilon$ expansion. We comment on relevance of the present result to a phase diagram of a doped organic compound, $(DI\text{-}DCNQI)_2Ag_{1-x}Cu_x$.

I-S Competition among Different Charge and Lattice Ordering States in One-Dimensional Metal Complexes

The quasi-one-dimensional halogen-bridged binuclear metal complexes (MMX chains) have strong electron-lattice coupling and electron-electron interaction. They have various electronic phases. Recently, a metallic phase has been found for the ligand dta. Meanwhile, for the ligand pop, a new insulator phase is experimentally suggested in addition to the charge density wave phase. Generally considered, possible electronic phases are i) an averaged-valence (AV) state, ii) a charge-density-wave (CDW) state, iii) a charge-polarization (CP) state, iv) an alternate-charge-polarization (ACP) state, and v) a bond-charge-density-wave state. To understand the competition among these electronic states, we need to include the on-site, two kinds of nearest-neighbor, and next-nearest-neighbor repulsion strengths between electrons in M *d* orbitals, as well as two types of electron-lattice couplings. To reproduce both of the electronic phases for the ligand dta and those for the ligand pop, we need two types of elastic couplings among M and X ions. Thus we have several parameters to control the electronic phases. So far, changing these parameters qualitatively reproduces general tendency for the dependence of the electronic state on the interdimer distance. However, the low-temperature phase of $Pt_2(dta)_4I$ has the crystal structure of the ACP phase and considerably large spin susceptibility, which remain mysterious and need further investigations.

I-S-1 Numerical Studies of Ground State Phase Diagrams for the MMX Chains

KUWABARA, Makoto; YONEMITSU, Kenji

[*Mol. Cryst. Liq. Cryst.* in press]

We study ground state phase diagrams for the MMX chains, using a one-dimensional dimerized 3/4-filled extended Hubbard-Peierls model with site diagonal and off-diagonal electron-lattice interactions. The ground states are obtained mainly in the Hartree-Fock approximation, and their accuracy is checked by the exact diagonalization of small clusters. We find a new phase in addition to frequently considered phases and compare our results with experimental results. Without electron-electron interactions, the charge-density-wave, alternate-charge-polarization (ACP), and bond-charge-density-wave (BCDW) phases are realized depending on the electron-lattice interactions. As the site off-diagonal electron-lattice interaction increases, the ACP phase becomes stable. As the site diagonal electron-lattice interaction increases, the BCDW phase becomes stable. The on-site repulsion stabilizes the averaged-valence and charge-polarization phases, while the nearest-neighbor and next-nearest-neighbor repulsion destabilizes them.

I-S-2 Magnetic Property of MMX Chains as Dimerized Quarter-Filled Systems

KUWABARA, Makoto; YONEMITSU, Kenji

[*Physica B* in press]

We study ground state phase diagrams for the MMX chains, using a one-dimensional dimerized 3/4-filled extended Hubbard-Peierls model with site diagonal and off-diagonal electron-lattice interactions. The ground states are obtained both in the Hartree-Fock approximation and by the exact diagonalization. It has been experimentally shown that the electronic structure of $Pt_2(CH_3CS_2)_4I$ is the alternate-charge-polarization (ACP) phase below 80K and has large magnetic susceptibility at low temperatures. Our results show the ACP phase is stable, if the site off-diagonal electron-lattice interaction, α, is strong enough, for any set of electron-electron interaction strengths. This phase is analogous to the spin-Peierls state. The spin gap is reduced as α becomes small because the lattice distortion becomes weak and the alternation of exchange interactions decreases. A possibility for the large magnetic susceptibility is that the ACP state is realized in the vicinity of the averaged-valence (AV) or charge-polarization (CP) phase. The boundary between the ACP phase and the AV or CP phase is sensitive to the interdimer-nearest-neighbor repulsion.

I-S-3 Structural and Magnetic Transitions in Quasi-One-Dimensional Halogen-Bridged Binuclear Metal Complexes

KUWABARA, Makoto; YONEMITSU, Kenji

[*Mol. Cryst. Liq. Cryst.* submitted]

We investigate ground state phase diagrams for the MMX chains, using a one-dimensional dimerized 3/4-filled extended Hubbard-Peierls model with site diagonal (α) and off-diagonal (β) electron-lattice interactions. Possible electronic states include an averaged-valence (AV) state, a charge-density-wave (CDW) state, a charge-polarization (CP) state, and an alternate-charge-polarization (ACP) state. The ground state can be controlled by choosing the metal (M), bridging halogen (X), ligand, and counter ion. The variation of X affects the model parameters, α, β, the interdimer transfer through X (t_{MXM}), the elastic constant between M and X (K_{MX}) and the interdimer repulsion strengths. As to the competition between the CDW and CP states, the boundary between them shifts upward as K_{MX} increases, and is almost independent of t_{MXM}. These results suggest the possibility of the phase control by changing the halogen ion. The CP state is characterized by coexistence of lattice distortion and gapless spin excitations. Therefore the structural transition between CDW and CP states is accompanied with a magnetic transition.

