EPJ Web of Conferences **41**, 03019 (2013)
DOI: 10.1051/epjconf/20134103019

Coherent Electron Dynamics in 10 fs Time Scale in Organic Charge Ordered and Dimer-Mott Insulators

*S. Iwai[1,2], Y. Kawakami[1], T. Ishikawa[1], Y. Sakurai[1], H. Itoh[1,2], K. Yamamoto[3], and T. Sasaki[2,4]

[1] Department of Physics, Tohoku University, Sendai 980-8578, Japan
*E-mail: s-iwai@m.tohoku.ac.jp
[2] JST, CREST, Sendai 980-8578, Japan
[3] Institute for Molecular Science, Okazaki, 444-8585, Japan
[4] Institute for Materials Research, Tohoku University, Sendai, 980-8577, Japan

Abstract. Coherent oscillations of correlated electrons were detected in the early dynamics of photoinduced melting and/or construction of the organic charge ordered/ferroelectric cluster and the dimer Mott insulator by using 3 optical-cycle infrared pulse.

1 Introduction

Optical responses of highly correlated insulators such as charge ordered (CO) and Mott insulators attract much attention because they exhibits ultrafast changes in the conducting and/ or magnetic natures upon photo-excitations. The layered organic charge transfer salts ET_2X (ET; bis[ehtylenedithio]-tetrathiafulvelene) are the key material for investigating such photoinduced phase transitions (PIPT)[1-3]. Recent progress of several fs ~10 fs laser enables us to capture the coherent dynamics of the correlated electrons. For example, coherent oscillations of the electrons in CO insulator α-$(ET)_2I_3$ play an crucial role in the photoinduced melting of CO[2].

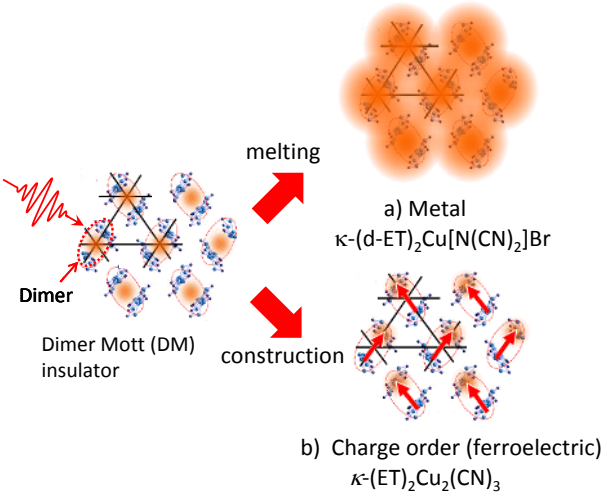

Fig. 1. Schematic illustrations of PIMT in dimer Mott insulators.
a) insulator to metal transition (κ-(d-ET)$_2$Cu[N(CN)$_2$]Br),
b) generation of ferroelectric CO cluster (κ-(ET)$_2$Cu$_2$(CN)$_3$).

Among the series of ET_2X, κ-type ET salts are particularly well known, because they exhibit wide variety of the electronic states such as Mott insulator, superconductivity, spin glass, relaxer ferroelectrics...etc. In κ-type ET salts, adjacent ET molecules are dimerized and a charge is localized on each dimer, as shown in Fig. 1(a), if the effective on-site Coulomb repulsion energy on each dimer is sufficiently greater than the

inter-dimer kinetic energy. Intra-dimer degree of freedom in such dimer-Mott (DM) system results in the phase-rich diagram. In these competing electronic and magnetic phases enables us to expect the various photoinduced melting and reconstruction of the electronic/magnetic orders. Clarification of the coherent electron dynamics in this phase-rich system is of great interest. Here, target materials are the DM insulators κ-(*d*-ET)$_2$Cu[N(CN)$_2$]Br (*d*- represents the deuteration of ET molecule) and κ-(ET)$_2$Cu$_2$(CN)$_3$, which exhibit photoinduced insulator to metal (I-M) transition [2](Fig. 1(a)) and photoinduced growth of CO/ferroelectric cluster (Fig. 1(b))[4, 5], respectively.

2 Experiment

3-cycle 12 fs pulse in the 1.2-1.8 μm wavelength region which was generated in optical parametric amplifier using type I BBO with degenerate configuration and chirped mirror compressor. Pulse width evaluated from the FROG pattern is 12 fs which corresponds to 3-optical cycle. The time resolution of the reflection detected pump-probe measurement is 15 fs [3].

3 Results and discussions

Fig. 2 shows the oscillating component in the time profile of the reflectivity change (ΔR/R) reflecting the primary dynamics of the PIPT. Figs. 2(a), 2(b) and 2(c) show the oscillations of the correlated electrons in the melting of CO (Fig. 2(a); α-(ET)$_2$I$_3$), and the DM (Fig. 2(b); κ-(*d*-ET)$_2$Cu[N(CN)$_2$]Br) insulators, and the generation of the CO-ferroelectric cluster (Fig. 2(c); κ-(ET)$_2$Cu$_2$(CN)$_3$), respectively.

In α-(ET)$_2$I$_3$, the oscillation with the period of 18 fs (Figs. 2(a) and 2(d)) is initially detected at 30 fs. This oscillation is attributable to the coherent motion of the CO electrons as shown in Fig. 3(a), because the spectral future of the oscillation (Fig. 2(d)) shows the gap-like structure which is analogous to the charge gap in the steady state optical conductivity (dashed curve). Then, at 60 fs, anti-resonant-dip is detected at 1400 cm^{-1}(dashed-dotted curve), indicating the Fano interference between the electron and C=C vibration.

