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Ultrafast probes of coherent oscillations in Fe-based superconductors

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Abstract

Forefront ultrafast experimental techniques have recently proven their potential as new approaches to understand materials based on non-equilibrium dynamics in the time domain. The time domain approach is useful especially in disentangling complicated coupling among charge, spin and lattice degrees of freedom. Various ultrafast experiments on Fe-based superconductors have observed strong coherent oscillations of an A_{1g} phonon mode of arsenic ions, which shows strong coupling to the electronic and magnetic states. This paper reviews the recent reports of ultrafast studies on Fe-based superconductor with a focus on the coherent oscillations. Experimental results with ultrashort light sources from the terahertz-infrared pulses to the hard X-rays from a free electron laser will be presented.

Keywords: Fe-based superconductors, coherent oscillations, ultrafast dynamics

1. INTRODUCTION

Recent development of ultrafast laser technology has enabled various probing tools to explore dynamics in a femtosecond time scale. Ultrafast pump-probe experiments reveal various couplings in the time domain that govern the nonequilibrium dynamics [1-4]. One of the most famous topics that researchers apply any newly developed method to investigate is the long-lasting mystery of the high temperature superconductivity (HTSC).

The Fe-based superconductors show complicated phase diagrams of structural and magnetic phase transitions together with nematic fluctuations of yet unknown origin in the vicinity of the superconducting dome [5, 6]. Therefore, understanding of couplings among those instabilities is essential to unveil the mechanism of HTSC. Various ultrafast techniques have revealed coherent lattice vibrations that strongly modulate the properties of Fe-based superconductors [1, 2, 7-12]. Although the electron-phonon coupling (EPC) on its own fails to account for the large gap and the high superconducting transition temperature, it is still considered as one of the important ingredients for the paring possibly in conjunction with the magnetic interaction [13-16]. In this paper, I will give a short review on recent reports on the coupling of phonon to other degrees of freedom based on coherent lattice vibrations of Fe-based superconductors.

2. PUMP AND PROBE EXPERIMENTS

The ultrafast dynamics in the non-equilibrium state is investigated by a pump and probe method. Various ultrashort light sources from the terahertz (THz) pulses to hard X-rays are available both for the pump and the probe. Figure 1(a) shows the scheme of pump-probe experiments. The sample of interest is excited by pump pulses. If the probe pulse arrives the sample surface earlier than the pump pulse, then the equilibrium property is probed. After the pump excitation, the recovery dynamics back to the equilibrium state is recorded by probe pulses at various time delay t_D.

The optical excitation by the pump pulses deliver huge excess energy to the electronic system of the sample. The excited charge carriers dissipate the energy via various scattering processes. The energy relaxation dynamics usually involves the electronic state at the Fermi level. Therefore, the dynamics depends sensitively on the ground state of the material. It makes the pump-probe experiments powerful for the investigation of coupling among charge, spin and lattice degrees of freedom.

In addition to the electronic excitation, the optical pumping can also give rise to coherent lattice vibrations.

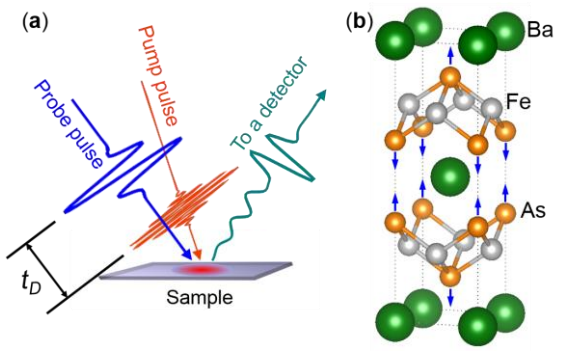

Fig. 1. (a) The scheme of pump-probe experiments. (b) The structure of $BaFe_2As_2$ and the A_{1g} mode of arsenic ions.

