I-P Photoinduced Phase Transitions in Molecular Materials

Photoirradiation may create electrons, holes or excitons, which are often accompanied by local structural deformation. Sometimes it causes spatially large structural transformations with the help of cooperativity possessed by interacting electrons and molecules. Thus, a nonequilibrium phase can be generated, which may not be reached by simply changing temperature or pressure because the energy of a photon is much higher than thermal energies. Such photoinduced phase transitions have been studied extensively, both experimentally and theoretically. Thanks to the great progress in laser spectroscopy techniques, charge and lattice dynamics are being clarified in many molecular materials on different time scales including ultrafast and/or coherent dynamics. We need to treat relevant itinerant-electron models, whose transfer integrals are off-diagonal elements giving transition amplitudes. This is in contrast to stochastic dynamics in classical statistical models, where transition probabilities are determined by the Boltzmann factors at finite temperatures.

I-P-1 Photoinduced Dynamics and Nonequilibrium Characteristics in Quasi-One-Dimensional Electron Systems: Mott Insulators *vs.* Band Insulators

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[*J. Phys.: Conf. Series* **21**, 30–37 (2005)]

Electron-electron interactions play an important role in nonequilibrium properties of molecular materials. First, we show differences between photoinduced ionic-to-neutral and neutral-to-ionic transitions in quasi-one-dimensional extended Peierls Hubbard models with alternating potentials. Cooperative dynamics lead to nonlinear ionicity in the former, while uncooperative dynamics lead to quite linear ionicity in the latter, as a function of the energy supplied from the oscillating electric field. Interchain electron-electron interactions bring about initial competition among metastable and stable domains in neighboring chains, slowing down the phase transition. Interchain elastic couplings are necessary to form a ferroelectric long-range order. Second, we show differences between field-effect characteristics of Mott insulators and those of band insulators in one-dimensional Hubbard models, to which tight-binding models are attached for metallic electrodes and scalar potentials are added for interfacial barriers. Ambipolar characteristics are found in the former, while unipolar characteristics generally appear in the latter. In the former, charge transport is cooperative so that the drain current is insensitive to the difference between the work function of the channel and that of the electrodes, and thus insensitive to the polarity of the gate bias.

I-P-2 Optical Responses of Photoexcited States in the One-Dimensional Ionic Hubbard Model

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[*J. Phys.: Conf. Series* **21**, 183–188 (2005)]

Photoinduced optical responses are studied in the one-dimensional ionic Hubbard model with the nearest-neighbor repulsion V. For $V = 0$, carriers introduced by photoirradiation move freely both in the Mott insulator phase and in the band insulator phase, giving rise to a Drude peak in the optical conductivity spectrum. The carriers in the Mott insulator phase remain conducting for $0 < V < 2t$ because their kinetic energy overcomes the binding energy. By contrast, the electrons and the holes in the band insulator phase are bound to form excitons for $V > 0$, which do not contribute to the charge transport unless the excitation energy allows them to be separated. The dependence of the Drude weight in the lowest-energy photoexcited state on V and the system size is investigated for both phases. Implications to experimental studies of halogen-bridged metal-complex chains are discussed.

I-P-3 Quantum Ising Model Coupled with Conducting Electrons

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[*J. Phys.: Conf. Series* **21**, 232–236 (2005)]

The effect of photo-doping on the quantum paraelectric $SrTiO_3$ is studied by using the one-dimensional quantum Ising model, where the Ising spin describes the effective lattice polarization of an optical phonon. Two types of electron-phonon couplings are introduced through the modulation of transfer integral *via* lattice deformations. After the exact diagonalization and the perturbation studies, we find that photo-induced low-density carriers can drastically alter quantum fluctuations when the system locates near the quantum critical point between the quantum paraelectric and ferroelectric phases.