In κ-(*d*-ET)$_2$Cu[N(CN)$_2$]Br, observation of the intense 25 fs oscillation (Figs. 2(b) and 2(e)) indicates that the intra-molecular C=C (ν_3, 1300 cm^{-1}) mode is immediately excited through the

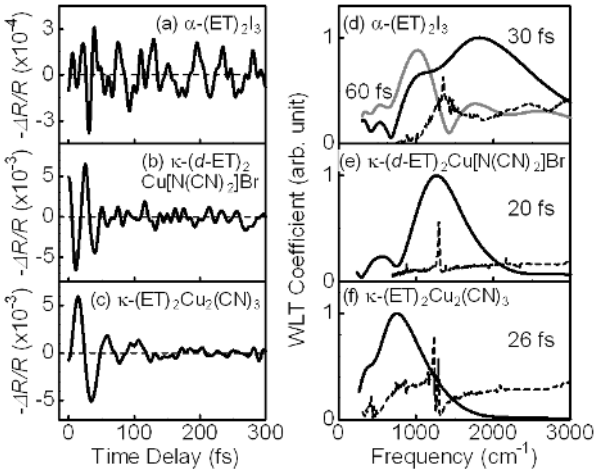

Fig 2. (a)-(c) Oscillating component of ΔR/R and (d)-(e) time resolved spectra which are obtained by the wavalet analysis. Dashed curves represent the steady state optical conductivity spectra.

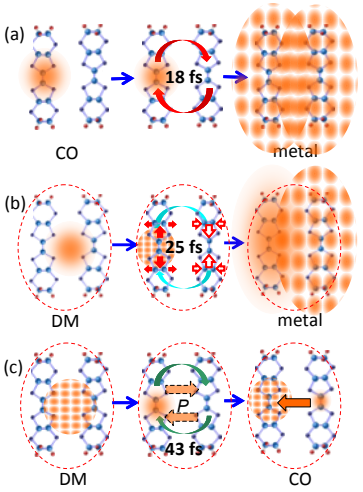

Fig.3. Schematic illustarations of the coherent oscillations. a) α-(ET)$_2$I$_3$, b) κ-(*d*-ET)$_2$Cu[N(CN)$_2$]Br, c) κ-(ET)$_2$Cu$_2$(CN)$_3$.

strong electron-molecular vibration (EMV) interaction in the dimer (Fig. 3b). Then, the intense C=C (ν_3) oscillation causes the nonlinear excitation of lower frequency intramolecular and intermolecular modes leading to the I-M transition [3].

In κ-$(ET)_2Cu_2(CN)_3$, photoinduced growth of the CO-ferroelectric clusters, that is a new type of PIPT, was reported. The energy (800 cm^{-1}) of the 43 fs oscillation (Figs. 2(c)and 2(f)) corresponds to charge gap which is related to the intra-dimer dipole oscillation as show by Fig. 3(c). This fact indicates that the coherence of the intradimer dipole oscillation is important in the growth of the CO/ferroelectric cluster.

According to these results, photoinduced melting of the CO and the Mott insulators are triggered by the coherent electron oscillation (α-$(ET)_2I_3$) and the C=C vibration interacting with the electrons (κ-(*d*-$ET)_2Cu[N(CN)_2]Br$), respectively. On the other hand, photoinduced growth of the CO/ferroelectric state is driven by the coherent dipole oscillation (κ-$(ET)_2Cu_2(CN)_3$). It is noteworthy that these coherent oscillations in the precursory dynamics of the PIPT are strongly dumped within 100 fs~200 fs. Such ultrafast dumping indicates that those modes strongly interact with other modes, i.e., the observed oscillations play essential role in the primary dynamics of the PIPT.

4 Summary

Ultrafast coherent oscillation of correlated electrons and electron-phonon were detected by using 12 fs, 3-optical cycle pulse in the early stage dynamics of the PIPT; photoinduced melting of CO (α-$(ET)_2I_3$), dimer Mott insulator (κ-(*d*-$ET)_2Cu[N(CN)_2]Br$) and the photoinduced growth of CO/ferroelectric cluster (κ-$(ET)_2Cu_2(CN)_3$). The PIPTs are initially triggered by the coherent oscillations of CO electrons in α-$(ET)_2I_3$, C=C vibration interacting with electrons in κ-(*d*-$ET)_2Cu[N(CN)_2]Br$, and the intra-dimer dipole oscillation in κ-$(ET)_2Cu_2(CN)_3$, respectively.

Reference

1. S. Iwai, K. Yamamoto, A. Kashiwazaki, F, Hiramatsu, H. Nakaya, Y. Kawakami, K. Yakushi, H. Okamoto, H. Mori, and Y. Nisho, Phys. Rev. Lett., **98**, 097402 (2007).
2. Y. Kawakami, T. Fukatsu, Y. Sakurai, H. Unno, H. Itoh, S. Iwai, T. Sasaki, K. Yamamoto, K. Yakushi, and K. Yonemitsu, Phys. Rev. Lett., **105**, 246402 (2010).
3. Y. Kawakami, S. Iwai, T. Fukatsu, M. Miura, N. Yoneyama, T. Sasaki, and N. Kobayashi, Phys. Rev. Lett., **103**, 066403 (2009).
4. M. Abdel-Jawad, I. Terasaki, T. Sasaki, N. Yoneyama, N. Kobayashi, Y. Uesu, and C. Hotta, Phys. Rev. B, **82**, 125119 (2010).
5. K. Itoh, H. Itoh, M. Naka, S. Saito, I. Hosako, N. Yoneyama, S. Ishihara, T. Sasaki, and S. Iwai, to be published.