

ACTIVITY REPORT

2014

Hiroshima Synchrotron Radiation Center, HiSOR Hiroshima University

HiSOR ACTIVITY REPORT

2014

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Edited by H. Sato

The annual report is available from

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Preface

The Hiroshima Synchrotron Radiation Center (HiSOR) promotes joint research with researchers from Japan and overseas, and human resource development as a "Joint Usage and Research Center (2010 - 2015)" in the field of synchrotron radiation (SR) science, authorized by the Minister of Education, Culture, Sports, Science and Technology.

The HiSOR has played pioneering work in the research of materials science centered mainly focused on solid state physics using SR in the range between ultraviolet and soft x-ray. Every March, the HiSOR widely communicates their research results with the international society through "Hiroshima International Symposium on Synchrotron Radiation".

The high-resolution angle-resolved photoemission spectroscopy (ARPES) at BL-1 and BL-9A, and highly-resolution spin ARPES at BL-9B using the low-energy SR have been recognized as significantly powerful tools for the precise observations of electronic and spin states of solids such as pure metals, high T_c superconductors and topological insulators. These experiments contribute to clear understanding of the macroscopic physical properties from the direct evaluations of interactions between electrons and elementary excitations (phonons, magnons), and degree of spin polarizations. In the research activity in 2014, especially the research on the electronic states of MoS_2 related to the valleytronics shows the high resolution ARPES and the high resolution spin ARPES have great ability to open new fields of materials science.

The HiSOR has been developing the experimental techniques related to the high resolution ARPES and the spin ARPES. The international review committee in 2012 pointed out that HiSOR is doing exceptionally well in continued upgrades and incessant maintenance of beamline components and end-stations, and this has been a key for the great success of the scientific programs at the facility up to now. The HiSOR research colleagues pointed out that the higher spatial resolution of ARPES is important to focus on the small samples or the single crystal surface on small part of cleaved sample for advanced experiments. The council of Joint Usage and Research Center of HiSOR has been discussing about facility upgrade plan to be able to respond to the users voice to realize advanced materials science. The research community proposed the introduction of VUV laser to start R&D of the low emittance SR experiment before renewal of the HiSOR. In 2014, the VUV laser systems and end-stations have been introduced for BL-9 experiment and the R&D experiment. The laser system for BL-9 is used for the advanced

experiment at BL-9A or BL-9B alternately at its unusable SR beamtime. The laser ARPES and laser spin ARPES systems were installed in the preparation hall. The laser ARPES shows good spatial resolution using simple focusing lens. The higher energy resolution (~250 μ eV) and high momentum resolution (~0.0048 Å⁻¹) was achieved at first R&D experiment. The laser spin ARPES system using new ARPES analyzer (VG-SCIENTA DA30) with 3D spin detector system was installed neighbor area of the new ARPES system sharing the same laser source.

Finally, international network for joint research expanded rapidly to 89 institutions (56 domestic and 33 overseas), mainly due to high performances of electronic and spin state analysis of solids as well as VUVCD analysis of biomolecules.

August 2015

Hirofumi Namatame

Director for Hiroshima Synchrotron Radiation Center

Hirofumi Namatame

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Current Status of HiSOR

Status of the HiSOR storage ring

1. Introduction

The HiSOR is a synchrotron radiation (SR) source of Hiroshima Synchrotron Radiation Center, Hiroshima University, established in 1996. It is a compact racetrack-type storage ring having 21.95 m circumference, and its natural emittance of 400π nmrad is rather large compared with those of the other medium to large storage rings. The most outstanding advantage of the facility lies in good combination with state-of-the-art beamlines (BL's) for high-resolution photoelectron spectroscopy in the photon energy ranges between VUV and soft X-ray. The principal parameters of HiSOR are shown in Table 1.

Table 1. Main parameters of the HISOR Storage ring.			
Circumference	21.95 m		
Туре	Racetrack		
Bending radius	0.87 m		
Beam energy at Injection	150 MeV		
at Storage	700 MeV		
Magnetic field at Injection	0.6 T		
at Storage	2.7 Т		
Injector	150 MeV Racetrack Microtron		
Betatron tune (v_x , v_y)	(1.72, 1.84)		
RF frequency	191.244 MHz		
Harmonic number	14		
RF voltage	200 kV		
Stored current (nominal)	350 mA		
Natural emittance	400π nmrad		
Beam life time	~10 hours@200 mA		
Critical wavelength	1.42 nm		
Photon intensity (5 keV)	1.2×10^{11} /sec/mr ² /0.1%b.w./300mA		

Table 1: Main parameters of the HiSOR Storage ring.

