IMPROVED PERFORMANCE OF THE NIJI-IV COMPACT VUV/IR FEL AND ITS APPLICATION TO THE SURFACE OBSERVATION*

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Abstract

Increase of the transmittance of the out coupling mirror as well as e-beam energy brought a sufficient free-electron laser (FEL) power around 200 nm for a real-time surface observation. By irradiating surfaces of transition-metals, such as Chromium and Palladium, with deep-ultraviolet (DUV) FEL, fine structure and surface chemical reaction were successfully observed with spatial and temporal resolutions of ~ 300 nm and 33 ms, respectively. Every efforts to shorten the lasing wavelength below 190 nm are being made continuously by correcting magnetic-field errors in the optical klystron and developing cavity mirrors with a smaller optical loss. A new optical klystron for use in the infrared (IR) region was installed into the north straight section of the NIJI-IV to extend the lasing range up to ~ 10 μm.

INTRODUCTION

At AIST, an FEL with ultra-wide wavelength range from the vacuum ultraviolet (VUV) to the far infrared (FIR) is being developed based on a compact storage ring NIJI-IV. Though its shortest lasing wavelength is still limited to 198 nm because of an unexpected gain smaller than that estimated by the analytical calculation [1], a sufficient margin of the gain enabled us to obtain one-order larger FEL power than before, by adopting an output-coupler with a higher transmittance as well as increasing the stored beam energy in the ring.

Since the work function of transition-metals lies around 5 eV, DUV FELs with a wavelength of ~ 200 nm are suitable as a intense light source to observe surface fine structure or surface chemical reactions on such transition-metals with high spatial and temporal resolutions in combination with the photoelectron emission microscopy (PEEM). The intensity of NIJI-IV FEL recently obtained is large enough to extract sufficient amount of photoelectrons for sub-micron scale and video-rate real-time observation from transition-metal surfaces.

Unexpectedly small FEL gain is probably caused by some demagnetization of magnet blocks in the 6.3-m optical klystron, which may deflect the beam and reduce the overlapping between electron beam and optical mode. We are preparing high performance dielectric multilayer mirrors optimized around 190 nm to compensate such an insufficient gain. We also plan to replace or remagnetize such degraded magnet blocks.

A 3.6-m optical klystron for lasing in the IR was recently installed into the north straight section of the NIJI-IV to extend the lasing wavelength range up to ~ 10 microns. One of our interests in the IR-FEL application is the microscopic Raman spectroscopy which can examine adsorbed molecules and their bonding conditions on the metal surfaces. Establishment of a total surface analysis system using NIJI-IV compact VUV/IR FEL combined with characteristic surface analysis techniques is one of our goals in the near future.

PERFORMANCE OF THE NIJI-IV FEL

We have been modifying the NIJI-IV FEL system to extend its lasing range. As shown in our previous paper [2,3], replacement of the NIJI-IV vacuum chambers to low-impedance-type ones as well as installation of thin sextupole magnets into the ring brought a higher peak current of the stored beam and a resultant higher FEL gain. This led to easy FEL lasing around 200 nm even with our compact system. To utilize such a DUV FEL, we tried to enhance the laser output power by increasing the transmittance of the output coupler of the optical cavity from 0.05 to 0.5 %. The beam energy was also increased by 10 % from 310 up to 340 MeV. As a result, the average output FEL power at 202 nm was successfully increased up to 0.5 mW per one laser port with a beam current of 15 mA, which is larger by an order of magnitude than before [2]. A higher power is expected through further increase of the beam energy as well as multi-bunch operation of the ring. Fig.1 shows the dependence of average output power on the beam energy.

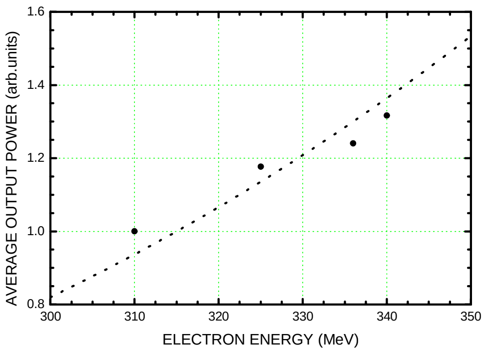

Fig.1. Dependence of average output power on the beam energy. Solid circles and solid curve indicate the observed power and an expected curve proportional to the 4th power of the beam energy.

