NSLS-II meV-IXS Program Overview

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The meV-IXS program at NSLS-II is served by beamline 10-ID dedicated for studies of atomic dynamics in materials systems of both applied and fundamental interest. The key instrument is a novel spectrometer with analyzer optics based on the combination of post-sample collimation and highly dispersive flat crystal optics that produces ~meV energy resolution at a medium energy of 9.13 keV with sharp tails and high momentum resolution. It offers unique strengths on soft materials with mesoscopic heterogeneity and complexity. Furthermore, the medium operation energy intrinsically provides enhanced surface sensitivity that is most suitable for systems of reduced dimensionalities (e.g., phonons on surfaces of single crystals, thin films, confined liquids, and HP DAC samples). In this overview, the current performance of the spectrometer will be presented and plan for future upgrades briefly discussed.

Implementing Terahertz Phonon Manipulation: What Can We Learn From IXS Measurements?

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In recent years, an increasing number of IXS investigations have been dealing with complex mesoscale structures, holding, in some cases, the promise of new functionalities. To outline some emerging trends in this field of research, I will discuss published or unpublished Inelastic X-Ray Scattering (IXS) investigations of the terahertz collective dynamics of inhomogeneous hybrid (liquid-solid) systems. These results indicate that the IXS spectrum has a non-trivial shape in which hydrodynamic acoustic-like modes are either suppressed or sided by additional, non-hydrodynamic ones. Most importantly, in the long run, the observed connection between this spectral behavior and the mesoscale arrangement might inspire new methods to steer acoustic propagation through the design of the nanoscale structure. Finally, I will highlight the critical role played by the Bayesian analysis in this emerging field of research, also illustrating some future directions.

Molecular picture of the transient nature of mesogenic domains

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The investigation of phonon excitations in soft and biological materials at the molecular scale has always been challenging due to limitations of experimental techniques in resolving meV vibrational modes. The understanding of collective mode behavior with a *Q*-dependence is the key to implement heat management based on the control of a sample structure. The latter has great potential for many energy-inspired innovations. As a first step toward this goal, we carried out high contrast IXS measurements on a liquid crystal (LC) sample, D7AOB, which exhibits solid-like dynamic features, such as the coexistence of longitudinal and transverse phonon modes. For the first time, we found that these terahertz phonon excitations persist in the crystal, smectic A, and isotropic phases. Furthermore, I will present data on the intermediate smectic A phase, which was shown to support a van der Waals-mediated nonhydrodynamic mode with an optical-like phonon behavior. Finally, I will show the coexistence of transverse acoustic and optical modes of 5CB LCs at near room temperature which was revealed through the emergent transverse phonon gap and THz light-phonon coupling taking place within the same energy range. The tunability of the collective excitations at nanometer–terahertz scales via selection of the sample mesogenic phase represents a new opportunity to manipulate optomechanical properties of soft metamaterials.

What's so special about cholesterol? Integrating simulations and IXS to identify collective modes in lipid membranes

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The membranes that enclose our cells are made of amphiphiles called lipids, which assemble into a bilayer structure to form a semipermeable barrier between the extracellular and intracellular spaces. The structural parameters of lipid bilayers — the thickness, area per molecule, the orientation of the director — have long been studied by static scattering of x-rays and neutrons, in order to determine how continuum elastic properties emerge from molecular level interactions. Now, recent advances in sources and detection have opened a new meV-IXS window onto membrane structure and dynamics. When integrated with molecular dynamics simulations, meV-IXS data reveal psec to nsec collective dynamics of membranes that are inaccessible by any other technique. A series of simulations and experiments on membranes containing varying amounts of cholesterol show how the dynamic response of membranes depends on this very special lipid, which makes up roughly 30% by mole of our membranes, and which simultaneously fluidizes and orders lipid bilayers.

Ultra-fast collective dynamics in biological membranes

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Biological membranes exhibit a great deal of compositional and phase heterogeneity due to hundreds of chemically distinct components that form them. Their phase behavior is enormously complex and, as a result, structural and dynamic processes in cell membranes are extremely difficult to study, especially at the molecular level. In the present work, we argue that high resolution inelastic X-ray scattering (IXS) can provide valuable insight into biophysics of lipid membranes. In the series of IXS experiments [1-3] on single, binary, and ternary lipid mixtures, we obtain experimental evidence of propagating in-plane transverse acoustic (TA) and optical (OP) phononic modes in lipid systems. These phonon modes exhibit low-Q/low-E phononic gaps (the absence of phonons within a particular Q- or E-range). The energy and the Q-cutoff of the phononic gaps depends on the temperature and compositional variation in those systems. We show that phononic gaps behavior is directly related to such processes as passive transport, lateral lipid diffusion, and formation of phase separated domains and allows for quantitative study of these fundamental properties of a cell membrane.

[1] M. Zhernenkov, D. Bolmatov, D. Soloviov, K. Zhernenkov, B. P. Toperverg, A. Cunsolo, A. Bosak, Y. Q. Cai, *Nat. Commun*. **7**, 11575 (2016)

[2] D. Bolmatov, D. Soloviov, M. Zhernenkov, D. Zav'yalov, E. Mamontov, A. Suvorov, Y. Q. Cai, J. Katsaras, Langmuir **36**, 4887–4896 (2020)

[3] D. Soloviov, Y. Cai, D. Bolmatov, A. Suvorov, K. Zhernenkov, D. Zav'yalov, A. Bosak, H. Uchiyama, and M. Zhernenkov**,** *Proc. Natl. Acad. Sci. U.S.A.* **117**, 4749-4757 (2020).