The originally designed maximum stored current of HiSOR was 300 mA. However, after the improvement of the control system and the RF system in 2003, HiSOR has been in operation with 350 mA maximum stored current since. Fig. 1 shows an example of typical one-day operation. Beam injection for HiSOR is executed twice a day, at around 9:00 and 14:30.



Fig. 1: Typical daily operation status.

HiSOR has two 180-deg. Normal-conducting bending magnets which generate a strong magnetic field of 2.7 T. This storage ring is equipped with two insertion devices, a linear undulator and a quasi-periodic APPLE-II undulator which replaced to the previous helical undulator in summer 2012. Major parameters of these undulators are listed in Table 2. The photon energy spectra of the SR from HiSOR are shown in Fig. 2.

ruble 2. Wall parameters of the undulators.				
Linear undulator (BL-1)				
Total length	2354.2 mm			
Periodic length λu	57 mm			
Periodic number	41			
Pole gap	30-200 mm			
Maximum magnetic field	0.41 T			
Magnetic material	Nd-Fe-B (NEOMAX-44H)			
Quasi-Periodic APPLE-II				
undulator (BL-9A,B)				
Total length	1845 mm			
Periodic length λu	78 mm			
Periodic number	23			
Pole gap	23-200 mm			
Maximum magnetic field	0.86 T (horizontal linear mode)			
	0.59 T (vertical linear mode)			
	0.50 T (helical mode)			
Magnetic material	Nd-Fe-B (NEOMAX-46H)			

Table 2: Main parameters of the undulators.



Fig. 2: Photon energy spectra of the SR from HiSOR.

2. Operation status in FY 2014

Fig. 3 shows monthly operation time of HiSOR storage ring in FY 2014. HiSOR has a long term cease period to have routine inspections in summer every year. The total user time of FY2014 achieved 1221 hours. Fig. 4 shows bar graph of total operation days in each fiscal year. It shows that days for user operation as well as the total operation days in FY2014 was decreased similar to that in FY2012 in comparison with an average year. Operation times of the storage ring and the Microtron from FY 2005 to FY 2014 are shown in Fig. 5.

In regard to the total operation time of except FY2012 and FY2014, it has been kept in almost constant hours, around or over 1800 hours, and user time was over 1500 hours in each Fiscal Year consecutively for five years from 2007 since the accelerator system has been well maintained. However, it has been only 1119 and 1221 hours in FY2012 and FY2014, respectively due to four (three in FY2012, one in FY2014) vacuum leak incidents to the storage ring chamber. After incidents in FY2012, the water-cooled SR absorbers in bending magnet sections was replaced to new ones during the summer shutdown period in 2013. As the result, the operation time and user time in FY2013 recovered to the almost same level as the average though the beam life time has shortened and radiation dose increased due to worse vacuum, and hence. The user run was conducted at the maximum current about 250 mA after January, 2013. The vacuum failure due to a water leak from a replaced new absorber happened again one week after starting the fall user run in October 2014. And, hence, the user



run ceased till the end of December and resumed in January, 2015.





Fig. 4: Operation days of HiSOR storage ring.



Fig. 5: Annual operation time of Storage ring and Microtron.

Beamlines

A total of 13 beamlines has been constructed so far; three normal-incidence monochromators, seven grazing-incidence monochromators, two double crystal monochromators and apparatus for white beam irradiation (Fig. 1). Table 1 lists the beamlines at present together with the main subject, energy range and monochromators.