I-S-4 Electric-Field Response of Exciton in Electroluminescent Polymer

LI, Lei[1]; FU, Rouli[2]; SUN, Xin[1]; YONEMITSU, Kenji
([1]Fudan Univ.; [2]Natl. Lab. Infrared Phys.)

[*Phys. Status Solidi B* **214**, 337 (1999)]

In electroluminescent polymers, the exciton is polarized by a weak electric field E and dissociated by strong E. These effects can quantitatively elucidate the observed dependence of the luminous intensity of the luminescent polymer on the electric field and the field-induced charge generation in conjugated polymers.

I-T Transport and Magnetic Properties of Two-Dimensional Metal-Complex, Organic and Oxide Conductors

Antiferromagnetism and superconductivity are realized in two-dimensional, strongly correlated, organic materials. In κ-(BEDT-TTF)$_2$X with the quarter-filled HOMO band and strong dimerization, the electronic phases are controlled by the ratio of the effective on-site repulsion strength to the bandwidth. The metal-assembled complexes, $Et_nMe_{4-n}Z[Pd(dmit)_2]_2$, has also strong dimerization. In contrast to the former organic materials, they have a rather one-dimensional LUMO band and a two-dimensional HOMO band, both of which are largely split due to the dimerization. The phase diagram is richer than that of κ-(BEDT-TTF)$_2$X in that applied pressure induces a

metal or superconductor phase depending on the cation $Et_nMe_{4-n}Z$. By using a strong-coupling expansion and mapping to an effective model, we find that the electronic phases are controlled by dimensionality and frustration due to the much more complex molecular configuration than that of κ-(BEDT-TTF)$_2$X. For example, for the cation Et_2Me_2Sb, a metallic phase appears under pressure and the antiferromagnetic order is absent because the transfer integral between next-nearest-neighbor dimers are comparable to that between nearest-neighbor ones. In addition to the materials above, many other organic conductors with various degrees of dimerization have been synthesized and the mechanisms of their insulator phases are being studied. To clarify the importance of the long-range electron-electron interactions, we are going to study optical spectra as well. The importance of the long-range electron-electron interactions is clarified also in ab initio molecular orbital, Hartree-Fock, and exact diagonalization studies of (BEDT-TTF) clusters and κ-(BEDT-TTF)$_2$X. Furthermore, the important effect of umklapp scattering processes on the quasiparticle weight is studied in the two-loop renormalization-group analysis.

I-T-1 Possible Magnetic Orders and Cation Dependence of $(Et_nMe_{4-n}Z)[Pd(dmit)_2]_2$

MORI, Michiyasu; YONEMITSU, Kenji; KINO, Hiori[1]
([1]JRCAT)

[*J. Phys. Soc. Jpn.* submitted]

We study the cation dependence of the insulating phase of Pd(dmit)$_2$ salts, based on the dimerized two-band Hubbard model. By using a strong coupling expansion, we obtain an effective Heisenberg model on an anisotropic triangular lattice. Two different magnetic orders are found possible depending on interaction strengths and the energy difference between the two bands. The variation of the cation leads to the variation of the ratio of a transfer integral in one direction to one in another direction, resulting in the variation of the ratios among the magnetic interaction strengths and that of the magnetic frustration. We calculate a sublattice magnetization at zero temperature within the linear spin wave theory and find that the antiferromagnetic order is stable except for the cation Et_2Me_2Sb, where the magnetic frustration is important. Our results are consistent with the ESR measurement.

I-T-2 Role of Dimensionality in Dimerized Two-Band Systems

MORI, Michiyasu; YONEMITSU, Kenji; KINO, Hiori[1]
([1]JRCAT)

[*Mol. Cryst. Liq. Cryst.* in press]

We have studied the ground state properties of Pd(dmit)$_2$ salts using an effective dimer model. This model describes low-energy excitations of the two-band Hubbard model and is derived by a strong coupling expansion. Substituting the cation simultaneously controls dimensionality of the Fermi surface, density-of-states singularity, and magnetic frustration in the dimer model. For the cation Me_4P, the effective Fermi surface has much better nesting property and the effective Fermi level is located in the vicinity of the van Hove singularity. For the cation Et_2Me_2Sb, the effective Fermi surface is rather isotropic and the effective Fermi level is far from the singularity. Such cation dependence implies that the salt with Me_4P (Et_2Me_2Sb) prefers an insulating (metallic) phase, which is consistent with the experiments.

I-T-3 Quasi-One-Dimensional Natures in $(Et_nMe_{4-n}Z)[Pd(dmit)_2]_2$

MORI, Michiyasu; YONEMITSU, Kenji; KINO, Hiori[1]
([1]JRCAT)

[*J. Low Temp. Phys.* in press]

We studied one of possible ground states for Pd(dmit)$_2$ salts in a dimerized two-band Hubbard model. By using a strong-coupling expansion, we obtained an effective dimer model, which has a quasi-one-dimensional Fermi surface. In the insulating phase, it leads to the Heisenberg model on an anisotropic triangular lattice. We calculated effective exchange coupling strengths within the second order perturbation theory and found that both ferromagnetic and antiferromagnetic couplings are possible. Similarities to and differences from Ni(dmit)$_2$ salts are discussed.

