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TIME-RESOLVED EXPERIMENTS AT NSLS II: MOTIVATION AND MACHINE CAPABILITIES*

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Abstract

NSLS-II is a 3-GeV third-generation synchrotron light source at Brookhaven National Lab. The storage ring has been in routine operations for over six years and hosts 28 operating beamlines. The storage ring performance has continuously improved, including 500-mA with limited insertion devices closed, and routine 400-mA top off operation with 90% uniform filling pattern. Recently, we are exploring different operation modes, uniform multi single-bunch mode, and camshaft mode with a high single-bunch charge, to support timing-resolved user experiments. In this paper, we explore the potential for scientific experiments using the pulsed nature of the NSLS, summarize the user requirements on the beam parameters and the progress of accelerator studies.

NATIONAL SYNCHROTRON LIGHT SOURCE II STATUS

The National Synchrotron Light Source II (NSLS-II) is a 3 GeV, ultra-small emittance (H: 1 nm-rad and V: 8 pm-rad), high brightness third generation light source at Brookhaven National Laboratory. It is to deliver a broad range of light, from IR to hard X-ray, with the brightness of 10^{22} photons/s/mm^2/mrad^2/0.1%BW to 60-70 beam lines at full built-out.

The storage ring was commissioned in 2014 and began its routine operations in the December of the same year [1, 2]. Figure 1 shows the trend of beam current increment for operation and machine study, operation reliability and ID beamline sources commissioning. Over the past years, beam current and operation IDs sources have been steadily increased, while reach and maintain beam in high reliability, >95% [3-6]. After three RF cavities in operation, we have machine runs at 400 mA (project goal 500 mA) with 2.5 minutes periodic top off injection to maintain SR current stability within ±0.5%. Meanwhile we have demonstrated 500 mA in 2019 and performed several radiation survey insertion device and front end to prepare for high current operation since then. Now, there are 28 beamlines in top off operation at 400 mA, with more new beamlines under construction. We support ~5000 hrs operation time per years and keep the reliability above 95%.

With more and more operation beamlines running in mature state, a discussion on possibilities for timing-resolved experiments at the NSLS-II rings has been initiated among accelerator and beamline scientists. A taskforce team were formed since Mar. 2020 to identify science applications that will require the use of SR pulse structures at NSLS-II, define a set of operation modes at NSLS-II to server both high-intensity and timing experiments and specify the instrumentation needs in order to enable these special experiments for accelerator to deliver such timing mode, availability of timing signals at beamlines, tuneable delay lines etc.

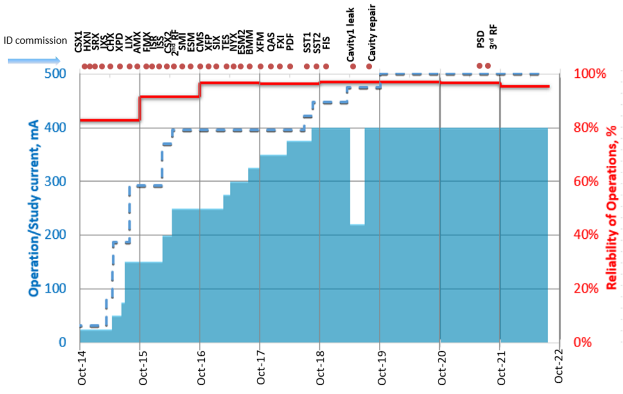

Figure 1: NSLS-II operation trend.

So far, seven beamlines have explored the potential timing-based experiments, including 1) full field imaging of structure in CDI beamline, 2) timing resolved Infra-Red science of materials in FISMET beamline, 3) timing resolved magnetization dynamics imaging in ESM beamline, 4) timing resolved pump-probe STXM to study the linear and nonlinear, magnetic excitations in magnetic micro- and nano-structures in SXN beamline, 5) timing resolved x-ray spectroscopy in ISS beamline, 6) timing resolved experiment for collective dynamics in CSX beamline and 7) high energy x-ray scattering in PDF beamline.

SCIENTIFIC CASES FOR TIMING MODES AT NNSLS-II

Time-resolved studies probe the dynamical properties of systems after being excited away from equilibrium. Exposure to light is among the most common methods for driving a system out of equilibrium—with the photon energy (wavelength) determining the nature of the initial excitation—and serves as an illustrative example of the more general "pump-probe" methodology.

