Photoinduced insulating of layered organic metal driven by strong electric field of 1.5-cycle, 7 fs infrared pulse

S. Iwai[1,2*], T. Ishikawa[1], Y. Sagae[1], Y. Naito[1], J. Ichimura[1], Y. Kawakami[1], H. Itoh[1,2], S. Ishihara[1], K. Yamamoto[3], K. Yakushi[4], T. Sasaki[5,2], H. Kishida[6,2], and K. Yonemitsu[7]

[1]Deaprtment of Physics, Tohoku University, Sendai 980-8578, Japan
[2]JST-CREST, Sendai 980-8578, Japan
[3]Deaprtment of Applied Physicse, Okayama Science University, Okayama, 700-0005 Japan
[4]Toyota Physical and Chemical Research Institure, Nagakute, 480-1192, Japan
[5]Institute for Material Research, Tohoku University, Sendai, 980-8577, Japan
[6] Department of Applied Physics, Nagoya University, Nagoya 464-8603, Japan
[7]Department of Physics, Chuo University, Tokyo 112-8551, Japan
**s-iwai@m.tohoku.ac.jp*

Abstract: Photoinduced metal to insulator (M-I) change were realized by strong electric filed (10 MV/cm) of 1.5-cycle (7 fs) near infrared pulse. Threshold for the M-I change decrease near the M-I transition temperature. The M-I change is driven by coherent charge oscillation with a period of 20 fs.

OCIS codes: (320.7120)Ultrafast Phenomena; (320.7130) Ultrafast processes in condensed matter, including semiconductors

1. Introduction

Photoinduced insulator to metal transition (PIMT) or equivalently the melting of the charge ordering (CO) insulators attract much attentions, because they can trigger various ultrafast changes in conducting and/ or magnetic properties. Such ultrafast melting of CO is driven by the immediate changes of the band filling and /or the electron kinetic energy. On the other hand, there are few studies on the reverse process, i.e., the metal to insulator (M-I) change. Considering the thermodynamic nature of the CO, i.e., the entropy for the CO is smaller than that for metal, photoinduced formation of the CO has been considered to be difficult. However, an instantaneous strong electric filed >10 MV/cm which is realized by the few optical-cycle fs laser enables us to modulate the electronic structures directly before the increase of electron temperature.

The layered organic conductor α-$(ET)_2I_3$ (ET; bis[ehtylenedithio]-tetrathiafulvelene) exhibiting ferroelectric CO insulator(Fig. 1(a))-metal(Fig. 1(b)) transition (transition temperature T_{CO}=135 K) has been the key material for investigating PIMT[1, 2]. In this study, we have demonstrated the photoinduced M-I change as shown schematic illustration in Figs. 1(a)(b) by using the 7 fs, 1.5-cycle near infrared pulse. Ultrafast reflectivity change showing the M-I change were discussed in terms of the "dynamical localization" [3-5].

2. Experiment

Super broadband infrared spectrum covering 1.2-2.3 μm was obtained by focusing the idler pulse (1.7 μm, carrier-envelope phase is self-stabilized) from the optical parametric amplifier to the hollow-fiber set in Kr filled chamber. Pulse compression was performed using both active mirror and chirped mirror techniques. Pulse width evaluated from the second harmonic generation autocorrelation is 7 fs which corresponds to 1.5-optical cycle. Instantaneous electric filed on the sample surface (excitation diameter ~160 μm) reaches to > 10 MV/cm.

