# Nonequilibrium Theory of Conductors

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When low-dimensional correlated electron systems such as organic conductors are placed in nonequilibrium environments, they sometimes show novel phenomena that never appeared in conventional conductors based on rigid bands. One example is found in the current-voltage characteristics and the field-effect characteristics caused by metal-Mott-insulator interfaces. Another example is found in photoinduced phase-transition dynamics, where the induced transient state is not reached by simply changing temperature or pressure. It is possible because the energy of a photon is much higher than thermal energies. Our theoretical researches are focused on the mechanisms of such nonlinear phenomena.

## 1. Suppression of Rectification at Metal–Mott-Insulator Interfaces[1)]

Charge transport through metal–Mott-insulator interfaces is studied and compared with that through metal-band-insulator interfaces. For band insulators, rectification has been known to occur owing to a Schottky barrier, which is produced by the work-function difference.

For Mott insulators, however, qualitative different current-voltage characteristics are obtained. Theoretically, we use the one-dimensional Hubbard model for a Mott insulator and attach to it the tight-binding model for metallic electrodes. A Schottky barrier is introduced by a solution to the Poisson equation. The current density is calculated by solving the time-dependent Schrödinger equation. We mainly use the time-dependent Hartree-Fock approximation and also use exact many-electron wave functions on small systems for comparison. Rectification is found to be strongly suppressed even for large work-function differences.

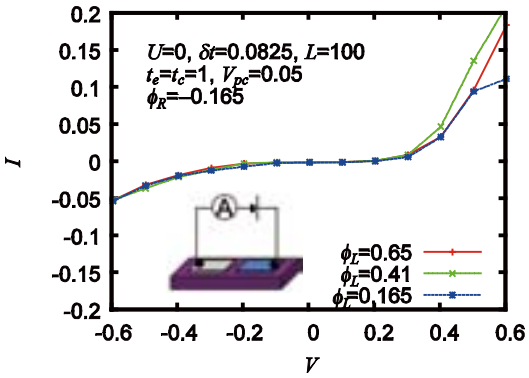


**Figure 1.** Current-voltage characteristics for a band insulator. Rectification is clearly seen.

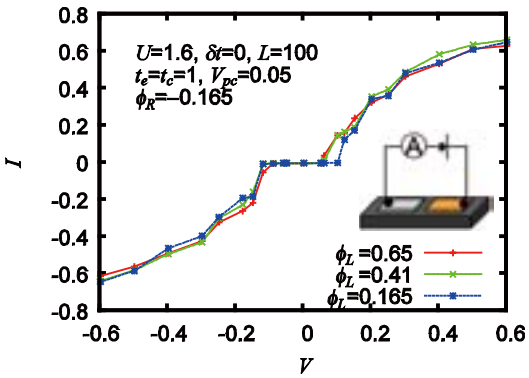


**Figure 2.** Current-voltage characteristics for a Mott insulator. Rectification is suppressed.

Its close relationship is shown with the fact that field-effect injections into one-dimensional Mott insulators are ambipolar. Experimentally, we fabricated asymmetric contacts on top of single crystals of quasi-one-dimensional organic Mott and band insulators. Rectification is strongly suppressed at an interface between metallic magnesium and Mott-insulating (BEDT-TTF)($F_2$TCNQ).

## 2. Charge Order with Structural Distortion in Organic Conductors: Comparison between θ-(ET)$_2$X and α-(ET)$_2$X Salts[2)]

Charge ordering with structural distortion in quasi-two-

dimensional organic conductors θ-(BEDT-TTF)$_2$RbZn(SCN)$_4$ and α-(BEDT-TTF)$_2$I$_3$ is investigated theoretically. We use the Hartree-Fock approximation, strong-coupling perturbation theory, and exact diagonalization of a 3/4-flled extended Hubbard model for an anisotropic triangular lattice. The model includes on-site and inter-site Coulomb interactions together with Peierls-type electron-lattice couplings. We examine the effect of lattice degrees of freedom on charge order. It is found that the experimentally observed, horizontal charge order is stabilized by lattice distortion in both compounds. In particular, the lattice effect is crucial to the realization of the charge order in θ-(BEDT-TTF)$_2$RbZn(SCN)$_4$, while the peculiar band structure of α-(BEDT-TTF)$_2$I$_3$ whose symmetry is lower than that of θ-(BEDT-TTF)$_2$RbZn(SCN)$_4$ is also an important factor in α-(BEDT-TTF)$_2$I$_3$ together with the lattice distortion. For α-(BEDT-TTF)$_2$I$_3$, we obtain a transition from the metallic phase with charge disproportion to the horizontal charge order with lattice modulations, which is consistent with the latest X-ray experimental result.

