# Spontaneous formation of correlated charge coherence induced by 1.5-cycle pulse in 1-D organic metal $(TMTTF)_2AsF_6$

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**Abstract:** Ultrafast optical response of organic metal $(TMTTF)_2AsF_6$ induced by 1.5 cycle (7 fs) infrared pulse was investigated. Intense coherent oscillation of correlated charge (18 fs) grows in the time scale of 50 fs, reflecting the spontaneous-formation of the electronic coherence before the electronic thermalization .
**OCIS codes:** (320.7120)Ultrafast Phenomena; (320.7130) Ultrafast processes in condensed matter, including semiconductors

## 1. Introduction

Since the first observation of the increase of electron temperature in inorganic metals, ultrafast optical responses in metal have been discussed in terms of the increase of an electron temperature in the framework of two-temperature model [1]. On the other hand, in strongly correlated materials, ultrafast charge dynamics have been investigated in insulators such as Mott insulator and charge order (CO) [2-4], because various ultrafast changes in the conducting and/or magnetic natures can be realized. However, there are few studies on correlated metals. Charge dynamics reflecting the strong electron correlation are expected to be captured before the electronic thermalization is completed.

The organic salt $(TMTTF)_2AsF_6$ (TMTTF; tetramethyltetrathiafulvelene) are well known strongly correlated 1-D metal or Tomonaga-Luttinger liquid at > 250 K (Fig. 1(b)), although this compound shows the ferroelectric CO below 110 K (Fig. 1(a)). In this study, we have observed the ultrafast coherent dynamics reflecting the correlated charge motion before the thermalization of the electron system by using the 7 fs, 1.5-cycle near infrared pulse.

## 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 into the hollow-fiber in Kr filled chamber. Pulse compression was performed using active mirror and chirped mirror. Pulse width evaluated from the second harmonic generation autocorrelation is 7 fs which corresponds to 1.5-optical cycle. Spectrum of the 7 fs pulse for pump and probe are shown as a dotted curve in Fig. 2(a).

## 3. Results and discussions

Fig. 2 shows the steady state reflectivity ($R$)(Fig. 2(a)), and transient reflectivity $\Delta R/R$ (open circles in Fig. 2(b)) at 300 K, respectively. Decrease of $R$ at 0.2-0.75 eV was observed immediately after the excitation by 150 fs, 0.89 eV pulse. As shown in the time profile in Fig. 2(c), a build-up time and the a recovery time of $\Delta R/R$ are in the time scale of <100 fs, and <1 ps, respectively. Such ultrafast responses are attributable to the increase of electron temperature. We could reproduce the time profile, assuming that the electron temperature increases up to 400 K. Here, we extended the two-temperature model to "three temperature model", because contributions from both intra-molecular vibrations (~1500 $cm^{-1}$) and inter-molecular phonons (< 200 $cm^{-1}$) should be considered in the analysis, i.e., we can define the temperatures for electron, intra-molecular vibration and intermolecular-phonon, respectively. The time evolution of $\Delta R/R$ can be reproduced by the analysis. Here, the coefficients of specific heat for electron, for intra-molecular vibration, and for inter-molecular phonon are 0.01042 $mJ/K^2/mol$, 0.64 $mJ/K^2/mol$, 4 $mJ/K^2/mol$, respectively. Coupling constants for electron-vibration and electron-phonon are 200 $meV^2$ and

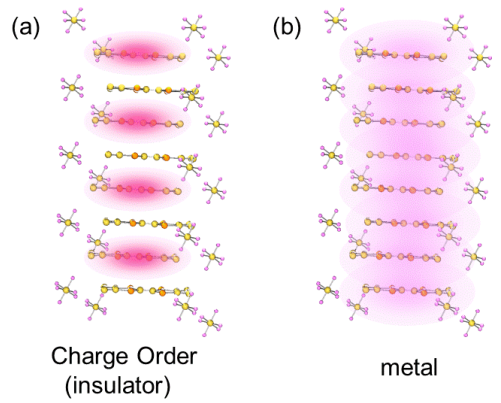


**Fig. 1**.Schematic illustration of **(a)** charge ordering phase and **(b)** metallic phase in $(TMTTF)_2AsF_6$.

0.63 meV$^2$, respectively. However, ultrafast dynamics before the electronic thermalization remains unclear in the 100 fs experiments.