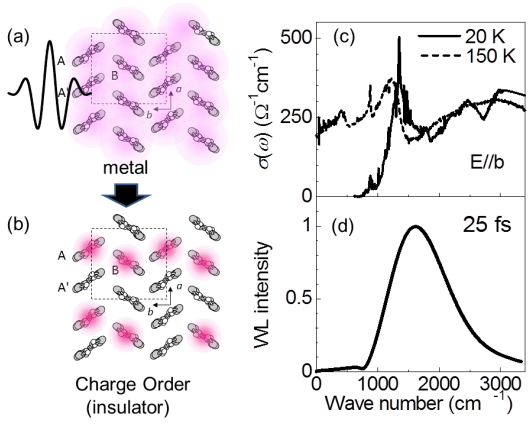

Fig. 1. **(a), (b)**Schematic illustrations of photoinduced change from metal**(a)** to charge ordering (CO) insulator**(b)**. **(c)**Steady state optical conductivity. **(d)** Wavelet analysis of 20 fs oscillation in time profile shown in Fig. 3(See text).

3. Results and discussions

Fig. 2 shows the steady state reflectivity (R) spectrum (Fig. 2(a)), spectral differences between different temperatures (Fig. 2(b)), and transient reflectivity $\Delta R/R$ spectra (Fig. 2(c)), respectively. An increase of R (positive $\Delta R/R$) at 0.6-0.75 eV was observed at 35 fs after the excitation by 7 fs pulse (closed circles and gray shade in Fig. 2(c)), although R decreases in 0.6-0.65 eV at 300 fs, indicating the negative $\Delta R/R$. The spectral shape of $\Delta R/R$ at 30 fs is analogous to the temperature differential spectrum $[R(40\text{K})-R(140\text{ K})]/R(140\text{ K})$ in Fig 2(b), reflecting the decrease of temperature or the M-I change. On the other hand, $\Delta R/R$ spectrum at 300 fs is analogous to the $[R(250\text{K})-R(140\text{ K})]/R(140\text{ K})$, indicating the increase of temperatures. Fig. 3(a) shows the excitation intensity (I_{ex}) dependence of $\Delta R/R$ (138 K, 35 fs). Increase of R is detectable only for I_{ex}>0.5 mJ/cm^2 (6.3 MV/cm), showing the threshold-like behaviour. The negative $\Delta R/R$

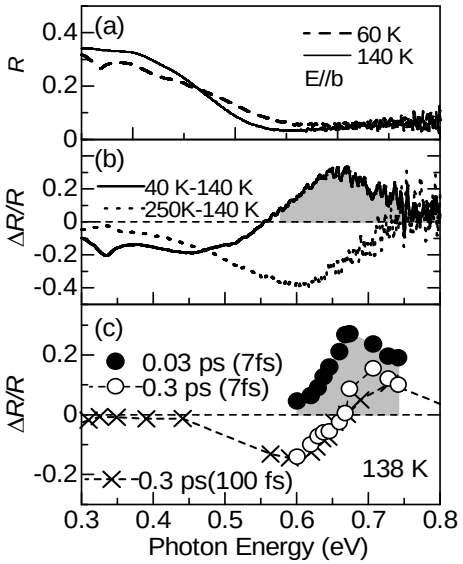

Fig 2. **(a)**Steady state reflectivity spectra (E//b) of α-$(ET)_2I_3$ at 60 K (CO) and 140 K(metal). **(b)** Spectral difference R(40K)-R(140 K)]/R(140 K) (solid curve) and [R(250K)-R(140 K)}/R(250 K) (dashed curve). **(c)** Transient reflectivity ($\Delta R/R$) spectra at 0.03 (closed) and 0.3 ps (open) after excitation by 7 fs pulse. Dashed curve and crosses show the $\Delta R/R$ st 0.3 ps after 100 fs excitation.