Exploring topological phonons in CdTe

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In condensed matter physics, the age of topology has led to the discovery of Weyl and Dirac fermions in electronic band structures with great potential for new types of electronic transport. In recent years, efforts have been made to extend the topology to bosonic systems in order to realize new types of vibrational/heat transport. Inelastic X-ray scattering (IXS) with meV resolution is a very powerful tool to study the topological phonons in condensed materials. Here, we report IXS measurements on CdTe single crystal. By mapping out the bulk phonon dispersions of CdTe, we show the observation of type-II Weyl points near the Brillouin zone boundaries. Moreover, topological phonons can lead to novel surface states, resembling those of the Weyl fermions such as Fermi arcs. Complimented with theoretical calculations, we try to explore the predicted surface state making use of IXS under grazing incidence conditions.

Segmented Adaptive Gap Undulator for Inelastic X-ray Scattering Beamlines

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Segmented Adaptive Gap Undulator (SAGU), in which different segments have different gaps and periods (see fig. 1), promises a considerable spectral performance gain over a conventional undulator with constant gap and period over its length. According to calculations (see fig. 2), this gain can be comparable to the gain due to the use of a superior undulator technology (e.g. a room-temperature in-vacuum hybrid SAGU would perform as a cryo-cooled hybrid in-vacuum undulator with uniform gap and period). SAGUs are particularly attractive for IXS beamlines, that typically don't require frequent changes of photon energy / undulator gaps in segments. Besides the calculation results, progress on construction of an SAGU prototype at NSLS-II will be reported.

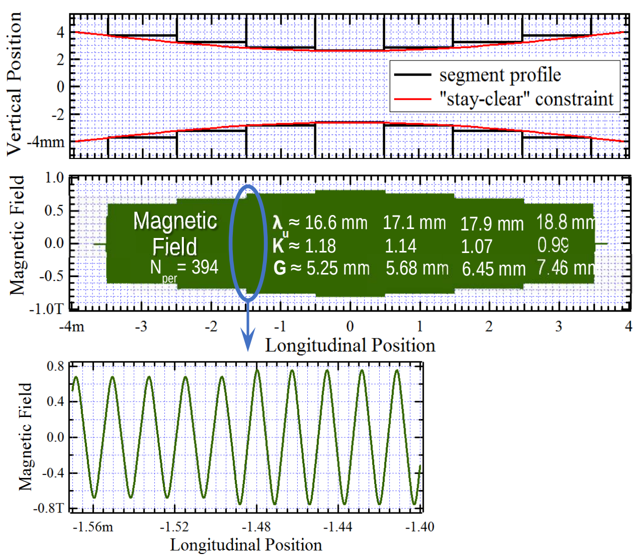

Figure 1: Electron beam vertical "stay-clear" constraint and the corresponding SAGU structure (upper and middle graphs), and the magnetic field in the region of a segment junction (bottom graph). Tentative parameters of a room-temperature hybrid permanent-magnet SAGU for IXS beamline are shown in the middle graph.

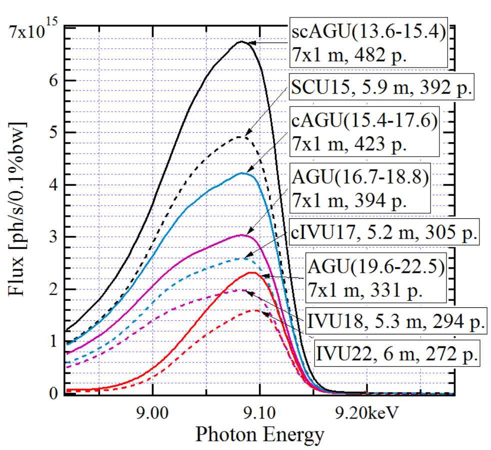

Figure 2: Calculated undulator radiation spectra in the vicinity of 9.1 keV photon energy, for constant-period undulators of different technologies (dashed curves): room-temperature hybrid (red and magenta), cryo-cooled (blue), and superconducting (black), optimized for producing maximal flux at that photon energy, and their corresponding SAGU versions (solid curves).

Performance evaluation of the IXS analyzer at the NSLS-II 10-ID beamline.

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NSLS-II meV-IXS spectrometer has a unique design aimed at studying dynamics in soft and hard matter with medium energy x-rays [1]. Its analyzer scheme is based on the combination of a post-sample collimating mirror and highly asymmetric crystal optics to deliver ~meV resolution with high spectral contrast for inelastic x-ray scattering. As a pioneering facility in the construction and operation of such a unique meV-IXS spectrometer, we have gained exceptional experience and detailed knowledge in optimizing and operating our spectrometer. In my presentation, I will demonstrate a detailed analysis of the performance of the analyzer optics based on theoretical computation and comparison with experimental observations. I will also provide the basis for potential future improvements of the analyzer.

[1] Y.Q. Cai, D.S. Coburn, A. Cunsolo, J.W. Keister, M.G. Honnicke, X.R. Huang, C.N. Kodituwakku, Y. Stetsko, A. Suvorov, N. Hiraoka, K.D. Tsuei, and H.C. Wille, "The ultrahigh resolution IXS beamline of NSLS-II: Recent advances and scientific opportunities", Journal of Physics: Conference Series 425, 202001 (2013)