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Combined system of synchrotron radiation and laser for solid-state research

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Two combined systems of synchrotron radiation and laser have been constructed for solid-state research. One is a laser-induced fluorescence system to observe synchrotron radiation-induced desorption of alkali atoms from ionic crystals, which consists of a laser diode with a high repetition rate and synchrotron radiation under a single-bunch operation. The other is a system of two-photon spectroscopy, which is based on the combination of synchrotron radiation pulses with a low intensity and high repetition rate and Nd:YAG laser pulses with a high intensity and low repetition rate. The experimental systems and the preliminary results are presented in this report.

Keywords: lasers; laser-induced fluorescence; two-photon spectroscopy.

1. Introduction

Since the first reports of synchrotron radiation in 1947 and of ruby laser operation in 1956, synchrotron radiation and lasers have been utilized in a variety of scientific fields. The combination of these two light sources is therefore very interesting and is a promising technique for solid-state research, but only a few pioneering studies have been reported so far. Saile (1980) measured photoelectric yields from Kr films excited by synchrotron radiation from DORIS and an N_2 laser, and observed several excitonic structures in the two-photon-yield spectra. Pizzoferrato *et al.* (1986) and Pizzoferrato & Casalboni (1987) observed two-photon absorption of KI, KCl and NaCl using synchrotron radiation from ADONE and a *Q*-switched Nd:YAG laser. Mitani *et al.* (1989) reported transient absorption of R6G dye using synchrotron radiation from UVSOR and a mode-locked Nd:YAG laser. The transient surface photovoltages induced by a pulsed laser on Si (111) were observed by means of photoelectron spectroscopy with synchrotron radiation from NSLS (Long *et al.*, 1990). The laser-induced absorption changes in far IR regions were also reported on oxide superconductors (Ederer *et al.*, 1992). In recent years, the present authors have successfully carried out two kinds of synchrotron radiation–laser combined experiments for solid-state research. In this report, the synchrotron radiation–laser combined systems are presented along with the preliminary experimental results.

2. Laser-induced fluorescence system for synchrotron radiation-stimulated desorption

A laser-induced fluorescence (LIF) experiment was carried out on alkali halide surfaces with synchrotron radiation at undulator beamline 3A1 of the UVSOR facility. Fig. 1 shows a scheme of the apparatus used for the LIF experiments. Intense quasi-monochromatic radiation of 36 eV from the undulator was used as an excitation light source. A typical photon flux of the incident synchrotron radiation was about 10^{14} photons s^{-1} mm^{-2}. Ground-state potassium atoms desorbed from the synchrotron radiation-irradiated KCl surface were excited with a laser diode (LD) (Hamamatsu PLP-02). The peak power and the pulse width of the LD were 260 mW and 30 ps, respectively. The photon energy of the LD was tuned to the transition from the ground state (4*s*) to the excited state (4*p*) of K atoms by using a potassium-gas cell. The fluorescence spectrum was observed using a

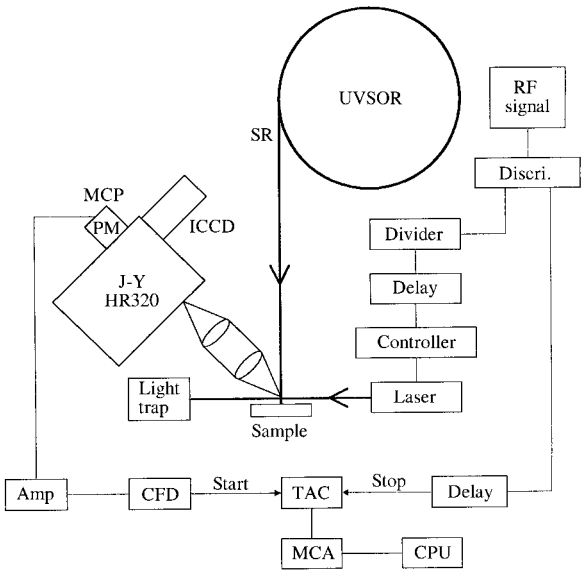

Figure 1
A schematic diagram of the LIF experiment.

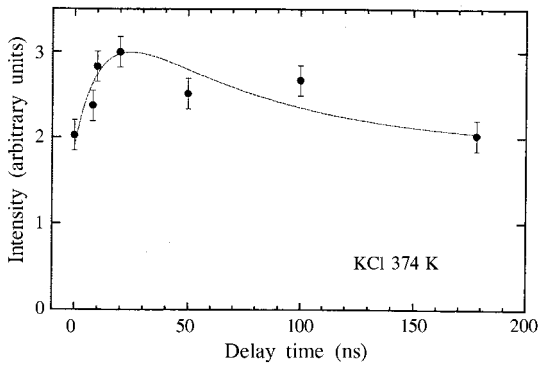

Figure 2
The time response of potassium-atom desorption from KCl.

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conventional visible monochromator (Jobin-Yvon HR-320) and a position-sensitive detector (Princeton Instruments Inc. ICCD-576). The time response of potassium-atom desorption was investigated by means of a pump-probe method with synchrotron radiation and LD pulses. The interval between successive synchrotron radiation pulses under single-bunch operation was about 180 ns and the time width of the pulses was about 500 ps. The radiofrequency signal from the UVSOR electron-storage ring was divided into 1/3 in order to synchronize the LD with synchrotron radiation pulses. The delay time between synchrotron radiation and the LD was changed with an ns-delay circuit, and the real value of each delay time was confirmed by observing scattered light of synchrotron radiation and the LD with a time-correlated single-photon counting method.

