Dynamical study of photoinduced structural phase transitions

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Abstract

Structural phase transition, which inherently reflects the multi-stability of condensed matter, is governed by cooperative and non-linear interactions between electronic and lattice systems. Phase transition triggered by electronic photoexcitation, known as photo-induced phase transition (PIPT), impacts multiple research fields because of following reasons. First, studies of PIPT can directly reveal ultrafast dynamics of cooperative interaction in real- time and space, using specific optically prepared excited states. Second, PIPT research opens a promising route to create new material phases which cannot be reached by conventional excitation under thermal equilibrium [1]. For these reasons, PIPT has been extensively studied world-wide. Here I review present status of the PIPT researches, with putting some emphasis on laser-induced order-disorder phase transitions in semiconductors and metals.

The central issue in PIPT is the dynamics of structural-order transformation under non-equilibrium conditions (electronically excited materials). In order to trace dynamics involved, and to understand then from microscopic point of view throughly, one need to study following three primary aspects of the whole PIPT process; one is the ultrafast dynamics of electronic excited states generated by photoexcitation in fs-temporal domains, the second is the ultrafast structural dynamics triggered by electronic-order changes, and the third is the atomic-scale characterization of photoinduced structural phases. In Fig.1, I show schematically the methodology typically used in PIPT studies.

In Fig.1, I would like to emphasize the two points. First, the time-resolved two-photon photoemission spectroscopy provides more direct information on the ultrafast dynamics of electronic systems in time- energy- and momentum spaces, than any other optical methods [2]. In fact, this method has been applied to study photoinduced metal-insulator transitions in Mott insulators [3]. The second is that time-resolved electron diffraction in femtosecond temporal domains is a powerful technique to capture ultrafast structural dynamics [4,5]. We have recently constructed an ultrafast transmission electron diffractometer with outstanding performance [6]. This instrument, by overcoming serious space-charge effects in previous studies, has shown 90 fs temporal resolution and a high sensitivity for single-shot diffraction measurements. In order to demonstrate this excellent performance, we show in Fig.2 the diffraction pattern measured by a single shot for Au single crystals. The three lens system of

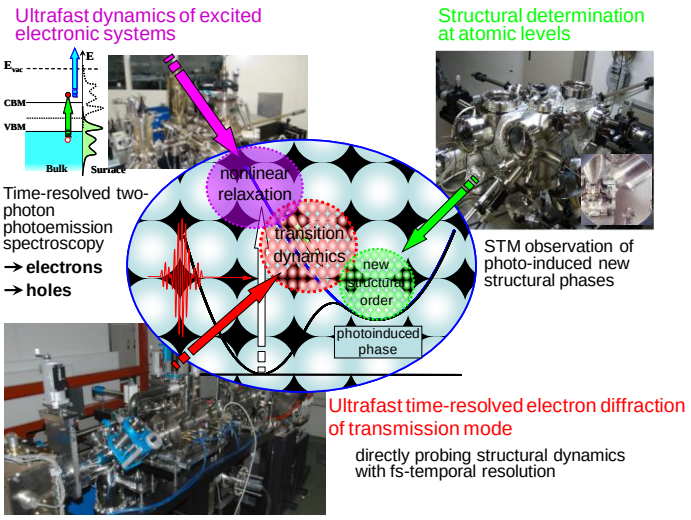

Fig.1 Experimental methods used in PIPT studies

the diffractometer makes it possible to measure different image modes; wide-momentum mode and high precision mode. Image (a) acquired by the former mode shows that the maximum scattering vector is greater than 1.56 Å-1, which is larger than any previously reported. Image (b) acquired by the latter mode displays the diffraction-peak width as narrow as 0.02 A-1, showing an excellent spectral resolution, which makes it possible to determine precisely small shifts of Bragg-peak widths and positions.

Capability of single-shot diffraction measurements is best suited to study irreducible phase-transition processes. Laser- induced melting (order-disorder transition) of solids is a typical example of such processes. We studied the laser- induced melting process of Au, a typical fcc metal, to which extensive studies have been performed. Intensities of Bragg peaks from single-crystalline Au were measured as a function of time delay between pump and probe pulses, and of the excitation laser intensity. The results were analyzed theoretically by combining two-temperature model to describe non-equilibrium states of excited metals with MD calculation. Electronic specific heat and electron-lattice coupling constant were obtained by ab-initio calculation. The structural factor, which corresponds to the Bragg-peak intensity, were calculated from pair density function at each time step of MD calculation.

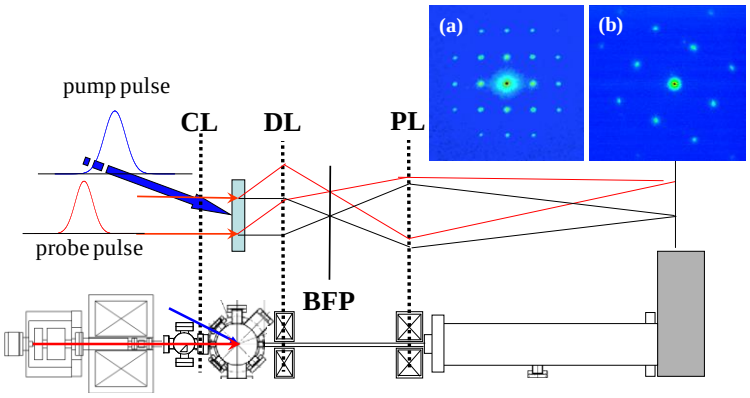

Fig.2 A schematic diagram of Ultrafast electron diffracto-meter and single-shot images for Au single crystals.

The results show following important features of melting of Au, which qualitatively depends on the excitation intensity:

(1) When the absorbed energy is just above the total heat Q_m required to thermal melting, melting starts from the surface, and melt front propagates inside crystals (heterogeneous melting). It takes about 100 ps to melt completely the 10-nm Au film.

(2) When the absorbed energy increases to 1.5 times of Q_m, we first observe a rapid expansion of samples with accompanying surface melting. This is followed by a rapid creation of homogenously distributed small molten pockets in the bulk (homogeneous melting). Within 20 ps after excitation, sample melts entirely.

(3) When the absorbed energy increases further to more than 3 times of Q_m, melting process shows an interplay between lattice heating effects by the electron-phonon coupling and non-thermal effects which leads to variation of potential energy surface of Au nuclei.

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