I-T-4 Possible Ground State Phases of Pd(dmit)$_2$ Salts

MORI, Michiyasu; YONEMITSU, Kenji; KINO, Hiori[1]
([1]JRCAT)

[*Physica B* in press]

We have studied possible ground state phases of Pd(dmit)$_2$ salts using the two-band Hubbard model. From a strong coupling expansion, an effective model is derived and used to describe low-energy excitations. It is found that mutual interactions and dimensionality of effective bands cooperatively determine the transport and magnetic properties.

I-T-5 Collective Excitations around Charge Ordering States and Coexistent States with Different Orders

MORI, Michiyasu; YONEMITSU, Kenji

[*Mol. Cryst. Liq. Cryst.* submitted]

Novel ground states have recently been studied in molecular conductors. One is the coexistence of a spin density wave (SDW) and a charge density wave (CDW) in $(TMTSF)_2PF_6$, which has a quasi-one-dimensional electronic state and is the quarter-filled system with dimerization. The Hartree calculation for the one-

dimensional extended Hubbard model found the coexistence of SDW and CDW. It is known that not only the on-site but also the nearest-neighbor and the next-nearest-neighbor repulsive interactions are significant. Another is the charge ordering state in θ-(BEDT-TTF)$_2$MM'(SCN)$_4$, which has a rather-two-dimensional electronic state and is the quarter-filled system with dimerization. The Hartree calculation for a two-dimensional extended Hubbard model found various charge ordering states depending on the anisotropy of the transfer integrals and that of the intersite Coulomb interaction strength. In this paper, we study collective excitations around such novel ground states and clarify effects of the dimensionality, the dimerization and the intersite Coulomb interaction on the excitation spectra. First, the ground states of one- and two-dimensional extended Hubbard models are calculated in the fully unrestricted Hartree-Fock (UHF) level to confirm the former results. Next, response functions are calculated in the random phase approximation on the basis of the UHF states to obtain the qualitative features of optical and magnetic excitations. Owing to the intersite Coulomb interaction, excitons appear in the optical spectra. These peaks are sensitive to the dimensionality of the electronic state. The dimerization, the anisotropy of transfer integrals and the intersite Coulomb interaction affect overall feature of collective excitation spectra.

I-T-6 Ab Initio Molecular Orbital, Hartree-Fock, and Exact Diagonalization Studies of Structures and Electronic Phases of (BEDT-TTF) Clusters and κ-(BEDT-TTF) Salts

IMAMURA, Yutaka[1]; TEN-NO, Seiichiro; YONEMITSU, Kenji; TANIMURA, Yoshitaka
([1]GUAS)

[*J. Chem. Phys.* **111**, 5986 (1999)]

Electronic and geometrical structures of bis-(ethylenedithio)tetrathiafulvalene (BEDT-TTF) molecules are studied using ab initio molecular orbital methods. The optimized structure of a BEDT-TTF monomer is close to the experimental one within errors of 0.02 Å and 0.5 degree in bond length and angle, respectively, expect the ethylene group. Ab initio parameters such as transfer integrals and Coulomb interactions are determined from the BEDT-TTF dimer and tetramer calculations. Using model Hamiltonians with the ab initio parameters, we investigate the electronic states based on the exact diagonalization method. The results show that the ground state has antiferromagnetic correlation, which is consistent with experimental results. We study the effects of long-range Coulomb interactions employing the 2-D extended Hubbard model with the Hartree-Fock approximation. It is found that the ground state shows various phases; antiferromagnetic, charge ordering, and paramagnetic ones, controlled by the long-range interactions.

I-T-7 Two-Loop Renormalization-Group Analysis of Two-Dimensional Electron Systems

KISHINE, Jun-ichiro; FURUKAWA, Nobuo[1]; YONEMITSU, Kenji
([1]Aoyama Gakuin Univ.)

[*Recent Prog. Many-Body Theories* **3** submitted]

Recently there has been renewal of interest in understanding two-dimensional (2D) electron systems based on the renormalization-group (RG) framework. In these attempts, possible instabilities occurring in the system have been discussed based on the *one-loop* RG analysis and little attention has been given to the RG flow of the quasiparticle weight that is treated in the *two-loop* level. In this paper, we consider the two-dimensional electron systems where the two-loop RG analysis works well. We take up the following two examples. As the first example, we consider a 2D Fermi surface that consists of flat regions and round-arc regions. We found that the RG flow of the quasiparticle weight depends on the location of the electron wave number, **k**, on the Fermi surface due to the kinematical restriction to the logarithmically divergent processes in the flat regions of the Fermi surface. As the second example, we consider a 2D Fermi surface which touches the Umklapp surface at the 4 points ($\pm\pi/2, \pm\pi/2$). In this case, growth of the umklapp processes between ($\pm\pi/2, \pm\pi/2$) points cause the vanishing quasiparticle weight at these points.