beamline	source	monochro- mator	subject	energy range (eV)	status
BL-1	LU	GIM	Polarization dependent high-resolution ARPES	22-300	in use
BL-3	BM	DCM	Surface XAFS	1800-3200	in use
BL-4	BM		White beam irradiation		closed
BL-5	BM	GIM	ARPES and PEEM	40-220	in use
BL-6	BM	GIM	Gas-phase photochemistry	200-1200	in use
BL-7	BM	GIM	ARPES	20-380	in use
BL-8	BM		Beam diagnosis		
BL-9A	HU/LU	NIM	UV-VUV high-resolution ARPES	5-35	in use
BL-9B	HU/LU	GIM	High-resolution spin-resolved ARPES	16-300	in use
BL-11	BM	DCM	XAFS	2000-5000	in use
BL-12	BM	NIM	VUV-CD of biomaterials	2-10	in use
BL-13	BM	GIM	Surface photochemistry	60-1200	in use
BL-14	BM	GIM	Soft-XMCD of nano-materials	400-1200	in use
BL-15	BM	NIM	VUV-CD of biomaterials	4-40	closed
BL-16	BM		Beam profile monitor		

Table 1: List of Beamlines

At present, nine beamlines BL1, BL3, BL6, B7, BL9A, BL9B, BL11, BL12, BL13 and BL14 are opened for users. Furthermore, three off-line systems, spin- and angle-resolved photoemission (SARPES) spectrometer, inverse-photoemission (IPES) spectrometer and low-temperature scanning tunneling microscope (LT-STM) system have also opened for users (Fig. 2).



Fig. 1: Schematic view of the experimental hall.



Fig. 2: Experimental apparatus for Joint usage and Joint research. (a) BL-1, (b) BL-3, (c) BL-6, (d) BL-7, (e) BL-9A, (f) BL-9B, (g) BL-11, (h) BL-12, (i) BL-13, (j) BL-14, (k) Spin resolved photoemission (offline), (l) Resonant inverse photoemission (offline), (m) LT-STM (offline).

Research Activities

-Accelerator Studies -

Magnetic design of Knot-APPLE undulator for HiSOR-II and its performance

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Keywords: Undulator

In order to generate a low energy photon beam in a high energy storage ring with an insertion device, it is necessary to increase the undulator's deflection parameter, K. If this is the case, a high heat load on beamline elements is a serious problem because the on-axis radiation power increases drastically as increasing the value of K parameter for a linear undulator. To reduce the on-axis heat load , the Figure-8 undulator, the Knot undulator and other exotic undulators were proposed [1-4].

In a Figure-8 undulator, the period length of horizontal magnetic field is twice as large as that of vertical field, and hence the projected beam orbit draws a figure of number 8. The direction of velocity vector of electrons always deviates from the undulator axis, and projected motion of electrons draws alternately a clockwise and anti-clockwise orbit, which results in the cancellation of circular polarization of the photons and leaves a remanent linear polarization.

In a Knot undulator, the magnetic field period in horizontal direction is 1.5 times larger than that of the vertical direction. The direction of velocity vector never coincides to but rather always deviates from the undulator axis. The spatial distribution of corresponding power density draws a knot-like figure.

However, neither Figure-8 nor Knot udulator has the capability to change polarizations. On the other hand, the APPLE undulator is capable to generate variable polarization, but is not capable to reduce on-axis power density in linear modes. We propose a novel Knot-APPLE undulator which is capable to reduce an on-axis high heat load and generate every polarization state. Firstly, we designed Knot-APPLE undulator for Shanghai Synchrotron Radiation Facility (SSRF). Figure 1 shows comparison of photon flux densities. Obviously, intensity of higher harmonic radiation is much smaller in the Knot-APPLE undulator than that in the planar undulator.



FIGURE 1. This is comparison of radiation spectra, solid line represents flux density from a Knot-APPLE undulator in horizontal linear mode and dotted line represents it from a planar undulator. Calculation was performed using SPECTRA based on the storage ring of SSRF.

Second, we modified magnet structure suitable for low energy storage ring such as the HiSOR-II ring to evaluate the performance of Knot-APPLE undulator. In the presentation, a new magnetic structure of Knot-APPLE undulator and its expected radiation spectrum and power density are presented.

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Magnetic design and performance evaluation of variably polarizing Leaf undulator

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Keywords: undulator

To generate the radiation having a low photon energy from a linear undulator inserted in a high-energy electron storage ring such as the SPring-8, it is necessary to increase the value of undulator parameter, K, which is proportional to the peak magnetic field B_0 . With a high K-value, the harmonic intensity of the radiation on the beam axis is increased. As the result, the heat load on optical elements installed in the beam-line is increased which may cause serious problems for synchrotron radiation user's experiments. In this context, a Figure-8 undulator was developed. The Figure-8 undulator is capable of reducing the on-axis power density due to higher harmonics by displacing them to off-axis. However, there is still a room for improving its performance by modifying magnetic structure.