Using a pulsed source for both the excitation—the "pump"--and interrogation—the "probe"--allows the study of fast processes. The light from a synchrotron storage ring is intrinsically pulsed due to the bunching of the stored electrons. We explore the potential for scientific

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experiments focused on revealing dynamical behavior of systems on picosecond to nanosecond time scales using the pulsed nature of the NSLS II light source. During normal operations of NSLS-II, the nominal RMS bunch length is in the 4 to 5 mm range, producing ~15 ps RMS duration light pulses. This value serves as a basic limit on the available time resolution. The wide spectral range of synchrotron light at NSLS-II--from < 1 meV to > 10 keV--is truly unique and enables the probing of a diversity of phenomena associated with structure and electronic configuration.

Here we show how existing NSLS II beamlines that utilize a wide variety of techniques can contribute into a wide range of scientific areas, including physics, chemistry, materials science, and biology. We consider some dynamical behaviors of materials and separate out them into collective electronic phenomena, structural dynamics, and chemical interactions.

Dynamics in Quantum Materials, Condensed Matter, and Electronic Device

Collective electronic properties are at the core of quantum materials as well as the operation of novel electronic devices. Such collective properties stem from the interactions that lead to the formation of Cooper pairs in superconductors, charge density waves, polarons and the organization of electron spins to form skyrmions or magnons. New quantum phenomena can emerge from the competition of other phases. Adding energy to the electronic system using a pulsed perturbation, like a laser pulse for example, creates a non-equilibrium condition that can favor one phase over another, with the relaxation dynamics offering a guide to how the energy is redistributed and the equilibrium condition re-established. Photodoping, where a laser pulse excites electrons from a valence band to a conduction band, is another example where variations in the free carrier density affect physical properties and the time-dependent relaxation can be used to understand the role of electronic interactions. In other cases, quantum confinement in nanostructures can lead to novel electronic behaviors of relevance for photovoltaics or non-linear optical devices. Here, the time-dependent properties of excitons and polarons can affect performance. It should be noted that many quantum phenomena occur at low energies for which relaxation processes can be many picoseconds or longer due to the limited avenues for the energy to be removed. This is particularly true for energies below the optic phonons. Given the advanced capabilities that NSLS-II offers in the infrared to soft x-ray energy range, the synchrotron is uniquely positioned to solve the problems in this area.

Structural Dynamics

Properties of stimuli responsive materials are often governed by complex dynamical structural changes that span multiple time and length-scales. For example, charge transfer during reactions in photoelectrocatalysis involving metal centers can strongly affect the local bonding, while also changing the charge distributions and dipole moments of the constituent systems which further result in rearrangement of the solvent molecules. On the other hand, macroscopic properties of materials are governed by the strain and deformation patterns present on the nanometer scale. Visualizing the changes in the structure across multiple lengths spanning sub-nanometer-to-nanometer scales is an important tool to map out structure-function relationships in functional materials and can be achieved using advanced scattering methods available at NSLS II.

Chemical Dynamics, Reactions, Interfaces

Chemical dynamics play a central role in the design and performance of novel materials for catalytic and energy applications. Transition metal complexes often serve as building blocks of such systems due to the rich nature of chemical interactions between metals and organic ligands allowing fine tuning of their chemical properties. Understanding the element-specific dynamics in such systems can help in development of mechanistic models of excited state reactivity and can be achieved by hard x-ray spectroscopic experiments.

REQUIREMENTS OF BEAM PARAMETERS

Considering both high-flux and timing users simultaneously, we select the filling modes of the ring in two main categories, symmetric and asymmetric.

For symmetric filling pattern, multi single bunch are evenly distributed in machine buckets with the same time gap and the same amount of charge, so that each bunch could be used for time experiments. This pattern presents advantages in terms of reproducibility and timing. The ring's harmonic number of 1320 (=2x2x2x3x5x11) determines the feasible symmetric fill patterns, thus can provide any combination of these prime factors. For example, we can deliver one bunch every 6 buckets, corresponding to 220 bunches, each separated by 12 ns. Some beamlines select 41.67 MHz Amplitude Systemes fiber laser for NSLS-II timing activities. The limitation of this mode, comparing with normal operation, is the average beam characteristics, such as lower total beam current, shorter beam lifetime, etc. due to vacuum heating and beam stability limitation

