

# Development of Sub-100-fs Time-Resolved Spin-ARPES and Frequency-Domain Analysis of Electronic-Structure Modulation in BiTeBr

Nagi Suzuki<sup>a</sup>, Shin Yokoyama<sup>a,b</sup>, Ko Sasaki<sup>a,b</sup>, Takuma Iwata<sup>a,b</sup>,  
Akio Kimura<sup>a,b,c</sup>, Koji Miyamoto<sup>d</sup>, Taichi Okuda<sup>b,c,d</sup>, Mario Novak<sup>e</sup>,  
and Kenta Kuroda<sup>a,b,c</sup>

<sup>a</sup>*Graduate School of Advanced Science and Engineering,*

*Hiroshima University, Higashi-Hiroshima, Hiroshima 739-8526, Japan*

<sup>b</sup>*International Institute for Sustainability with Knotted Chiral Meta Matter (WPI-SKCM2),*

*Hiroshima University, Higashi-Hiroshima, Hiroshima 739-8526, Japan*

<sup>c</sup>*Research Institute for Semiconductor Engineering,*

*1-4-2 Kagamiyama, Higashi-Hiroshima, Hiroshima 739-8527, Japan*

<sup>d</sup>*Research Institute for Synchrotron Radiation Science (HiSOR),*

*Hiroshima University, 739-0046 Higashi-Hiroshima, Japan,*

<sup>e</sup>*Department of Physics, Faculty of Science, University of Zagreb, 10000 Zagreb, Croatia*

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The ultrafast control of spin-polarized electronic states using light that leads to modulation of lattice and electronic structures has not yet been firmly established as a distinct strategy in optical spintronics. One promising approach is the photoinduced coherent phonons, which modulate spin-polarized electronic structure by electron-phonon coupling. To establish this route, it is essential to directly capture the ultrafast temporal response of photoinduced electron-phonon coupling and to trace the full temporal evolution of the resulting modulation in spin-polarized electronic states. Here we developed a time- spin- and angle-resolved photoemission spectroscopy (Tr-SARPES) apparatus which achieved temporal resolution sufficient to resolve ultrafast electron-phonon-coupling dynamics (92 fs) by incorporating a femtosecond laser and a pump-probe optical setup into the SARPES instrument at HiSOR [1] and applied it to reveal the ultrafast dynamics of electronic states coupled to coherent phonons excited by femtosecond pulses in the polar semiconductor BiTeBr.

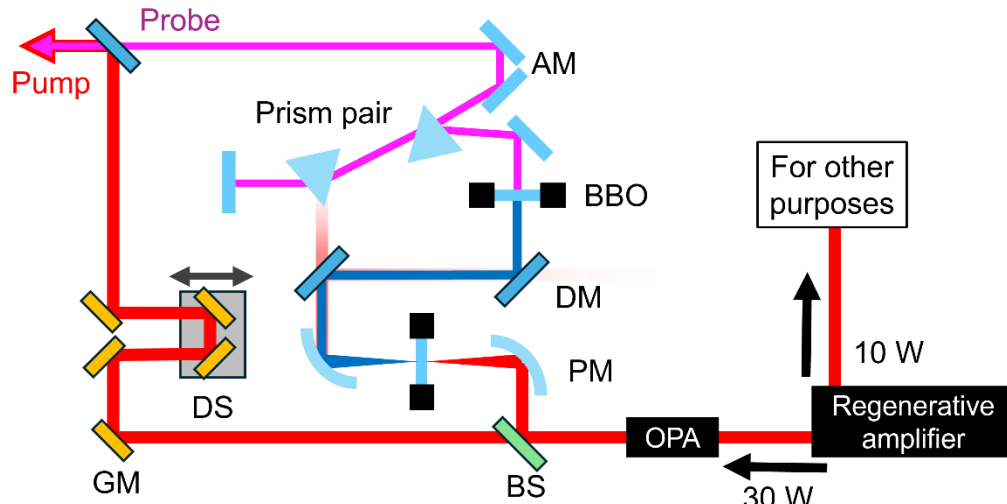
The developed pump-probe optical layout is shown in Figure 1. The setup comprises a laser source and two optical paths: one for the pump and one for the probe. As a laser source, we implemented a regenerative amplifier, delivering 1030-nm pulses with a pulse duration of 190 fs and an average power of 40 W at 100 kHz repetition rate. For Tr-SARPES measurements, 30 W is used, which is carried into an optical parametric amplifier (OPA). This OPA provides tunable pulses covering from 650 to 2500 nm. In this work, we used the signals from OPA at 840 nm with temporal width of 45 fs. A beam splitter splits the IR-beam into pump and probe optical paths. In the probe optical path, ultraviolet probe pulses were generated by second-harmonic generation and subsequent fourth-harmonic generation (FHG) in BBO crystals. After FHG, dispersion was compensated with a prism-pair compressor to optimize probe pulse duration. In the pump optical path, the pump beam was sent through an optical delay stage to control the pump-probe delay time. Finally, the pump and probe beams were recombined using a dichroic mirror to form a collinear beam path and were focused onto the sample inside the ultrahigh-vacuum chamber.