Now, we proposed a realistic magnetic structure for a Leaf undulator with the performance that exceeds the capability of the Figure-8 undulator, and discuss about various radiation performances including spectra, polarization properties, power density distributions. Under these circumstances, an ideal magnetis field distribution the Leaf undulator was proposed by Qiao, et. al. By devising the layout of magnetic blocks in the Leaf undulator, the electron beam draws more of an ideal trajectory, and hence the on-axis radiation may contain even less higher harmonics than those from a Figure-8 undulator. Furthermore, unlike the Figure-8 undulator, magnetic period lengths of vertical(y) and horizontal(x) components are the same. Each single period contains 3×2 magnet blocks, an empty space of 3/10 period to introduce a phase shift of field distribution, and an empty space of 1/10 period to compensate peak field variations. Thus, both the vertical and horizontal magnetic fields have $\pi/2$ phase shift of sinusoidal variations alternately(Leaf mode). And then, by shorten the 3/10 empty space to 1/10, this device can generate circularly polarized radiation(Helical mode).



FIGURE 1. (a), (b) show magnetic structures for Leaf mode and Helical mode, respectively.



FIGURE 2. (a), (b) show magnetic field distributions for Leaf mode and Helical mode, respectively.



FIGURE 3. (a), (b) show expected on-axis radiation spectra for Leaf mode and Helical mode, respectively.

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Design for New Type Quasi-Periodic Undulator

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The synchrotron radiation spectrum generated by a periodic linear undulator contains higher harmonics of integral multiple of fundamental harmonic in the spectrum. These higher harmonics are usually an obstacle to user's experiments. Therefore, to reduce higher harmonic contamination, such a user has to use many optical elements such as monochromator, band-pass filter, mirror to eliminate unwanted harmonics. However, such kind of beamline setup from an undulator to an end-station may cause a reduction of photon beam intensity. In order to ease such difficulties, quasi periodic undulator was developed in which higher harmonics appear at irrational positions instead of rational positions in the spectrum. A combination of a quasi-periodic undulator (QPU) and a single monochromator can provide purely monochromatic photon beam for end-users.

Until today, quasi periodic undulators used in all over the world are designed such that the peak magnetic field strength at certain positions representing the quasi-periodicity is reduced to introduce a smaller phase advance of photon wave at every QP position in an undulator. In this study, we design a new type quasi periodic undulator by increasing the magnetic field strength at every QP position to introduce a larger phase advance of photon wave. Figure.1 shows the magnetic structure of new QPU. The field variation of this undulator is presented in Fig.2. The spectral feature of this new QPU is shown in Fig.3.



Fig.1. RADIA model of a new QPU



Fig.2. Magnetic field distribution of new QPU



Fig.3. Radiation spectrum from new QPU calculated by the code SPECTRA

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Research for introducing a perfect quasi-periodicity for all polarization modes in an APPLE-II undulator.

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keyword: undulator, synchrotron radiation, accelerator

In general, a periodic undulator emits quasi-monochromatic radiation that contains odd-integer higher harmonics in addition to the fundamental radiation even if the undulator is installed in low emittance ring. In many case, it is not so easy to eliminate unwanted higher harmonics by using a single grating monochrometer. In order to solve this problem, a scheme of QPU(Quasi-Periodic Undulator) was proposed ^[1,2]. The QPU is realized by modifying the magnetic structure so that the sequence of phase slip in each half period is corresponding to the order of quasi-periodicity^[3]. The concept is valid for any undulator scheme including a variably polarizing undulator, an electromagnetic undulator, and any exotic type of undulators. In this paper, we investigate a realistic magnet structure of a quasi-periodic variable polarization APPLE-II undulator and calculate the magnetic field distribution at respective polarization that APPLE-II can make. But we cannot solve this problem in vertical polarization mode even if the concept is installed. In figure 1 and figure 2, the magnetic distributions of previous and new quasi-periodic undulator models in vertical polarization modes are shown, respectively.





