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Tunable Mid-infrared Pump Pulse Realized by Optical Parametric Amplification in Time Resolved ARPES System

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Angle-resolved photoemission spectroscopy (ARPES) is one of the most powerful techniques for mapping the electronic structures of solid-state materials by measuring the energy and momentum of photoemitted electrons. Time- and angle-resolved photoemission spectroscopy (TARPES) is a combination of pump-probe spectroscopic technique and ARPES, with which nonequilibrium electronic structures and ultrafast dynamics of solids can be directly observed. Pump-probe spectroscopy is a general method to detect ultrafast electronic dynamics. One beam (pump) is used to excite a sample, driving it out of equilibrium, and another beam (probe) is used to monitor light induced changes caused by pump. A series of relaxation processes of the electron population can be traced in time domain by sweeping the delay time between the two pulses. Owing to the very short time duration of the used laser pulses, dynamics in timescale of femto-seconds can be observed.

Usually we use near-infrared light pulses with the center wavelength of 800 nm for the pump in TARPES setup because it is fairly easy to be installed by just delivering the output of Ti:Sapphire lasers. However, the pump pulses in this range of wavelength excite mainly electron systems with the excess energy for the most metallic, semimetallic, or narrow-gapped semiconducting materials. That results in the unavoidable heating effects and destroy many electronic or magnetic orders. Different from near-infrared light pulses, mid-infrared light pulses can excite electron systems with less excess energy and suppress above-mentioned heating effects. Furthermore, they can resonantly excite electrons and optically active lattice vibrations in some materials and their amplitudes can be enhanced as high as several percent of interatomic distances.

Since most of laser sources provide pulses at a specific wavelength, especially in the visible spectrum, a great effort has been dedicated to the development of tunable sources. In this sense, optical parametric amplification (OPA) is a practical solution that can provide an adjustable source in an IR-spectrum window beyond 800 nm. OPA is a well-known nonlinear optical process occurring in second-order nonlinear crystals and involving energy transfer among beams at different frequencies. Energy is transferred from a beam with high frequency (pump beam) to a beam with lower frequency (signal beam), thus resulting in an amplification of the signal. To fulfill energy conservation, the third beam (idler beam) is generated. In addition, by generating another beam of which frequency corresponds to the difference between signal and idler beams from another nonlinear optical crystal, a mid-infrared beam can be obtained.

In our lab, the pump pulse used for OPA is generated with a commercial extremely stable Ti: sapphire regenerative amplifier system with the center wavelength of 800 nm, the repetition rate of 10 kHz, and the pulse duration of ~35 fs. The used OPA is also commercial with a two-stage parametric amplifier of white-light continuum. The output power of OPA is given in Fig. 1 (a) as a function of wavelength. With the pump, the OPA is supposed to provide tunable signal pulses from 1100 nm to 1600 nm, and idler pulses from 1700 nm to 2400nm. Due to the low efficiency of the nonlinear optical process, the power obtained is about several hundred milliwatts, but it is sufficient for the TARPES experiment. In the future we will also install non-colinear difference frequency generator (NDFG) extension, whose output is supposed to cover from 2500 nm to over 10 microns, to realize mid-infrared pumping.