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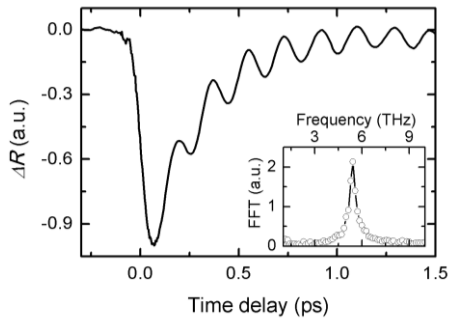

Fig. 2. The photo-reflectivity of $BaFe_2As_2$ at room temperature and $\Phi \approx 230$ μJ/cm^2. The inset shows the Fourier transform of the oscillatory components.

The generation of coherent phonons suggests that the phonon mode is coupled to the electronic state involved in the pump excitation. More interesting is that the generated phonons are all in-phase over the excited volume because the excitation is triggered by coherent ultrashort pulses. Such a uniform lattice deformation due to coherent vibrations could modify the electronic structure although it is transient. Therefore, the coherent lattice vibrations provide a great opportunity to investigate the EPC and relevant phenomena.

3. NIR-OPTICAL PROBE

The most common and easily accessible ultrashort light source is the near infrared (NIR)-optical pulses directly available from an ultrafast Ti:Sapphire laser system. For the pump-probe experiments, the laser pulses are split into two branches of pump and probe, respectively. Although the mono-color NIR-optical pump-probe experiment does not provide any dispersion, its high signal to noise ratio is ideal to investigate small changes of the pump-induced signal.

Figure 1(b) shows the structure of $BaFe_2As_2$, one of the most studied Fe-based superconductors. The iron ions at the centers of tetrahedra of four arsenic ions form a two dimensional square lattice. In Fe-based superconductors, the electronic structure near the Fermi level is dominated by the Fe-d states strongly hybridized with the As-p states. It has been suggested that the lattice vibration of arsenic ion should be strongly coupled to the electronic state [17-19].

Figure 2 shows the photo-reflectivity of $BaFe_2As_2$ at room temperature. In addition to the decrease and relaxation of the overall electronic signal, strong regular oscillations are superimposed. The Fourier transform of the oscillatory signal is shown in the inset of Fig. 2. Its frequency agrees well with the A_{1g} mode of arsenic ions along c-axis shown in Fig. 1(b). Although the strong pumping destroys the superconducting and magnetic orders of the Fe-based materials, the oscillation amplitude stays linear to the pump fluence Φ up to as high as several mJ/cm^2 [1, 7]. Assuming that the relaxation of the electronic signal is dominated by scattering to phonons, P. B. Allen derived the electron-phonon coupling constant

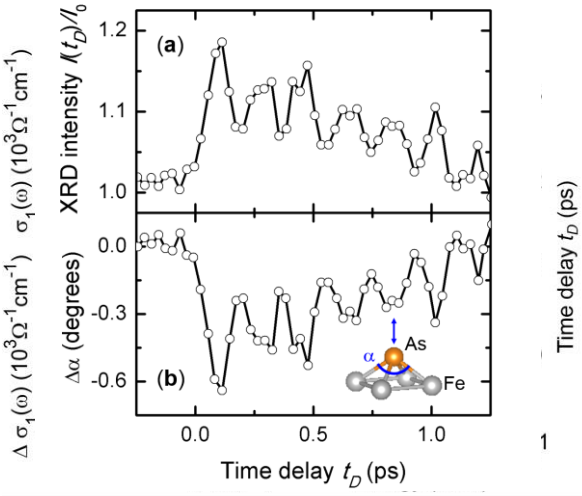

Fig. 3. The NIR pump-IR probe spectra of $BaFe_2As_2$ at T_L = 10 K and $\Phi \approx 63$ μJ/cm^2. (a) Stationary optical conductivity spectra $\sigma_1(\omega)$ in the SDW state at 10 K and in the normal state at 140 K. The transient $\sigma_1(\omega)$ at 180 fs is marked by green circles. (b) Two dimensional color plot of $\Delta\sigma_1(\omega)$, the photo-induced change of $\sigma_1(\omega)$. (c) Spectrally integrated $\Delta\sigma_1(\omega)$ over regions of A, B, C shown in (b). Black lines are single exponential decay fits to the data. Reproduced from [1].

from the relaxation time [20]. Considering the 5.5 THz A_{1g} mode, electron-phonon coupling constant of 0.1-0.2 has been suggested from various pump-probe measurements [7, 10, 21]. Although the A_{1g} mode strongly modulate the electronic structure, the obtained coupling constant cannot account for the high transition temperature of the Fe-based materials by the conventional BCS theory.