Fig.1 magnetic field distribution of previous model.

Fig.2 magnetic field distribution of new model.

By using calculated field distributions in various polarization modes, we calculate spectra and evaluate the performance of this undulator including the fundamental peak intensity degradation

and polarization by assuming the undulator is installed in the next generation light source HiSOR-II storage ring.

3.0x10¹² From Fig.1 -From Fig.2 [phs/s/mm²/0,1%b.w.] 2.5 Flux Density 2.0 1.5 1.0 0.5 ď 0.0 3 5 1 7 $\epsilon_{ph}/\epsilon_{1st}$

Fig.3 shows the spectral radiation from undulator of Fig.1and Fig.2.

Fig.3 Radiation spectra of previous model and new model.

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Approaches to detect the photon phase of bending magnet radiation by interference measurements

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Keywords: HiSOR, photon phase, bending magnet radiation.

When the particles having relativistic velocity move in the bending magnetic field, the particles receive a force in a direction perpendicular to both the direction of the velocity and the magnetic field by the Lorentz force. As the result, particles emit radiation in the forward tangential direction of trajectory. Although the bending magnet radiation has only the horizontally polarized component in the orbit plane, the off-plane radiation has vertically polarized component as well. The wavefront phases upper and lower radiation are shifted by $\pm \pi/2$ in respect to that of the horizontally polarized radiation on the plane. The upper and lower off-plane radiations are elliptically polarized with opposite helicity.

This study was conducted with the interference experiment of light in the visible region at the beam line BL-8 of HiSOR. In the experiment, a band-pass filter, a polarizer, a double slit, and a CCD camera were used. With this set-up, images of the interference fringes were captured by a CCD camera, and difference between the interference pattern of the vertical component and the horizontal component was observed. In this experiment, we attempted to verify a π difference of the wavefront phases between vertical polarization components above and below the electron orbit plane.

Figure 1 shows the intensity distributions of observed interference fringes. The horizontal axis represents the pixel number of CCD camera, and the vertical axis represents intensities of interference fringes. Different color lines represent the polarizer angle.

Although a clear reversed interference pattern has not been observed, this experimental technique may be used to detect the phase information related to the light's orbital angular momentum in higher harmonics from a helical undulator.



Figure 1. The intensity distributions of observed interference fringes.

Research Activities

- Instrumental Developments -

LASER SPIN-ARPES

STATUS OF LASER SPIN-ARPES

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Keywords: spin detection, spin mapping

In the past year, a project "LASER-SPIN ARPES" was developed. The project consisted in designing, assembling and partial commissioning of spin and angle- resolved photoemission analyzer. The analyser combines the efficiency of the spin VLEED detectors [1, 2] with the new functionalities of DA30L SCIENTA hemispherical analyser [3].

DA30L SCIENTA analyzer has horizontal and vertical deflectors to perform band and spin mapping in k space ($\pm 15^{\circ}$ from normal emission) of the full or partial surface Brillouin zone without the need to rotate the sample. The photoelectrons are channeled through single entrance aperture and via a lens system to both spin resolving detectors, unlike previous setup that uses of two apertures and spin transfer lenses [2]. The acquisition from the two VLEED detectors (BLACK and WHITE) is in parallel or in a single channel as determined by the switch parameters. The experimental apparatus will be accessorized by two photon sources: He lamp and laser of photon energy 5.5-6.5 eV, currently with *s*-polarized light, spot size 0.4 mm (with aperture) and with the possibility to be focused down to some 10 μ m² (with lenses) and pulse width ~10 ps.

The analyzer is hosted in a new chamber, connected in UHV to surface sample preparation facilities. The analyzer and new chambers were delivered in March 2014.

Status up to February 2015

Source: Non monochromatized He lamp is used as photon source for the apparatus.

Sample positioning and temperature: The main chamber was accessorized by a 4 axes manipulator that can be cooled down up to 100K with liquid N.

DA30L analyser: The analyser was commissioned and the performance of the horizontal and vertical deflector lenses was determined by acquiring the band mapping of Bi_2Se_3 at 21.2 eV. Energy resolution tests were made at 100K on Ag(111).