I-P-4 Photoinduced Metallic Properties of One-Dimensional Strongly Correlated Electron Systems

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[*J. Phys. Soc. Jpn.* **74**, 2671–2674 (2005)]

We study photoinduced optical responses of one-dimensional strongly correlated electron systems. Optical conductivity spectra are calculated for the ground and photoexcited states in a one-dimensional Hubbard model at half filling by the exact diagonalization method. It is found that, in the Mott insulator phase, the photoexcited state has large spectral weights including the Drude weight below the optical gap. As a consequence, the spectral weight above the optical gap is markedly reduced. These results imply that a metallic state is induced by photoexcitation. A comparison between the photoexcited and hole-doped states shows that photoexcitation is similar to chemical doping.

I-P-5 Interchain-Coupling Effects on Photoinduced Neutral-Ionic Transition Dynamics in Mixed-Stack Charge-Transfer Complexes

YONEMITSU, Kenji

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

Effects of interchain electron-electron interactions on the photoinduced ionic-to-neutral and neutral-to-ionic transition dynamics are studied in a quasi-one-dimensional extended Peierls-Hubbard model with alternating potentials for mixed-stack charge-transfer complexes. The ionic-to-neutral transition dynamics depend on the strengths of interchain couplings. For weak couplings, the interchain coherence is destroyed. For strong couplings such as those corresponding to TTF-CA, once neutral domains are nucleated above an increased absorption threshold, they grow spontaneously and cooperatively till the whole system is converted. In contrast, interchain couplings slightly enhance nonlinearity of the otherwise uncooperative neutral-to-ionic transition dynamics.

I-P-6 Theory of Photoinduced Phase Transitions

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[*J. Phys. Soc. Jpn.* in press]

Theories of photoinduced phase transitions have developed along with the progress in experimental studies, especially concerning their nonlinear characters and transition dynamics. At an early stage, stochastic dynamics are explained on the basis of statistical physics. Recently, a variety of dynamics observed in different electronic states are described by relevant electronic models. Especially, coherent motion of a macroscopic domain boundary needs appropriate interactions among electrons and lattice displacements. We describe the history of theories of photoinduced phase transitions and discuss a future perspective.

I-Q Collective Transport through Metal-Insulator Interfaces

Molecular materials are used in many device structures. Charge transport is always through an interface between two materials with different electronic states and work functions. In field-effect transistors fabricated on an insulating material with coherent charge transport under electric fields, the insulator-(source/drain) electrode interface barrier potentials, known as Schottky barriers, play an important role. For band insulators, the Schottky barriers indeed govern the current-voltage characteristics. Because the work function of the electrodes is generally different from that of the material, the characteristics are generally very asymmetric with respect to the polarity of the gate bias and therefore unipolar. Quite recently, ambipolar characteristics are found in field-effect transistor device structures based on organic single crystals of a quasi-one-dimensional Mott insulator. The ambipolar characteristics imply very similar effects on electron and hole injections of the Schottky barriers when combined with electron correlation effects. We need to combine interacting electron models with electrostatic potentials that originate from the long-range Coulomb interaction and are responsible for the interface barrier potentials.

I-Q-1 Mechanism of Ambipolar Field-Effect Carrier Injections in One-Dimensional Mott Insulators

YONEMITSU, Kenji

[*J. Phys. Soc. Jpn.* **74**, 2544–2553 (2005)]

To clarify the mechanism of recently reported, ambipolar carrier injections into quasi-one-dimensional Mott insulators on which field-effect transistors are fabricated, we employ the one-dimensional Hubbard model attached to a tight-binding model for source and drain electrodes. To take account of the formation of Schottky barriers, we add scalar and vector potentials, which satisfy the Poisson equation with boundary values depending on the drain voltage, the gate bias, and the work-function difference. The current-voltage characteristics are obtained by solving the time-dependent Schrödinger equation in the unrestricted Hartree-Fock approximation. Its validity is discussed with the help of the Lanczos method applied to small systems. We find generally ambipolar carrier injections in Mott insulators even if the work function of the crystal is quite different from that of the electrodes. They result from balancing the correlation effect with the barrier effect. For the gate-bias polarity with higher Schottky barriers, the correlation effect is weakened accordingly, owing to collective transport in the one-dimensional correlated electron systems.