Closed circles in Fig. 2(b) shows the $\Delta R/R$ spectrum measured using 7 fs pulse. Fig. 3 shows the time evolution of $R/R$ (Fig. 3(a)) measured at 0.83 eV, oscillating component (Fig. 3(b)) and the time resolved spectra obtained from the wavelet (WL) analysis (Fig. 3(d)). i) Intense oscillation with a period of 18 fs (1850 cm$^{-1}$) was observed in the timescale of < 100 fs in Figs. 3(a)(b). The broad spectra in Fig.3 (d) reflects the fast damping of the oscillation. This short-lived 18 fs oscillation is attributable to the coherent intermolecular charge motion, because the oscillating energy in Fig. 3(d) corresponds to the intermolecular charge transfer excitation in the optical conductivity spectrum(Fig. 3(c)). ii) complicated waveform in time profile (Fig. 3(b)) and spectral dip (Fig. 3(d)) reflecting the destructive interference between the charge and intra-molecular C=C ($\nu_4$) vibration shows the beginning of the electron-molecular vibration (EMV) coupling. Such coherent electron and electron-vibration (EMV) dynamics are beyond the two-temperature and three temperature model. In particular, the electron oscillation grows in the time scale of 50 fs as shown in Figs. 3(a)(b) after the excitation by 0.89 eV pulse. Considering that the 0.89 eV excitation can generate the charges with large excess energy (~0.8 eV) which are analogous to the free-carriers [5], the growth of the oscillation indicates that the coherence of the correlated charge is spontaneously organized after the excitation. The formation of the electronic coherence observed here is considered to be the elementary dynamics in correlated electron system.

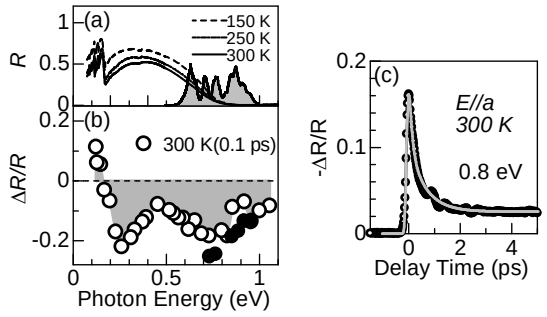


**Fig 2**. **(a)**Steady state reflectivity ($R$) at various temperatures. **(b)** Transient reflectivity ($\Delta R/R$) at 0.1 ps (300 K) measured by 100 fs pulse (open circles). $\Delta R/R$ spectrum measured by 7 fs pulse at 30 fs (closed circles). **(c)** Time evolution of $\Delta R/R$ (measured by 100 fs pulse). The gray curve was calculated by using two-temoerature model (See text).

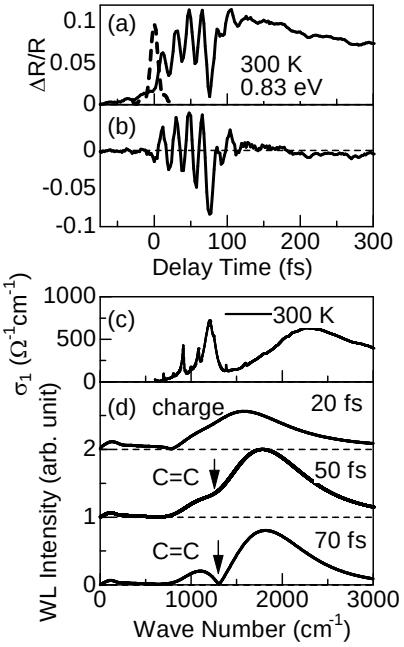


**Fig.3**. **(a)(b)** Time evolutions of $\Delta R/R$**(a)** at 0.83 eV and the oscillating component**(b)** after excitation by 7 fs pulse, respectively. **(c)(d)** Steady state optical conductivity spectrum **(c)** and transient spectra obtained by the wavelet analysis of the oscillating component **(d)**.

## 4. Summary

We have observed the ultrafast coherent dynamics reflecting the correlated charge motion before the thermalization of the electron system in $(TMTTF)_2AsF_6$ by using the 7 fs, 1.5-cycle near infrared pulse. Spontaneous formation of the electronic coherence is induced in the time scale of 50 fs.

## 5. References

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