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Infrared experiments under laser excitations at SPring-8

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Abstract

The infrared (IR) beamline BL431R at SPring-8 has recently become available to general users. It has four experimental stations, one of which is the absorption and reflection spectroscopy station (ARSS). ARSS is designed primarily for absorption and reflection IR spectroscopies under laser excitations, in particular for laser-pump, IR SR-probe time-resolved spectroscopies. We describe the instrumentations at ARSS, and the expected future projects.

Keywords: Time-resolved spectroscopy; Infrared synchrotron radiation; SPring-8

The infrared (IR) synchrotron radiation (SR) has many advantages over the conventional IR sources [1]. It is a polarized, broad-band source, which is much brighter than the conventional IR sources. It is also a pulsed source on the ns time scale. Several SR facilities in the world have developed beamlines designed exclusively for the use of IR SR. The BL43IR [2] at SPring-8, the large synchrotron radiation facility of the Japan Synchrotron Radiation Research Institute, is one of such IR beam lines, which has become available

to general users since April 2000. It consists of a front-end section with unique optical designs to extract the IR SR from the SPring-8 storage ring [3], a Fourier-transform interferometer (Bruker IFS HR120), and four different experimental stations [2,4,5]. Among them is the absorption and reflection spectroscopy station (ARSS), which is intended primarily for absorption and reflection IR spectroscopies under laser excitations, in particular, laser-pump, IR SR-probe time-resolved experiments with a sub-ns temporal resolution [6]. These experiments take advantage of the pulsed nature of the IR SR, and they give the unique opportunity of studying various time-dependent phenomena on the ns time scale in wide range of materials. Below, we first describe the basic features of the ARSS, then expected future research projects.

Fig. 1 shows the photograph of a part of the ARSS. The sample chamber is UHV-compatible, with many view ports for an easy access of the excitation laser beam as well as for monitoring the beam spots on the sample. The sample chamber is

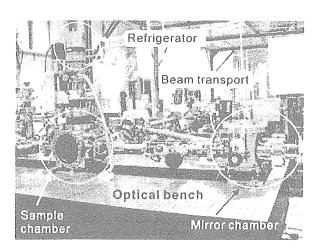


Fig. 1. Photograph showing a part of the absorption and reflection spectroscopy station (ARSS) at the infrared beamline BL431R of SPring-8.

placed on an optical bench, so that the excitation laser beam can be easily carried to the sample by a set of mirrors. The collimated IR SR carried through the beam transport from the interferometer is focused by an off-axis paraboloidal mirror (placed in the mirror chamber seen in Fig. 1) onto the sample, which is mounted on the cold finger of a closed-cycle He refrigerator (temperature range 5-320 K). The refrigerator is inserted into the sample chamber through a three-axis manipulator, so that several samples can be studied at the same time. The transmitted or reflected IR beam from the sample is focused by another off-axis paraboloidal mirror onto a HgCdTe photoconductive detector (mid-IR) or a Si bolometer (far-IR). Optical windows are used to separate the high vacuum of the sample chamber and the low vacuum of the beam transport/detector sections. BaF₂ windows are used for the mid-IR, and CVDgrown diamond windows for the far-IR, which have a 1.2° wedge to avoid interference due to multiple internal reflections.

Fig. 2 shows the schematic diagram of the ps pulsed laser system and the pulse timing detection electronics at the ARSS. The laser system is based on a mode-locked Ti:sapphire laser (Spectra

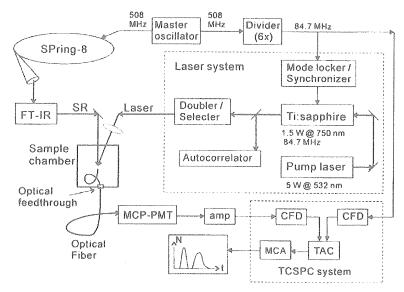


Fig. 2. Schematic diagram for the time-resolved pump-probe experiments at BL431R of the SPring-8. The abbreviations in the diagram stand for the following. FT-IR: Fourier-transform infrared interferometer, MCP-PMT: microchannel-plate photomultiplier tube, CFD: constant fraction discriminator, TAC: time-to-amplitude converter, MCA: multichannel analyzer, TCSPC: time-correlated single photon counting.

Physics Tsunami 3950), pumped by a CW solid state laser (Spectra Physics Millenia). This system generates 1.5 ps pulses at 84.7 MHz in the 720-900 nm range (with standard optics in the Ti: sapphire) or in the 880-1100 nm range (longwave optics). A doubler/selector is used for the second harmonics generation and/or the reduction of repetition rate. In order to synchronize the laser pulses with the SR pulses, an electronic divider is used to select every sixth pulse out of the 508 MHz RF pulses from the SPring-8 master oscillator, then the selected 84.7 MHz pulses are input to a synchronizer electronics that is combined with the mode locker. Most typically, the experiment is done under an operation mode where only every sixth RF bucket is filled with the electrons, so that the IR SR pulses from the ring have exactly the same time interval with the laser pulses. For other operation modes with a longer SR pulse interval, the repetition rate for the laser pulses is reduced using the selector. The timing of the laser pulses relative to that of the SR pulses can be varied using either an external delay line or an electronic delay in the synchronizer. To monitor the arrival times of laser and SR pulses simultaneously, an optical fiber is attached to the sample holder. The SR and laser pulses carried through this fiber are detected by a microchannel-plate photomultiplier (MCP-PMT, Hamamatsu R3809U). This timing monitor can be done without breaking the vacuum, by interchanging the samples and the fiber by the manipulator. The electrical pulses from the MCP-PMT are time-resolved using a standard time-correlated single photon counting (TCSPC) electronics [7]. Fig. 3 shows an example of the measured temporal profiles for laser and SR pulses. From these data, the combined transit time spread of the TCSPC electronics and the MCP-PMT is $\sim 100 \,\mathrm{ps}$ (note that the actual laser pulse width is 1.5 ps), and the SR pulse width at the sample position is estimated to be $\sim 400 \,\mathrm{ps}$.

At ARSS we plan to perform various timeresolved IR experiments. It has been known that the IR spectroscopy is a particularly powerful probe for the free carrier dynamics in solids and for the molecular/lattice vibrations. Using the laser-pump, IR-probe time-resolved spectroscopy,

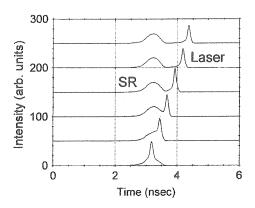


Fig. 3. Examples of the temporal profiles of SR and laser pulses detected at the sample position, with different time delays. For clarity each curve is offset along the vertical axis. The combined transit time spread of the TCSPC electronics and the MCP-PMT is $\sim 100\,\mathrm{ps}$ and the actual laser pulse width was $\sim 1.5\,\mathrm{ps}$. Hence the SR pulse width at the sample position is estimated to be $\sim 400\,\mathrm{ps}$.

their transient behaviors can be studied. Potential future research projects at ARSS include the following. (i) The temporal dynamics of photo-induced carriers in semiconductor nano-structures (quantum wells, wires, etc) and in organic conductors (poly-acetylene, β -calotene, etc). (ii) Fine structures and temporal evolutions in the vibrational spectra of localized centers in ionic crystals. (iii) Transient vibration spectroscopies of photo-reactive/photosensitive biological systems (photosynthetic centers, rhodopsin, etc.).

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