PHOTO-INDUCED DYNAMICS OF Pt(dmit)$_2$ SALTS

“Molecular movie”

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The observation of the photoinduced dynamics of the structural change with sub-ps time scale must be an interesting challenge for studying the photo-functionality of the molecular crystals. It must be complementary to the conventional time-resolved optical spectroscopy which can detect the electronic structural change. Femtosecond electron diffraction (FED) has been appeared as a powerful tool for the direct measurement of the structural dynamics [1]. We studied the photoinduced dynamics of the charge-transfer and the structural change in the Me$_4$P[Pt(dmit)$_2$]$_2$, which shows a unique phase transition; the charge separated (CS) phase transition ($T_c = 218$ K) [2]. In the low temperature CS phase, tightly bounded neutral and loosely bounded divalent dimers are ordered in the two-dimensional plane in the crystal, due to the strong electron-phonon and electron-electron correlations [3]. The optical pump-probe time-resolved spectroscopy in the CS phase (100 K) suggested the melting of the ordering of the dimers. But the observed dynamics is rather complicated beyond the simple melting of the ordering. The FED result clearly indicates the occurrence of the molecular motions in the sub-ps time-scale, which is corresponding to the observed dynamics by the optical spectroscopy. We analyzed the time-dependence of the electron diffraction patterns and succeeded in making the “molecular movie” which can demonstrate the photoinduced molecular motions. “Molecular movie” shows us the occurrence of the melting of the ordering of the dimers which suggests the charge-transfer between the dimers. And also, it clearly shows the unexpected motions which could not be revealed by the time-resolved optical spectroscopy. The combination of the conventional time-resolved optical spectroscopy and the FED measurement must be powerful in order to reveal the various aspects of the photoinduced dynamics in the molecular crystals.

References