4. THz-IR PROBE

While NIR-optical experiments provide different dynamics depending on various order parameters of the material, direct evidence of the order parameter itself is missing in the mono-color transient. The time domain spectroscopy (TDS) can provide complex optical constants over the spectral window. Therefore, the TDS with broadband THz-infrared (IR) pulses is very powerful to resonantly probe dynamics of various order parameters with distinct low energy spectral features such as an infrared phonon and energy gaps of superconductivity or density waves [1, 4]. Broadband TDS spectra have confirmed that the ultrafast dynamics of cuprates and Fe-based materials at low temperatures indeed correspond to the transient response of superconducting and spin density wave energy gaps [1, 4].

Figure 3 shows the dynamics of the spin density waves (SDW) in $BaFe_2As_2$ measured at a lattice temperature T_L = 10 K. As shown in Fig. 3(a), the SDW order is accompanied by a spectral redistribution around 80 meV due to the SDW gap. Figure 3(b) clearly proves that the NIR pump pulses destroy the order transiently, which

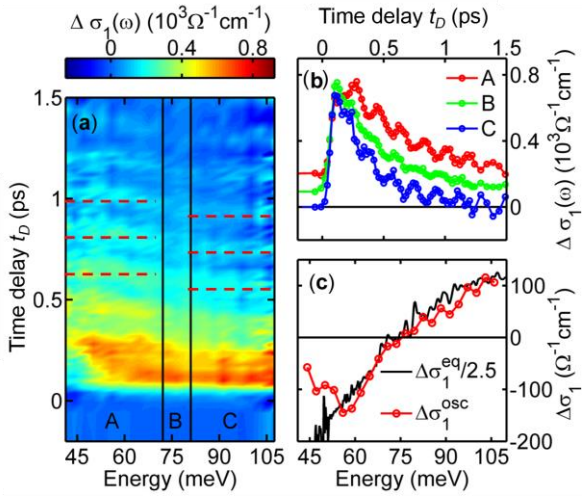

Fig. 4. The NIR pump-IR probe spectra of $BaFe_2As_2$ at T_L = 134 K and $\Phi \approx 530$ μJ/cm^2. (a) Color plot of $\Delta\sigma_1(\omega)$ and (b) spectrally integrated $\Delta\sigma_1(\omega)$ over regions of A, B, C shown in (a). (c) Comparison of $\Delta\sigma_1(\omega)$ by temperature change across the phase transition in the equilibrium state and by the coherent lattice oscillations in the transient state. Reproduced from [1].

recovers in a time scale of about 1 ps. One may hardly notice small signature of the coherent oscillations in Fig. 3(c). Although the coherent oscillations show up at all temperatures, the huge SDW signal dominates the transient change. That is, the oscillations add small modulations to the SDW gap closure-opening signal.

However, the situation becomes different when the SDW order is absent. Figure 4 shows the transient conductivity in the normal state at T_L = 134 K just above the transition temperature. The overall spectral change does not show the SDW gap closure-recovery dynamics shown in Fig. 3. The overall increase of $\sigma(\omega)$ is consistent with the heating of the electronic system. In addition to the heating and cooling of the electronic system, regular oscillation patterns are clearly observed during the recovery in Fig. 4. More interesting is that the oscillations have a dispersion. Figure 4(b) clearly shows that $\Delta\sigma_1(\omega)$ changes the sign in the middle region of B marked in Fig. 4(a). Strikingly, this dispersion agrees with $\Delta\sigma_1(\omega) = \sigma_{100K} - \sigma_{140K}$ in the equilibrium state that shows the SDW gap develops. It suggests that the SDW order parameter is modulated depending on the displacement of the A_{1g} phonon mode.

