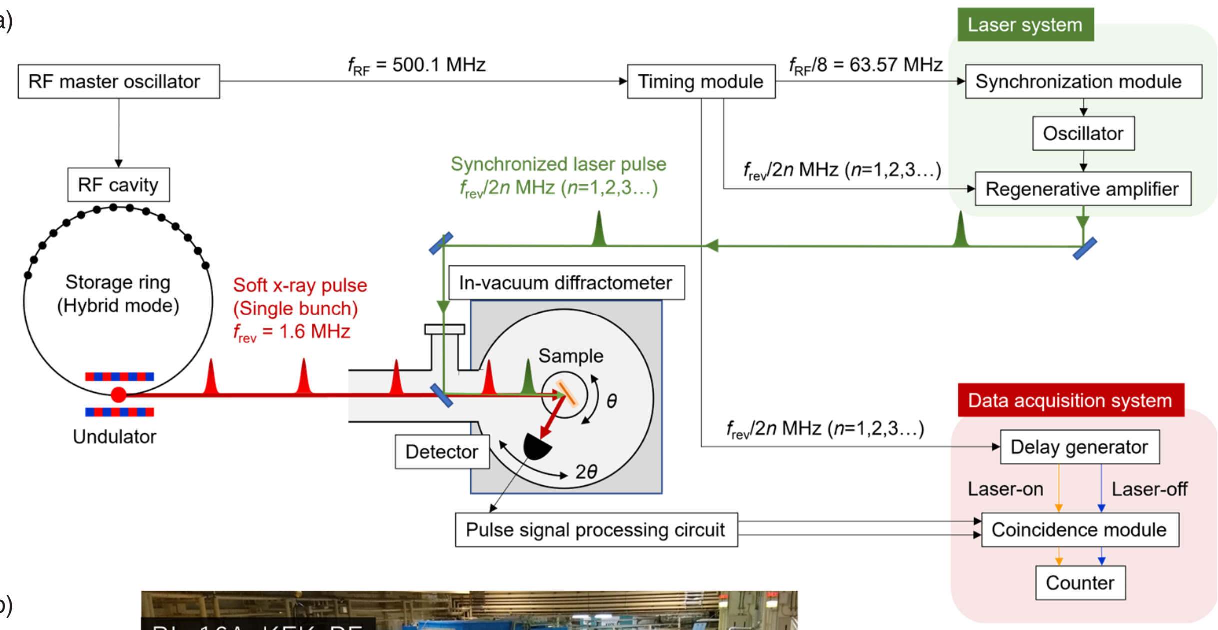
Development of High-Repetition-Rate Time-Resolved Resonant Soft X-Ray Scattering Measurement System at the Photon Factory

The high-repetition-rate high-power compact laser system has been installed to improve efficiency via flexible data acquisition to a sub-MHz frequency in a time-resolved resonant soft X-ray scattering experiment at the Photon Factory. By using the high-repetition-rate measurement system, we could demonstrate the elementally selective detection of the transient resonant soft X-ray scattering intensity variation related to the photoinduced dynamics of the magnetic order in a multiferroic manganite. The development of this system enables us to conduct time-resolved resonant soft X-ray scattering experiments on materials that were previously difficult to measure due to the weak scattering signals.

Controlling physical properties such as conductivity, electricity, and magnetism in strongly correlated electron system by using ultra-short laser pulses on femtosecond timescale is an important topic in condensed matter physics because the emergence of novel photoinduced phenomena expect the creation of a new field involving both fundamental material science and device applications [1]. To understand the physical properties, it is essential to reveal the electronic ordered states. Resonant soft X-ray scattering (RSXS) is a useful probing method that reveals the electronic order characteristics by tuning the photon energy of the X-ray to the absorption edges of the constituent elements. Therefore, time-resolved RSXS (trRSXS) using a pump and probe technique has a great advantage for capturing ultrafast dynamics of electronic order in strongly correlated electron system.

(a)



(b)

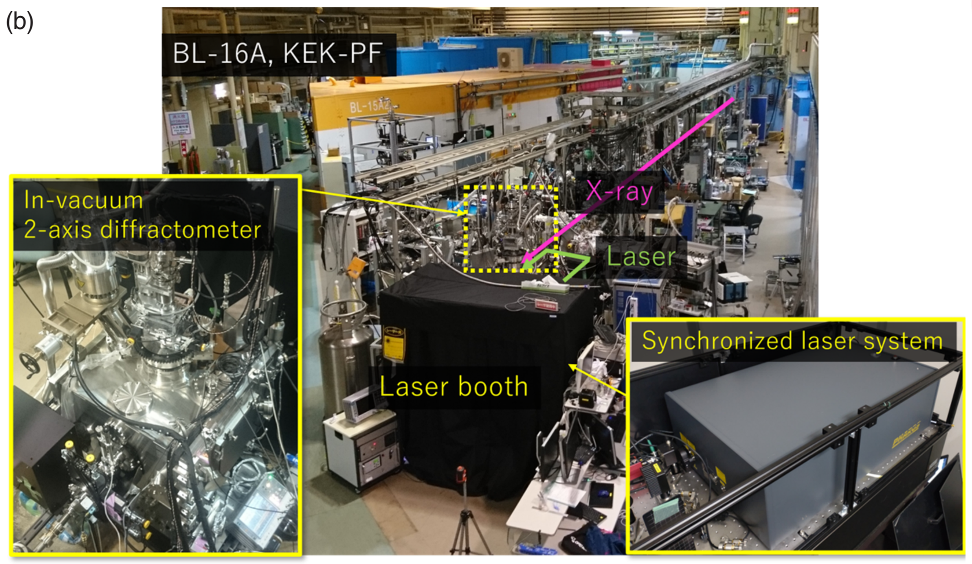


Figure 1: (a) Schematic overview and **(b)** equipment photo of the trRSXS measurement system constructed on BL-16A. The repetition rate of the laser system is synchronized with the radio-frequency (RF) master oscillator of the synchrotron radiation. The laser beam is introduced into the vacuum path passing through the soft X-ray beam and it then irradiates the sample in the in-vacuum diffractometer almost coaxially with the soft X-ray beam. The detector is a channel electron multiplier with a pulse-counting mode. The signal from the sample is divided into two signals by pulse signal processing circuit. One of two signals is recorded in integration mode with a coincidence module through an electronic gate process corresponding to the laser-on timing triggered by a digital delay pulse generator and the other to the laser-off timing.