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Solid circles and curve respectively indicate the observed FEL power at 202 nm and an expected energy dependence of the laser power which can be proportional to the 4th power of the beam energy. In this figure, the observed average power was estimated from the peak height of the lasing line spectrum detected with a photodiode array attached to a 25-cm monochromator. Here, the peak height was corrected for the beam current. Absolute power was measured with a highly sensitive and stable CW power meter (Gentec TPM-310) with a wind block. It is found in Fig.1 that the observed output power was enhanced by at least 30 % during 10-% increase of the beam energy, which does not largely deviate from the expected curve. Now we plan to increase the beam energy from 0.31 up to at least 0.4 GeV which will bring three times power enhancement. A few-mW power in the DUV to the VUV range is in a sufficient level for PEEM measurement.

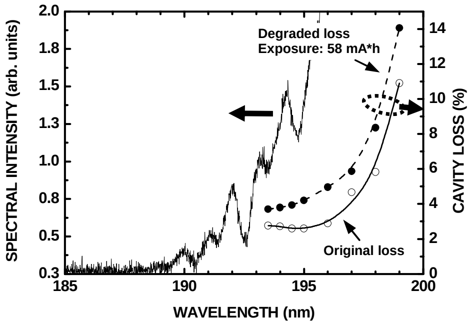


Fig. 2. Cavity-loss as a function of wavelength. A spectrum of the spontaneous emission resonating inside the optical cavity is also indicated.

Lasing around 190 nm and below is also being tried using a low-loss optical cavity composed of two Al_2O_3/SiO_2 mirrors. Though the original loss of this cavity was 2.8 % at 193 nm, it rapidly increased up to 3.7 % with 58 mA-h exposure to the undulator radiation. Due to an unexpectedly smaller FEL gain, probably caused by some demagnetization of the undulator magnets, we have not obtained lasing around 190 nm yet. Now we are preparing robust mirrors with an optical loss smaller than 1 % and also planning to replace the degraded magnet blocks in the optical klystron. Fig.2 shows the loss curve for the above mentioned cavity as a function of wavelength. A spectrum of the spontaneous emission resonating inside the optical cavity, observed at a beam current of ~ 7 mA, is also indicated in the figure. Though the spectrum is strongly distorted by intense long-wavelength off-axis components and light absorption by the Schumann-Runge system of Oxygen molecules around 193.8 nm, spectral modulation peculiar to an optical klystron is clearly seen. Since such a spectral modulation becomes much more obvious and even spiky for a beam current more than ~ 14 mA, the lasing seems to be achieved soon.

A new optical klystron for lasing in the IR was installed in the north straight section of the NIJI-IV. Its performance is being checked spectroscopically [4].

APPLICATION OF NIJI-IV DUVFEL TO SURFACE OBSERVATION

NIJI-IV DUVFEL was applied to surface observation, in combination with a PEEM system (STAIB Instrumente, type 350). This PEEM system has three sets of electrostatic electron lenses and a micro channel plate (MCP) equipped with a fluorescent screen. By viewing the focused images on the fluorescent screen with a CCD camera, transient phenomena, such as chemical reactions on transition-metal surfaces, can be monitored with video-rate time resolution. Since the spatial resolution of this system is 80 nm and our FEL intensity is large enough to extract sufficient amount of photoelectrons to recognize the surface contrasts within 33.3 msec, we can examine real-time physical and chemical information on transition-metal surfaces in a sub-micron scale. Fig.3 shows the experimental setup for FEL-PEEM measurement. The FEL with a wavelength of 202 nm comes from left-hand side through a long focus lens and is reflected by a flat aluminium mirror to focus onto the sample surface. The FEL intensity on the sample surface is roughly estimated to be 500 mW/cm^2.

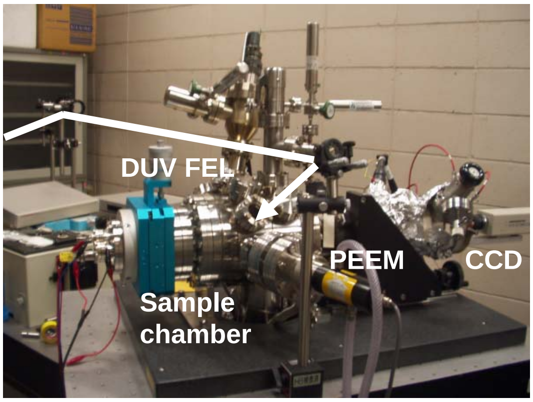
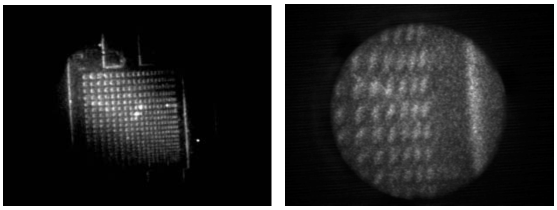


Fig.3 Experimental setup for FEL-PEEM measurement.