## 3. Photoinduced Change in the Charge Ordering Pattern in (EDO-TTF)$_2$PF$_6$ with Strong Electron-Phonon Interaction[3)]

The quasi-stable state in the photoinduced phase transition for the quasi-one-dimensional quarter-filled organic conductor (EDO-TTF)$_2$PF$_6$ has been examined by ultrafast reflective measurements and time-dependent model calculations incorporating both electron-electron and electron-phonon interactions. The transient optical conductivity spectrum over a wide probe photon energy range revealed that photo- excitation induced a new type of charge-disproportionate state. Additionally, coherent and incoherent oscillations dependent on probe photon energies were found, as predicted by the calculation.

The photoinduced state has a (1010) charge separation owing to competing long-range e–e and e–anion interactions.

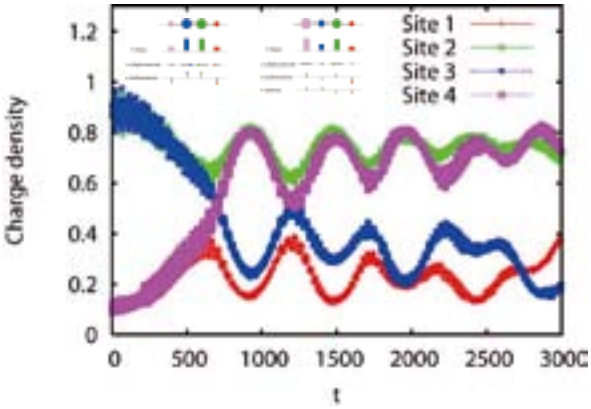


**Figure 3.** Time dependence of charge densities after the (0110)-charge-order ground state is photoexcited to a {0200} state.

On the low-energy side of the photoinduced peak, the transient spectrum shows irregular time dependences. It is because delocalized electrons are observed there, which are scattered by phonons on different molecules and times. Thus, the coherence is easily lost.

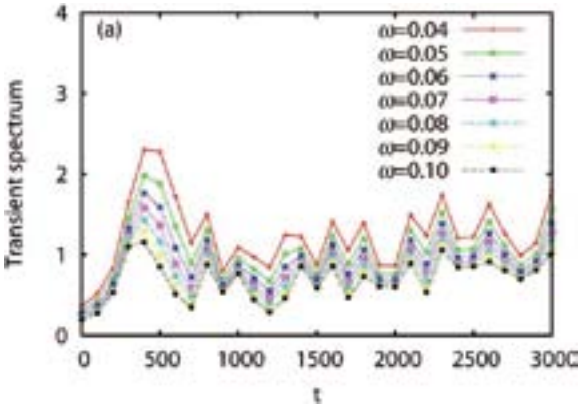


**Figure 4.** Time dependence of transient spectra on the low-energy side of a main peak, after photoexcitation from (0110) to {0200}.

On the high-energy side of the photoinduced peak, the transient spectrum shows coherent oscillations. It is because localized charge-transfer processes are observed there, which are governed by the instantaneous distribution of charge and displacements. The coherence is rather robust.

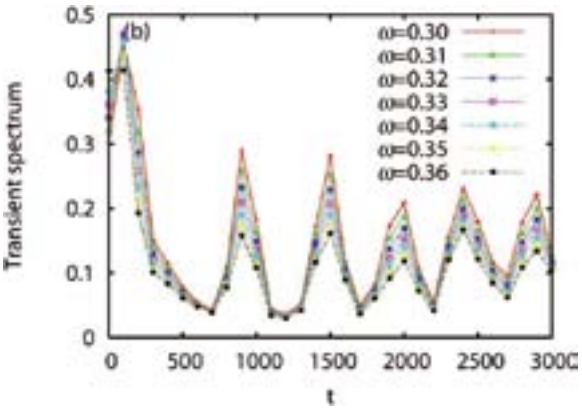


**Figure 5.** Time dependence of transient spectra on the high-energy side of a main peak, after photoexcitation from (0110) to {0200}.

### References

1) K. Yonemitsu, N. Maeshima and T. Hasegawa, *Phys. Rev. B* **76**, 235118 (6 pages) (2007).
2) Y. Tanaka and K. Yonemitsu, *J. Phys. Soc. Jpn.* **77**, 034708 (9 pages) (2008).
3) K. Onda, S. Ogihara, K. Yonemitsu, N. Maeshima, T. Ishikawa, Y. Okimoto, X. -F. Shao, Y. Nakano, H. Yamochi, G. Saito and S. Koshihara, *Phys. Rev. Lett.* **101**, 067403 (4 pages) (2008).

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