The SDW order in Fe-based materials arises from the nesting instability between hole and electron bands located at the zone center and zone boundaries (Γ and X/Y points in the Brillouin zone for the one iron square lattice unit cell), respectively. It has been suggested that the magnetic moment of iron ions and the band structure around the Fermi level are sensitive to a small change of the As distance from the iron square lattice [17, 18]. Indeed, the time and angle resolved photoemission measurements have shown that the chemical potential and the electronic bands near E_F are periodically modulated by the 5.5 THz A_{1g} mode vibrations [11]. Therefore, the phonon oscillations could strongly modulate the nesting instability such that a transient SDW order periodically should follow the ultrafast lattice motion as shown in Fig. 4.

5. X-RAY PROBE

The free electron laser (FEL) has opened a new era of the ultrafast investigation on the lattice dynamics. Intense and ultrashort X-ray pulses from FEL shorter than the oscillation period of phonon vibrations allows us to make an ultrafast film on the motion of the lattice, which could reveal the role of the lattice in realizing various material properties together with other ultrafast techniques.

As shown in previous sections, the strong coherent phonon oscillations of arsenic ions have been observed in many experiments. Although the experimental results could be understood in a qualitative manner, detailed mechanism was lacking without the information of the exact motion of the arsenic ions. Research efforts with both the FEL and synchrotron radiations have finally succeeded in monitoring the lattice dynamics from the ultrafast modulations of Bragg diffraction peaks [2, 12, 22].

Fig. 5. (a) Time resolved X-ray diffraction intensity from $(118)_T$ plane of $BaFe_2As_2$ at T_L = 140 K and $\Phi \approx 2.9$ mJ/cm^2. (b) Variation of the Fe-As-Fe angle depending on the phonon oscillations. Reproduced from [2]

Figure 5(a) shows that the Bragg peak intensity from the $(118)_T$ plane of the tetragonal symmetry is modulated at the frequency of the A_{1g} mode. The peak intensity modulations occur due to the change of the scattering form factor depending on the position of the arsenic ions. Therefore, the measurements provide us the transient position of the arsenic ions. Figure 5(b) shows the variation of the Fe-As-Fe bond angle α from the intensity modulation. It shows that the arsenic ions move away from the iron square lattice upon pumping. The FEL measurements suggest that the modulation of the angle α could be as large as about 1 degree under high fluence pumping. It is worth to note that the estimated displacement of arsenic ions based on the NIR-optical probe is comparable to the measured results by the FEL [1, 2]. Band structure calculations show that the decrease of α results in strong shift of hole bands at Γ and X/Y points. These shifts of bands modify the nesting character strongly such that another nesting between two xy bands at Y and M points gets strongly enhanced, which could give rise to a transient SDW order.

The magnetic order is strongly coupled to the structural phase transition in the equilibrium state. However, the ultrafast destruction of the magnetic order at low temperature or the transient SDW order in the normal state do not involve the structural phase transition [2, 12, 22]. The transient SDW state in the tetragonal state suggests that the orthorhombic structure is not the necessary condition for the magnetic order. Likewise, the transient state without the magnetic order in the orthorhombic structure tells us that the normal state without the SDW order is compatible

with the orthorhombic structure, although the structure prefer the magnetic order in the equilibrium state. The pump-induced structural transition occurs only under an extremely high fluence and only after the excess energy in the electronic system is transferred to the phonon system by a few tens of picosecond time scale [22]. That is, the structural phase transition is driven only by heating the lattice itself.

6. SUMMARY AND OUTLOOK

Various ultrafast experiments have shown that the coherent lattice vibrations strongly modulate the magnetic property of Fe-based materials. The generation of the transient SDW order depending on the displacement of the arsenic ions clearly demonstrates the strong coupling of the magnetic state and the lattice degree of freedom. The time and angle resolved photoemission and femtosecond X-ray diffraction experiments together with band calculations suggest that the transient SDW order could be explained by an enhanced nesting instability in the modified band structure by the lattice vibrations. However, it has to be noted that the transient SDW order was observed under much weaker fluence than the condition that the band calculation expects enhancement of the new type of nesting instability. Various transient states reveal a variety of combinations of the magnetic states and lattice structures. More details for the exciting transient phenomena and the link to the high temperature superconductivity is yet to be understood.

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