Fig. 2 shows the time response of the ground-state K desorption from KCl. It should be noted that the LIF intensity shows a peak around 20 ns and a large background. This indicates that the response time of the ground-state K desorption has a fast component in the ns range and a slow component (more than 180 ns) which is observed as the background due to pile-up. The present result is consistent with the time response of the excited-state alkali desorption, which shows a fast component in the ns range and a slow one between 180 ns and 3 ms (Hirose & Kamada, 1993). The present authors propose two desorption mechanisms: a fast one due to the lattice instability induced by an electronic transition in the surface layer (electronic process) and a slow one originating from the thermal instability of surface defects (thermal process).

3. Synchrotron radiation–laser combined system for two-photon spectroscopy

The experimental system for two-photon spectroscopy was constructed at beamline 1B. Fig. 3 shows a schematic diagram of the apparatus. A typical photon flux of the incident synchrotron radiation was about 10^{10} photons s^{-1} mm^{-2} around 100 nm. A single crystal of BaF_2 was simultaneously excited with a second harmonic (532 nm) of the *Q*-switched Nd:YAG laser and monochromated synchrotron radiation. The peak power and the pulse width of the laser were 1 MW and 5 ns, respectively. The luminescence at about 4.1 eV, which is due to self-trapped excitons (STEs), was observed using a conventional monochromator (Jobin-Yvon, HR-320) and a micro-channnel plate photomultiplier tube (MCP-PMT) (Hamamatsu, R2809U). The present detection method is a kind of zero method and therefore is more sensitive than the transmission method that was used to observe two-photon absorption of KI, KCl and NaCl by Pizzoferrato *et al.* (1986) and Pizzoferrato & Casalboni (1987).

Special care was taken in the present experiments to synchronize synchrotron radiation and laser, since synchrotron radiation and laser have different power and temporal specifications: synchrotron radiation pulses have a low intensity and high repetition rate, while the YAG laser pulses have a high intensity and low repetition rate. Fig. 4 shows a schematic timing scheme of the present system. The interval between successive synchrotron radiation pulses under the multi-bunch operation was about 11 ns and the time width of the pulses was about 500 ps. The radiofrequency signal (90 MHz) from the master oscillator of the UVSOR electron-storage ring was divided into 1/6 400 000 in order to synchronize the laser to synchrotron radiation pulses. The coincidence between synchrotron radiation and laser was achieved by controlling a timing circuit. The signals from the MCP-PMT were stored in two counters through the two-gate system. The time width of the gate system was chosen by referring to the lifetime of the luminescence. Gate 1 corresponds to the luminescence plus background signals, while gate 2 corresponds to the background signal alone. The true luminescence signal due to the two-photon excitation was obtained by numerical subtraction between the intensities of the counters 1 and 2.

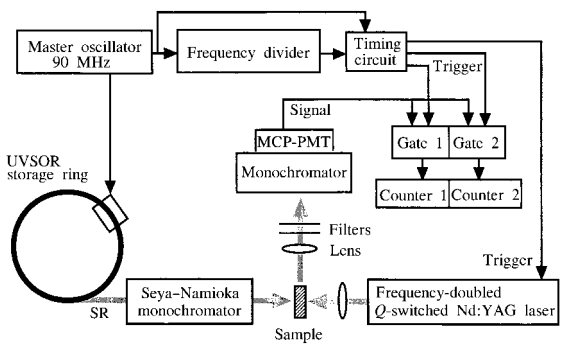

Figure 3
A schematic diagram of the experimental arrangement of two-photon spectroscopy.

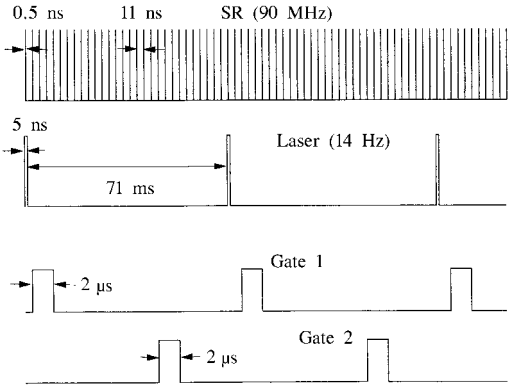

Figure 4
A schematic diagram of the timing scheme of two-photon spectroscopy. Note that the scheme is given for the different time scales for convenience.

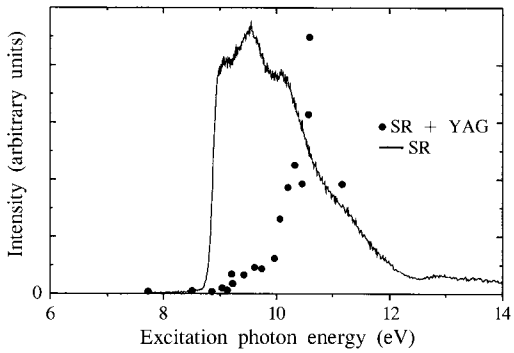

Figure 5
The two-photon excitation spectrum of BaF_2 at room temperature. The one-photon excitation spectrum is also shown by a solid curve for comparison.

Fig. 5 shows preliminary results on BaF_2, which is an interesting material since it shows so-called Auger-free luminescence under core-level excitation (Itoh *et al.*, 1988) and is useful as a scintillation material in high-energy physics. Two- and one-photon excitation spectra for the STE luminescence at room temperature are shown by solid circles and a solid curve, respectively. The two-photon excitation spectrum rises at 9.2 eV and increases towards 10.6 eV, while the one-photon spectrum shows a dip due to the 1 s exciton at 9.8 eV. Therefore, the binding energy of the exciton in BaF_2 is roughly estimated to be 0.8 eV from a simple assumption of the exciton energy levels. Detailed experiments at low temperatures are in progress using another Nd:YAG laser with a higher repetition rate and higher power.

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