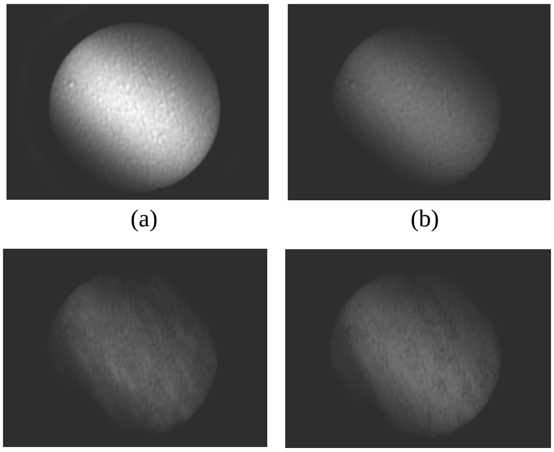
To examine the total resolution of our FEL-PEEM system, we observed a standard reference sample for scanning electron microscope (SEM) which has a fine structure formed with 70-nm thick CrO_2 on a Cr-overcoated SiO_2 substrate. A typical example of the video-rate real-time imaging of the standard SEM sample is shown in Fig.4 (a) and (b). It was found that the FEL was intense enough to obtain video-rate moving images on the Cr surface. From higher magnification image (Fig.4 (b)), the spatial resolution of our system is estimated to be ~ 300 nm at present. Suppression of mechanical vibration from a vacuum pump will be necessary to realize the specification of the PEEM system.



(a) (b)

Fig. 4. Typical example of the video-rate real-time imaging of the standard SEM sample for lower (a) and higher (b) magnifications. The narrowest feature width is 1 μm in both images.

As a preliminary experiment for the FEL-PEEM measurement, we observed a Palladium surface, introducing CO and O_2 gasses. In this case, CO_2 is created from adsorbed CO and O_2 molecules under the catalysis of Palladium and desorbed from the Palladium surface. Such a reaction process must be reflected in the contrast of the PEEM image [5]. Fig.5 (a) – (d) shows still shots for four deferent situations, sampled out from a real-time moving image. After cleaning with Argon sputtering and annealing at 1,100 K in a high vacuum below 5 x 10^{-7} Pa, pure surface of a Palladium standard sample (from Mateck) was exposed to the CO gas to be covered with CO molecules. Then, Oxygen gas was introduced to be reacted with adsorbed CO molecules to produce CO_2 which will desorb immediately. Fig.5 corresponds to the FEL-PEEM images for pure surface after cleaning (a), a CO-adsorbed surface (b), surface reaction of CO and O_2 (c), and the same surface reaction at a different moment (d). In these images the full width of the viewing field is 200 μm. During the measurement, the FEL was lasing at 202 nm with a beam current between 11 and 8.5 mA In Fig.5 (a), many crater-like structures are scratches which remains on the polished surface originally. One can see in Fig.5 (b) that the brightness of the image was reduced through the adsorption of CO molecules over the whole surface. On the other hand, the images indicated clearly different features after introducing Oxygen gases, Fig.5 (c) and (d), where a complicated black and white blindle appeared. The black small patches seen around upper right area of (d) imply Oxygen-adsorbed regions, while several bright islands seen from upper left to lower right of (c) seems to be the areas where the adsorbed CO molecules were desorbed to form CO_2 through chemical reaction with O_2. These patterns varied rapidly in a-few-second time scale and were observed only during FEL lasing. More detailed observation is being carried out under various conditions, such as different gas pressure and substrate temperature.



(a) (b)

(c) (d)

Fig. 5. Still shots, sampled from a real-time moving PEEM image, on a Palladium surface under deferent conditions; after surface cleaning by both Argon sputtering and annealing (a), with the surface covered by CO molecules (b), and during reaction of CO with O_2 (c)(d).

SUMMARY

Output power of the NIJI-IV FEL was enhanced by an order of magnitude in the DUV region around 200 nm. The DUV FEL was utilized to make real-time imaging of fine structures and chemical reactions on transition-metal surfaces in combination with a PEEM system. Moving images of reactions by CO and O_2 on a Palladium surface were visualized with video-rate time resolution. More detailed FEL-PEEM experiment is being carried out at present. Efforts to obtain FEL lasing at a wavelength shorter than 190 nm are being continued. Lasing in the IR using our compact ring is also expected to be realized. A total real-time analysis system based on the NIJI-IV compact VUV/IR FEL will be established in combination with characteristic surface analysis techniques in the near future.

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