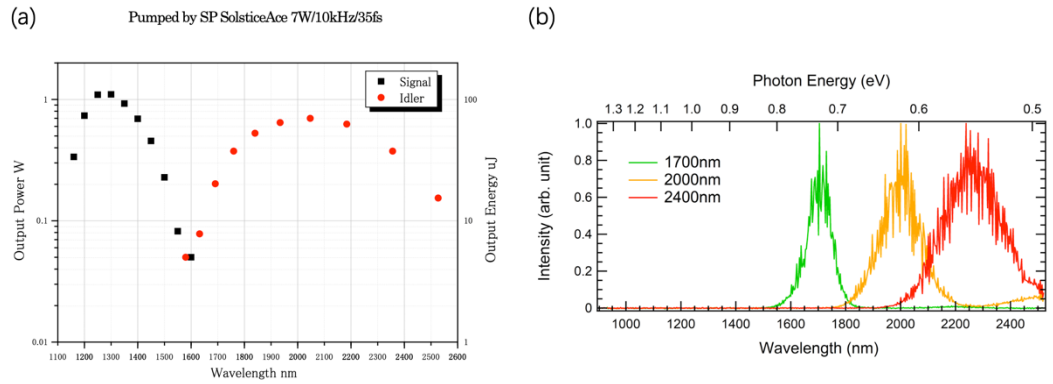

Fig. 1: The output spectra of OPA. (a) The output power spectra. The control of the output power can be achieved by adjusting the power of the pump light in the experiment. (b) Spectra obtained by spectrometer at three specific wavelengths.

After installation, we selected three specific center wavelengths of the idler pulses of OPA for performing the TARPES measurements: 1700 nm, 2000 nm, and 2400 nm, corresponding to photon energies of 0.73 eV, 0.62 eV and 0.52 eV, respectively. The spectra of the three wavelengths are shown in Fig. 1 (b). We first measure the beam size for each wavelength, which is necessary for the calculation of the pump fluence and has an influence on the quality of the experimental data. In principle, we want the beam spot to be as small as possible to reduce the muti-photon effects from the silver paste or cupper plate which surround the sample of interest. The beam profiles are expected to be Gaussian pulses, and we use a knife edge technique to do the measurement, where a blade is placed in front of the laser beam and measuring the power transmitted after the blade using a photodiode. By fitting the data, the radius of the beam spot in the direction of the edge movement is obtained.

Another significant parameter that needs to be measured is the temporal resolution of the TARPES system. We use highly oriented pyrolytic graphite (HOPG) for the measurement, and the results of 2400 nm pumping are given in Fig. 2. Fig. 2(a) shows the TARPES spectra of HOPG, and by integrating the area above E_F shown as a red box in Fig. 2(a) the temporal evolution of the photoemission intensity is obtained, as shown in Fig. 2(c). We fit the data with a function consists of a convolution of an exponential function with a Gaussian function and the FWHM of the Gaussian corresponding to the total temporal resolution of the TARPES system is 222.5 fs. The obtained temporal resolution for each wavelength is given in Fig. 2(d).

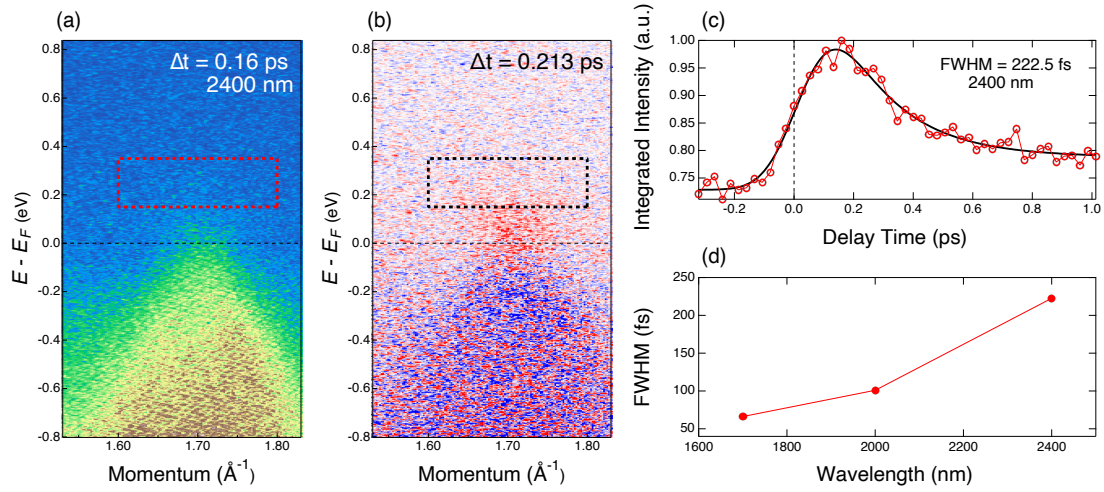

Figure 2: (a) TARPES spectra of HOPG around K point at 300K, the wavelength of pump is 2400 nm. (b) Differential spectra. (c) Temporal evolution of the photoemission intensity. The integrated area is marked with dashed boxes in (a) and (b). The black solid line is the fitting curve of the spectrum. (d) FWHM of each wavelength.