Spin detectors: Two VLEED spin detectors were assembled, installed and commissioned by measuring spin-resolved spectra of thin Bi film on Si(111).

VLEED preparation chamber:

The VLEED preparation chamber was equipped with two target heating stages, LEED optics, Fe evaporator and O gas line.

Preparation chamber:

The chamber is accessorized by a 4 axes manipulator, ion gun, LEED, evaporator ports, fast entry of sample and sample bank. Sample preparations available are: direct current heating for semiconductors and annealing through electron bombardment.

Further evolution plan

Main chamber: Installation of 5 axes manipulator with possibility to cool down up to 10K. Photon source: laser with *s* polarized light.

Determination of Sherman function



FIGURE 1. (a) Picture of the Laser Spin-ARPES lab. (b-d) In-plane spin polarization of Bi film (2-4ML) grown on Si(111) measured along $\overline{K} - \overline{\Gamma} - \overline{K}_{\text{(b-d) and}} \overline{M} - \overline{\Gamma} - \overline{M}_{\text{(e-f)}}$ directions with WHITE and BLACK VLEED detectors respectively.

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Current Status of Tunable VUV-laser-based µ-ARPES System at HiSOR

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Keywords: ARPES, Photoemission micro-spectroscopy

Angle-resolved photoemission spectroscopy (ARPES) plays an important role for the investigation of the physical properties of solids. Owing to greatly improved energy and momentum resolution of modern ARPES, one can now not only directly determine the energy (ω) and momentum (k) distribution of electrons, but also evaluate k-resolved energy-gaps, and many-body interactions in solids [1]. Since high-resolution ARPES can be achieved mostly using vacuum-ultra-violet (VUV) light source such as synchrotron radiation and gas discharge lamps, the surface quality of samples becomes a critical issue in VUV-ARPES experiments due to the significantly short mean free path (~10 Å) of photoelectrons in the VUV-region [2]. The sample surface for VUV-ARPES experiments can be obtained from *in situ* preparation in ultra-high vacuum by cleaving, fracturing, scraping, or sputtering and annealing the sample. However, it is not always possible to obtain a clean, flat, and homogeneous sample surface. The prepared sample surface often possesses micron-order defects, distortions, and impurities, all of which generally leads to the broadening of the spectral features and increase ambiguousness of ARPES data. One of the most straightforward ways to overcome this problem is the use of the well-focused light with a spot size smaller than a clean, flat, and homogeneous portion of the sample surface. We have therefore started to construct and develop a tunable VUV-laser-based μ -ARPES system at Hiroshima Synchrotron Radiation Center (HiSOR).

We adopted the commercial VUV laser system which can provide tunable low-energy photons from 191 nm (6.49 eV) to 210 nm (5.90 eV) with the high photon flux higher than $\sim 10^{14}$ photons/sec. ARPES with lowenergy photons has advantages of the longer electron escape depth (the increased bulk sensitivity) and the superior energy and momentum resolutions [3]. To reduce the energy broadening due to the space charge effects, the repetition rate of the laser as high as 80MHz was chosen. Furthermore, the pulse duration (pulse width) longer than 10 ps was selected for narrower spectral width of the laser light: the spectral width was expected to be smaller than 66 μ eV according to the Heisenberg uncertainty principle. By using this tunable VUV-laser, we have obtained extremely high energy and momentum resolutions (better than 0.5 meV and 0.0048 Å⁻¹), judging from the Fermi edge of evaporated gold (Fig.1) and the full width half maximum of momentum distribution curve at the Fermi level of the ARPES spectrum of optimally doped Bi₂Sr₂CaCu₂O_{8+δ} along the nodal direction (Fig. 2). The installation of focusing lens in air is also in progress and the calculated spatial resolution is less than 3 μ m. The current apparatus of the laser ARPES system and some ARPES results will be demonstrated.





FIGURE 1. Angle-integrated photoemission spectra from evaporated gold taken with 6.36 eV (195 nm) at 6.2 K, where solid line was obtained by curve fitting using Fermi-Dirac function convolved by Gaussian broadenings.

FIGURE 2. ARPES spectrum of optimally doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ taken with 6.36 eV (195 nm) below 10 K along the nodal direction, where white line denotes the momentum distribution curve at the Fermi level.

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