In trRSXS experiments utilizing a synchrotron radiation, however, the average X-ray photon flux is reduced generally because of the thinning out of the synchrotron X-ray pulse train with a MHz repetition rate in accordance with a kHz repetition rate of a conventional pumping laser system. Furthermore, since the repetition rate of a conventional laser system is fixed, it is difficult to easily tune the data acquisition frequency, which depends on the repetition rate of a laser system, to suit signal intensities and sample characteristics. Therefore, the number of synchrotron facilities where trRSXS experiments can be conducted has been limited.

With recent developments in laser technology, high-power pulse laser systems with tunable repetition rate from single-shot to the order of MHz frequency which is comparable to a synchrotron radiation X-ray pulse frequency have become commercially available. Combining MHz synchrotron X-rays and high-repetition-rate laser pulses improves the data acquisition frequency of probing trRSXS signals. In this study, we constructed trRSXS measurement system using a high-repetition-rate high-power compact laser system on BL-16A and BL-19B [2]. The overview of the experimental setup for the trRSXS measurement system and the instrumental photo are shown in **Fig. 1(a)** and **(b)**, respectively. The measurement system allows the laser repetition frequency to be tuned variably up to MHz synchrotron X-ray pulse frequency, according to the RSXS signal intensity and the sample characteristics. Therefore, the trRSXS experiments can be conducted efficiently at the optimum data acquisition.

By using the trRSXS measurement system, we measured the time evolution of transient resonant soft X-ray magnetic scattering signals of $SmMn_2O_5$, which is a typical example of multiferroic material [3, 4], after the laser pulse irradiation. A schematic of the experimental geometry of the incident and scattered soft X-ray and laser on the sample with a surface normal of (1 0 0) is shown in **Fig. 2(a)**. The laser repetition frequency, i.e. data acquisition frequency, is set to 22.9 kHz which is approximately 20 times higher than that of conventional measurement system, to avoid the sample damage caused by the high repeatedly laser irradiation. As shown in **Fig. 2(b)**, the RSXS signal with the (0.5 0 0) propagation vector around Mn L_{III}-edge at 644 eV reflecting the antiferromagnetic ordering decreased just after laser irradiation, suggesting the ultrafast melting of the antiferromagnetic order within 100 picoseconds. The fitting analysis shown by red line in the figure revealed that the relaxation process contains two different decay components with decay time at approximately 7 and 100 nanoseconds, representing the green and blue lines in the figure, respectively. As shown by the blue line in **Fig. 2(c)**, however, the time evolution around the oxygen K-edge at 535 eV can be well reproduced with the fitting function only for the latter decay component with decay time at c.a. 100 nanoseconds. Although further study is currently in progress to clarify the difference in dynamics between Mn and oxygen absorption edges, these experimental results clearly demonstrate the effectiveness of using the constructed trRSXS measurement system.

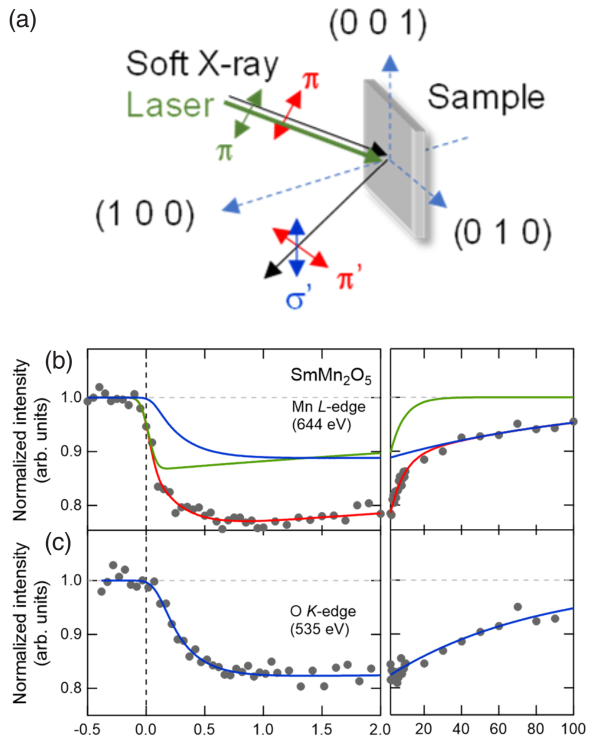


Figure 2: (a) Schematic of the experimental geometry of the soft X-ray and laser beams. **(b)** Time evolution of the (0.5 0 0) RSXS peak intensities around Mn L_{III}-edge and **(c)** around O K-edge at 535 eV measured at 7 K with a 22.9 kHz repetition rate.

The development of the high-repetition-rate trRSXS measurement system will greatly expand the range of target materials and measurement conditions. Moreover, it is expected not only to provide clear guiding principles for the development of new ultrafast control technology of physical properties using light, but also to dramatically accelerate research and development for an application of next-generation ultrafast switching and communication devices.

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BEAMLINES

BL-16A and BL-19B

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