Design of Terahertz Pump – Photoemission Probe Spectroscopy Beamline at UVSOR-II

Shin-ichi Kimura*,†, Eiken Nakamura*, Masahiro Hosaka**, Toshiharu Takahashi‡ and Masahiro Katoh*†

* UVSOR Facility, Institute for Molecular Science (BUNSHIKEN), Okazaki 444-8585, Japan
† School of Physical Sciences, The Graduate University for Advanced Studies (SOKENDAI), Okazaki 444-8585, Japan
** Graduate School of Engineering, Nagoya University, Chikusa-ku, Nagoya 463-8603, Japan
‡ Research Reactor Institute, Kyoto University, Kumatori, Osaka 590-0494, Japan

Abstract. At UVSOR-II, Institute for Molecular Science, a new beamline for novel pump-and-probe spectroscopy experiments combining terahertz (THz) coherent synchrotron radiation and vacuum-ultraviolet coherent harmonic generation from same electron bunches in the storage ring interacted with an amplitude-modulated pulse laser introduced from the outside of the storage ring. The purpose of the beamline is to elucidate the electronic structure relating to physical properties of solids by using selective excitations of low energy electronic and vibrational structure. The beamline design is reported.

Keywords: Terahertz; Coherent synchrotron radiation; Coherent harmonic generation; Photoemission; Pump-probe

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Recently, new quantum phases of solids are attracting attention, because new physical properties that have never been observed at ambient conditions and the advent of new functionalities are expected. The new physical properties usually appear below room temperature (300 K \(\sim 24\) meV). The electronic structure located in the energy region of several 10 meV below and above the Fermi level \((E_F)\) is the origin of physical (thermodynamic) properties as well as functionalities. Therefore it is important to study the electronic structure near \(E_F\) to clarify the origin of the new physical properties. The electronic structure can be directly and selectively excited by photons in the infrared (IR) and terahertz (THz) regions.

One of methods to create intense THz light is coherent synchrotron radiation (CSR). CSR has been observed at an electron linac at Tohoku University in 1980’s for the first time. [1] After that, the fundamental properties of CSR has been studied for a long time, but it has not been used for applications because of unstable intensity of light due to unstable electron beams of linacs. Since 2000, CSR has been observed in storage rings. [2] The spectroscopic applications using the storage-ring-based CSR were carried out at BESSY-II and so on. [3] However there is no application for an excitation source so far.

At UVSOR Facility, Institute for Molecular Science, Japan, fundamental studies on the generation of CSR by using a bursting mode [4] and by a laser slicing technique [5, 6] were performed. In addition, the quasi-monochromatic CSR was produced due to the periodic modulation of electron bunches created by an amplitude-modulated laser pulse introduced from the outside of the storage ring. [7] The obtained CSR was \(10^4\) times higher intensity than the “normal” (incoherent) SR. On the other hand, coherent harmonic generation (CHG) of the incident laser pulse can be produced by the same periodic modulation of electron bunches. [8]

On the other hand, there are some interesting phenomena due to the excitation of mid-IR and THz sources; one is the vibration in conductivity of manganite \((\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3)\) excited by a mid-IR pulse laser, [9] the other picosecond carrier dynamics in indium antimonide \((\text{InSb})\) in the THz region after a THz pumping. [10] However, there was no direct observation of the time-dependent electronic structure after the THz excitation so far.

To investigate electronic structure, photoemission spectroscopy (PES) is a powerful experimental method because the occupied electronic structure as well as the density of states can be directly detected. Recently, the energy resolution rapidly increases and the electronic structure of the origin of the physical properties can be observed. Therefore very small change in the electronic structure due to the THz excitation is expected to be detected by PES at present.

Then we are planning to perform the THz pump – PES probe spectroscopy using the quasi-monochromatic THz-CSR as an excitation source and the CHG as a probing light. The feature of the spectroscopy is to probe the time-dependent electronic structure after the resonant excitation of vibration modes and energy gaps. In the experiment, the
higher order of CHG light, for instance 6-th order (∼9 eV, VUV-CHG), is expected because the CHG is used for the light source for PES. Since both THz-CSR and VUV-CHG are emitted from the same electron bunch, there is no time jitter in between. The pulse width and the repetition rate of THz-CSR and VUV-CHG are several 100 fsec-to-several 1 psec and 1 kHz, respectively, due to the incident laser pulse. Since the energy resolution and the peak intensity of the VUV-CHG are expected to be less than 5 meV and 100 times higher than the “normal” SR, respectively, the VUV-CHG is available for the light source of high-resolution PES. In this paper, we report the design of the THz pump – PES probe spectroscopy beamline at UVSOR-II using the THz-CSR and VUV-CHG.

In Figure 1, the designed schematic top view of the THz pump – PES probe beamline, BL1, is depicted. The amplitude-modulated Ti:Sa laser pulse (1 kHz, 10 mJ/pulse) is introduced to the electron storage ring of UVSOR-II. Then a periodic energy modulation is created on an electron bunch at one of two undulators, namely “Modulator”, located at the straight section. VUV-CHG is emitted from the modulated electron beam at the downstream undulator, namely “Radiator”. The modulated electron beam also produces the quasi-monochromatic THz-CSR at the bending magnet located at the downstream of the undulators. Since both THz-CSR and VUV-CHG are monochromatic lights, no monochromator for both lights is needed. However, the spectral feature of the THz-CSR must be confirmed by a Fourier transform interferometer (FTIR). Both VUV-CHG and THz-CSR are directed to the same position on a sample in an ultra-high vacuum chamber with a photoelectron analyzer (VG-Scienta SES100) located at the outside of the radiation shielding wall. The THz pump – PES probe experiment is performed as follows; At first, THz-CSR is irradiated to the sample. Just after that, VUV-CHG is irradiated to the sample and PES measured with the time delay. The time delay between the THz-CSR and VUV-CHG is made by the delay line in the optical pass of VUV-CHG. Since the expected 6-th higher harmonics (∼9 eV) is not high photon energy, normal incident optics can be used for the delay line.

Figure 2 shows the form factor of THz-CSR at several frequencies as a function of the horizontal emission angle from the straight section. The form factor corresponds to the efficiency of the evolution of CSR. THz-CSR below 3 THz (∼100 cm⁻¹) is generated from 10 degree. Above 20 degree, the intensity of THz-CSR below 2 THz (∼67 cm⁻¹) is rapidly increased. In addition, according to the geometrical condition, the first mirror can be put at 34 degree in maximum. Therefore the acceptance angle of THz-CSR was adopted to be 20–34 degree (244 mrad) in horizontal direction. The vertical angle was set to ±40 mrad for collecting the widely expanded THz-CSR. To collect THz-CSR in the wide acceptance angle, a three-dimensional “magic mirror” that has been successively installed at BL43IR, SPring-8 [11] and BL6B, UVSOR-II [12] is employed.
To summarize, we designed a beamline for THz pump – photoemission probe spectroscopy using THz-CSR and VUV-CHG generated by an amplitude-modulated laser pulse at UVSOR-II. The purpose of the beamline is to elucidate the low-energy electronic structure that is the origin of the physical properties of solids. The beamline will be operated in 2011.

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REFERENCES