I-R Strongly Correlated Electron Systems with Frustrations

Strongly correlated electron systems have produced many exotic phases. Especially in low dimensions, there are always subtle balances between the tendency toward a long-range order through instabilities and the tendency against it due to quantum fluctuations. Geometrical frustrations and/or orbital degrees of freedom enhance quantum fluctuations to lead to intriguing phenomena. Phase diagrams are clarified by paying special attentions to low-energy excitations near quantum critical points.

I-R-1 Frustration-Induced η Inversion in the *S* = 1/2 Bond-Alternating Spin Chain

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[*Phys. Rev. Lett.* **93**, 127203 (4 pages) (2004)]

We study the frustration-induced enhancement of the incommensurate correlation for a bond-alternating quantum spin chain in a magnetic field, which is associated with a quasi-one-dimensional organic compound F_5PNN. We investigate the temperature dependence of the staggered susceptibilities by using the density matrix renormalization group, and then find that the incommensurate correlation becomes dominant in a certain range of the magnetic field. We also discuss the mechanism of this enhancement on the basis of the mapping to the effective *S* = 1/2 XXZ chain and a possibility of the field-induced incommensurate long-range order.

I-R-2 Field-Induced Phase Transitions and Long-Range Orders in the *S* = 1/2 Spin Bond-Alternating Chain with Frustrating Interaction

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[*J. Phys. Soc. Jpn.* **74**, Suppl. 63–66 (2005)]

We study field-induced phase transitions and long-range orders in the *S* = 1/2 spin bond-alternating chain with frustrating interaction. By using the inter-chain mean field theory combined with the finite temperature density matrix renormalization group, we investigate properties of field-induced long-range orders and phase transitions. We find that the inter-chain interaction strongly affects the realized phases of this system.

I-R-3 Field-Induced Incommensurate Order in Frustrated Spin Chain

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[*Prog. Theor. Phys.* submitted]

A mechanism of the incommensurate long-range order induced by external magnetic field is considered to propose its possible realization in the bond-alternating spin chain F_5PNN. Using the density matrix renormalization group analysis, we present several typical phase diagrams, depending on the interchain interaction and the frustration due to the next-nearest-neighbor coupling. A possible magnetization plateau at half the saturation moment is also discussed.

I-R-4 Phase Diagram of the Excitonic Insulator

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[*Physica B* in press]

Motivated by recent experiments, which give strong evidence for an excitonic insulating phase in $TmSe_{0.45}Te_{0.55}$, we developed a scheme to quantitatively construct, for generic two-band models, the phase diagram of an excitonic insulator. As a first application of our approach, we calculated the phase diagram for an effective mass two-band model with long-range Coulomb interaction. The shielded potential approximation is used to derive a generalized gap equation controlling for positive (negative) energy gaps the transition from a semi-conducting (semi-metallic) phase to an insulating phase. Numerical results, obtained within the quasi-static approximation, show a steeple-like phase diagram in contrast to long-standing expectations.

I-R-5 Effective Interaction between the Interpenetrating Kagome Lattices in Na_xCoO_2

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[*Phys. Rev. B* **71**, 214414 (19 pages) (2005)]

A multiorbital model for a CoO_2 layer in Na_xCoO_2 is derived. In this model, the kinetic energy for the degenerate t_{2g} orbitals is given by indirect hopping over oxygen, leading naturally to the concept of four interpenetrating Kagome lattices. Local Coulomb interaction couples the four lattices and an effective Hamiltonian for the interaction in the top band can be written in terms of fermionic operators with four different flavors. Focusing on charge- and spin-density instabilities, a big variety of possible metallic states with spontaneously broken symmetry are found. These states lead to different charge, orbital, spin, and angular momentum ordering patterns. The strong superstructure formation at x = 0.5 is also discussed within this model.

I-R-6 Magnetism in Strongly Correlated and Frustrated Systems

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[*Physica B* **359**, 626–632 (2005)]

In strongly correlated systems, some of the low-energy excitations are enhanced. In geometrically frustrated systems, due to the absence of long-range order, there remains (pseudo-) degeneracy associated with the low-energy excitations at low temperatures. Lifting of degeneracy in both Mott insulating systems and itinerant electron systems are discussed by considering the pyrochlore lattice as an example.