We evaluated the temporal resolution of the Tr-SARPES system using the topological insulator Bi<sub>2</sub>Te<sub>3</sub>. In the ground state [Fig. 2(a)], no photoemission intensity was observed above the Fermi level ( $E_F$ ). With pump and probe [Fig. 2(b)], intensity appeared above  $E_F$ , originating from unoccupied bulk bands, confirming successful detection of the excited-state electronic structure. We then estimated the instrument response by minimizing material-dependent contributions such as carrier relaxation dynamics. Specifically,

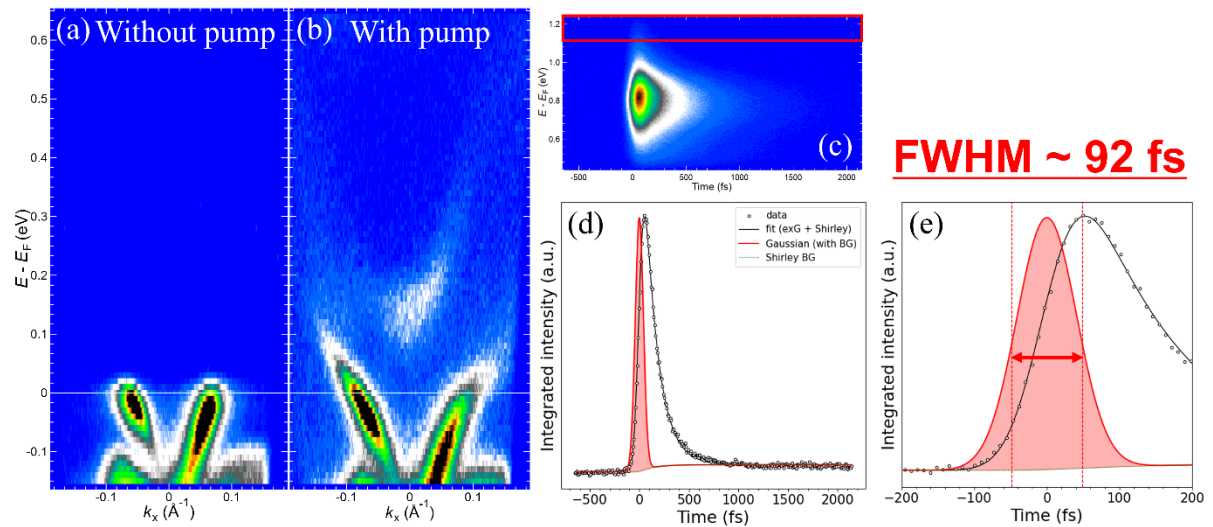
to reduce the influence of relaxation, we analyzed unoccupied states well above  $E_F$  [Fig. 2(c)]. By fitting the energy-window-integrated transient intensity, we obtained the pump-probe cross-correlation modeled as a Gaussian [Fig. 2(d, e)] and define the temporal resolution as the FWHM of the fitted Gaussian, yielding 92 fs.

Finally, using this apparatus we tracked the ultrafast dynamics of Rashba-split electronic states in the Rashba-type polar semiconductor BiTeBr and observed pronounced oscillations in the time-dependent photoemission intensity after photoexcitation, attributable to photoinduced coherent phonons.

The development of this apparatus thus unlocks sub-100-fs, time-resolved access to the interplay between lattice motion and spin-polarized electronic structure.



**FIGURE 1.** Schematic diagram of the pump-probe optical setup. (BS: Beam Splitter, GM: Gold Mirror, DS: Delay Stage, PM: Parabolic Mirror, DM: Dichroic Mirror, AM: Aluminum Mirror)



**FIGURE 2.** (a) ARPES image of  $\text{Bi}_2\text{Te}_3$  in the ground state observed with the probe beam only. (b) ARPES image of  $\text{Bi}_2\text{Te}_3$  in the excited state observed with both the pump and probe beams. The measurements were performed at 30 K with a 5.9 eV probe, a 1.48 eV pump, and a pump fluence of  $2.9 \text{ mJ/cm}^2$ . (c) Angle-integrated photoemission intensity in the unoccupied band located around 1 eV above the Fermi energy. (d) Observable obtained by integrating the region enclosed by the rectangle in (c), together with its fitting. The fitting function is given by the convolution of a Gaussian representing the input signal of the laser pulse and a response function reflecting the electron lifetime,  $f(t) = \Delta I_0 e^{-(t-t_0)/\tau} \Theta(t-t_0)$ . The Gaussian determined from the fitted parameters is shown as the red curve. (e) Magnified view of (d).

## REFERENCES

1. T. Iwata *et al.*, *Sci. Rep.* **14**, 127 (2024).