Another filling pattern to support timing experiments can be implemented by dedicating a subset of the bunches to produce the “probing” light pulse. This filling scheme is called a hybrid filling pattern and often has one high-current bunch, “camshaft” bunch, located in the middle of the ion-clearing bunches, or multiple camshaft bunches with repetition rate matching to the rate of beamline “pumping”, e.g., laser pulses. The maximum current of the camshaft bunch(es) is(are) determined by single-bunch instability and beam lifetime. This filling mode represents an appealing compromise between normal filling conditions for general user experiments, where a high total beam current and a long lifetime are desired, and timing experiments, where a short, bright probing pulse is desired. In addition, this mode can separate the camshaft bunch from the rest of the fill with time gaps of 100s of ns, easing the time-resolution requirements on experimental system, such as the detector, or allowing a longer relaxation time for the sample.

In Table 1, we list the beam parameters for the proposed timing mode experiments' filling pattern along with standard operation mode, including the beam current, filling pattern, single bunch peak current, bunch length, bunch spacing etc.

ACCELERATOR PERFORMANCE

To reach high single bunch current, we performed accelerator beam studies to set accelerator with reasonable injection efficiency and beam lifetime. Besides bunch-bunch feedback (BBF) optimization, we also generated lattice with high linear chromaticity to increase internal damping. With three family of chromatic sextupoles, we increased the chromaticity from +2 to +7. We also use six families of harmonic sextupoles for the nonlinear lattice optimization. Overall, the beam current of 11.3 mA was accumulated in the single-bunch mode with very short beam lifetime. Further current increase is limited by vacuum activities.

We characterized bunch length dependence [7] on bunch current and ID settings. Figure 2 shows the bunch lengthening measured at NSLS-II bare lattice and the operational lattice with all IDs closed. RF voltage also affects the bunch length.

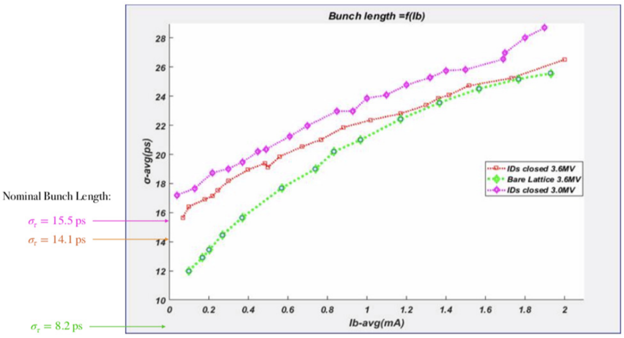

Figure 2: Measured bunch length as a function of single-bunch current.

We also put more efforts to develop filling pattern monitor to accommodate large range bunch charge in hybrid mode and bunch cleaning system to reach 10^{-5} bunch purity. Furthermore, we explored the possible scheme of timing system from master oscillator to individual beamlines for signal synchronization.

CONCLUSION

After years of operation, NSLS-II accelerator and beamlines are running in mature state. We started to explore timing-resolved mode to support experiments.

Time-resolved studies are a natural milestone and run in many storage ring user facilities. Source brightness and reliability development, along with high-efficiency x-ray optics, enable x-ray to probe samples at picosecond time scales, which is sufficient for a wide variety of high-impact measurements. At NSLS-II, we anticipate that advances in condensed matter physics, materials science, chemistry, and biology would follow our adoption of new storage ring fill patterns. This new capability would expand the studies already underway to better time resolution and create opportunities for the creation of new user communities.

We explored the possible science applications in beamlines, define the candidates of achievable timing filling pattern, conducted accelerator beam studies to evaluate machine performance, and specify the instrumentations in order to enable these special experiments.

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Table 1: NSLS-II Proposed Operation Mode Beam Parameters

Operation Mode	Standard	Diffraction limit	Hybrid mode		220/N* bunches
Beam Intensity		Moderate			
I [mA]	400/500	400/500	300 + 5 mA		~200
Filling pattern	90% (80%)	90% (80%)	Bunch trains (80%) + singlets (1)		Multi single bunch
I/bunch [mA]	0.33	0.33	0.2	3-5 mA	~1 mA (N=1)
Bunch gap [ns]	2	2	2	280	12*N
Emittance [nm/pm]	0.9/30	0.9/8	0.9/30	0.9/30	0.9/30
Lifetime [hrs]	8	3.5	~16 hrs	1 hr	5 hr
Bunch length (ps)	19	19	18	~40	~25

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