Having completed the measurement of these necessary parameters, we perform TARPES measurements with pump light at these three wavelengths. Another sample is Ta_2NiSe_5, which is known as a plausible candidate of excitonic insulators and exhibits a semiconductor-to-insulator transition at 328 K, accompanied by a structural distortion. In addition, recently we found a photo-induced insulator-to-metal transition for this material, and the nonequilibrium dynamics caused by an ultra-short laser pulse attracts much attention. However, we pumped this material at 800 nm (1.55 eV) previously while the band gap of this material is 0.16 eV. In that case, we unavoidably inject excess energy in electron and lattice system, which is considered to destroy multiple orders including excitonic insulator. That is our motivation to perform nearly-resonant excitation to band gap, which will decrease excess energy and suppress heating effect. We anticipate some nontrivial phenomena by such a condition.

The measurements are performed at 100K, in which the system is in the excitonic insulator phase. We used different wavelengths (photon energies) of pump pulses for the measurements and observed the lifetime limited insulator-to-metal transition process, as shown in Figs 3(a) and 3(b). We observed the dynamics of photoexcitation at different photon energies, and the drop time and relaxation time of each wavelength is shown in Fig. 3(d).

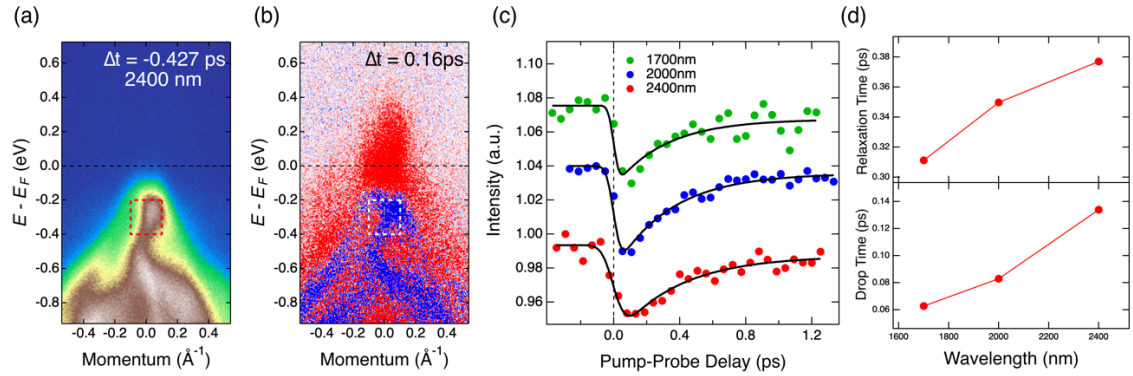

Fig. 3: (a) TARPES spectrum of Ta_2NiSe_5 before pumping (-0.43 ps). (b) Differential spectrum after pumping (0.16 ps). (c) Temporal evolution of the photoemission intensity integrated in dashed boxes in (a) and (b) for different pump wavelengths. (d) Extracted drop time and relaxation time of different pump wavelengths.

In the future, we plan to install a NDFG extension to obtain a pump beam covers from 2500 nm to over 10 microns. As the optical conductivity spectra indicates, the energy gap between the valence and conduction band is about 0.4 eV at a low temperature, and we look forward to reaching a longer wavelength. Besides, to maintain and improve the metrics of the TARPES system, we need to continue to try to stabilize the pump laser. As a material, we will investigate some other system such as another type of excitonic insulator or nontrivial CDW system, which have not been performed by wavelength-tunable excitation and thus might provide interesting playground.

[Conference Presentations]

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