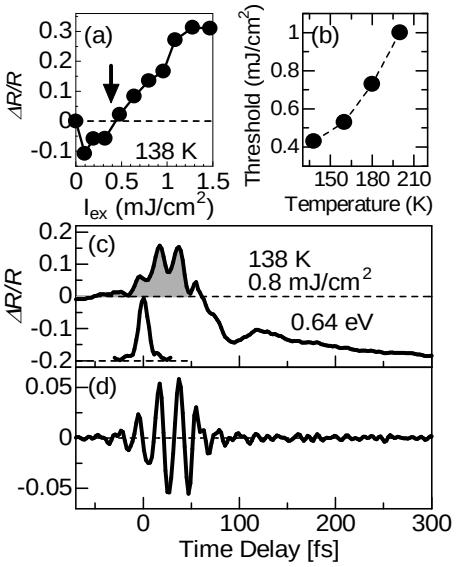

Fig.3. **(a)** Excitation intensity (I_{ex}) dependence of $\Delta R/R$ at 0.64 eV(138 K). The threshold intensity is indicated by the arrows. **(b)** Temperature dependence of the threshold intensity. **(c)** Time evolution of $\Delta R/R$ at 138 K(0.8 mJ/cm^2). The cross correlation profile of the pump and the probe pulse is also shown. **(d)** oscillating component obtained by using Fourier filter (>20 THz).

for I_{ex}<0.5 mJ/cm^2 is attributable to the increase of electron temperature. The threshold intensity (shown by arrows in Fig. 3(a)) decreases near the T_{CO}=135 K as shown in Fig. 3(b). Such threshold behaviour near the CO temperature also supports the interpretation in terms of the photoinduced M-I change.

Figs. 3(c) and 3(d) respectively show the time evolution of $\Delta R/R$ measured at 0.64 eV(Fig. 3(c)) and the high-frequency (> 20 THz) oscillating component(Fig. 3(d)) obtained by Fourier high-pass filter. The oscillation with a period of 20 fs is attributable to the coherent oscillation of the correlated charge[2], because spectral shape obtained by the wavelet (WL) analysis shown in Fig. 1(d) is analogous to that of the inter-molecular charge transfer excitation in optical conductivity (Fig. 1(c)). This coherent charge oscillation is closely related to the M-I change, because the charge oscillation disappears with the decay of the photoinduced insulating state.

Considering that the I-M change can be observed only for I_{ex} > 0.5 mJ/cm^2 (~ 6 MV/cm), the M-I change is induced by the strong electric filed. According to the "dynamical localization" theory [3-5] under strong field, intersite transfer integral (t) is modulated and can be reduced to zero for ~100 MV/cm [5]. The M-I change is considered to occur as a precursory phenomena of the dynamical localization. Small change of t can result in the M-I change ,because t and U, V are competing with each other near the M-I phase boundary in α-$(ET)_2I_3$.

4. Summary

Photoinduced insulating were realized in organic conductor α-$(ET)_2I_3$ by strong electric filed (10 MV/cm) of 1.5-cycle (7 fs) near infrared pulse. This phenomenon can be understood as a precursory phenomena for the dynamical localization effect near the M-I transition temperature.

5. References

[1] S. Iwai, K. Yamamoto, A. Kashiwazaki, F. Hiramatsu, H. Nakaya, Y. Kawakami, Y. Yakushi, H. Okamoto, H. Mori, and Y. Nishio, "Photoinduced Melting of a Stripe-Type Charge-Order and Metallic Domain Formation in a Layered BEDT-TTF-Based Salt", 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, "Early-Stage Dynamics of Light-Matter Interaction Leading to the Insulator-to Metal Transition in a Charge Ordered Organic Crystal", Phys. Rev. Lett. 105, 246402(2010).

[3] D. H. Dunlap and V. M. Kenkre, "Dynamic localization of a charged particle moving under the influence of an electric field", Phys. Rev. B34, 3625(1986).

[4] Y. Kayanuma and K. Saito, "Coherent destruction of tunnelling, dynamical localization and the Landau-Zener formula, Phys. Rev. A77, 010101(2008).

[5] K. Nishioka and K. Yonemitsu, "Intra- and Inter-dimer Transfer Integrals Effectively Modified by Pulsed and Continuous-Wave Lasers for Controlling Charge Transfers in Molecular Crystals", J. Phys. Soc. Jpn. 83, 024706 (2014)