
Kobe City Reservoir
Above Shin-Kobe Station

Perspective and Prospective from Momentum Resolved Studies of Atomic Dynamics

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Presented the EBS NRS Workshop
ESRF, Grenoble, France
11 March 2019

Who am I?

PhD doing NRS at Stanford University - data from 2nd generation sources
(everything after the HHL mono was the user's responsibility)

Rueffer's Group at ESRF from 1995 – 1997
New era in NRS studies (& SR): AC inelastic, AB coherent,

1998: Moved to Japan, SPring-8/JASRI, agreed to build a beamline for IXS
BL35XU, 4.5 m ID.   BL Comparable with others, but still too weak.

2004: Began to consider using 30m straight section for IXS (main issue: power load)

2006: Moved to RIKEN, began push to get money for new BL in 30m straight

2009: First funding came, began detailed design of BL43LXU
3 x 5m IDs, tunable from 14.4 to 25.7 keV in the fundamental

2012: Light from a single ID
2015: All IDs installed and BL mostly working:
Now: 0.8, 1.3, 2.8 and 25 meV res. for momentum resolved spectroscopy,
~3 to 80 GHz on the sample per meV resolution

Interests: Methods/information content, Implementation, X-Ray Instrumentation, Materials Science

In depth (and up-to-date) review of IXS: Springer Handbook & arXiv 1504.01098

The EBS Upgrade

Greatly Improved Brilliance (ph/s/meV/source size/source divergence)
But mostly the same flux (ph/s/meV)

The improvement is essentially all in the horizontal source properties
The horizontal at EBS will be similar to the vertical at oESRF (o=old)
Nearly a "round" x-ray beam

A win for experiments requiring:
- very small beams (smaller H size possible, & with less divergence)
- very parallel beams (possibly interesting for extreme optics and NSAXS)
- high transverse coherence or large coherent fraction.

Also a win for power on the monochromator (reduced by a factor of 1.5 to 2)

Meanwhile, any experiment that was well matched to oESRF operating conditions will remain mostly the same, except horizontal x-ray spot may shrink.

Strong base in present suite of experimental techniques.

The purpose of this workshop is to look for new ones.

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Coherence?

An "obvious" direction to go (in the context of other plans for EBS) is to make use of the improved coherence / coherent fraction for imaging.

Ex: Coherent Diffractive Imaging (CDI), Ptychography
~ 10 nm scale resolution (or better)

Nuclear Forward Scattering (NFS) in the Time Domain
Gives access to chemical and magnetic structure via hyperfine interactions

However, NFS imaging is not really possible since
a 2D time-resolving detector is needed, ideally with small pixel size

Note: 1D is possible so specific applications may be considered

-> Complementary method needed

Most Obvious: Scanning Microscopy (Tomography?)
Expect ~ 0.2 um beam size, eventually 0.1 um, maybe better
Hyperfine Structure (NFS or SMS) and even local phonon PPDOS

SMS = Synchrotron Mossbauer Source (approximately absorption spectroscopy)
Freq. Domain/Velocity scan w/Borate near Neel Point – Simpler but slower

Nuclear SAXS

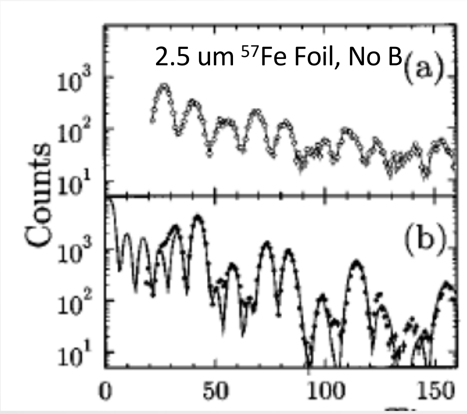
Instead of imaging, one can consider a SAXS geometry which gives related information: correlation lengths for hyperfine parameters

Here the EBS gives
1. Nearly round beams: similar divergence H&V
2. The potential to collimate more (~urad) and keep the beams small.

Possible Setup

HRM, Crossed Channel Cuts, Sample, Crossed Channel-Cuts

Also area detector could be considered (if existed) or 1D detector



Shvyd'ko et al PRB 1996

Expect: Correlation lengths ~um and smaller acessible

Caveat: Theory for NFS from thick (enriched) samples does not yet exist.

AB opinion: Maybe not so bad. Start kinematic and modify.
(Unmagnetized foil is perhaps an especially hard case.)

Crossed Polarizers

ELSEVIER

Nuclear Instruments and Methods in Physics Research B 103 (1995) 371–375

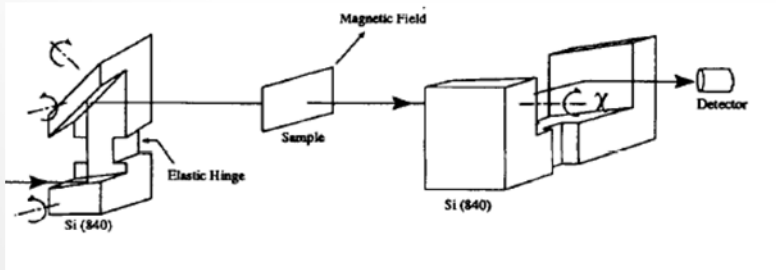
NIM B
Beam Interactions with Materials & Atoms

Mössbauer spectroscopy using synchrotron radiation: overcoming detector limitations

D.P. Siddons [a,*], J.B. Hastings [a], U. Bergmann [b], F. Sette [b], M. Krisch [b]

[a] National Synchrotron Light Source, Brookhaven National Laboratory, Upton, NY 11973, USA
[b] European Synchrotron Radiation Facility, Avenue des Martyrs, 38043 Grenoble, France

Received 16 December 1994; revised form received 9 June 1995



Selects only scattering which rotates the polarization using a 90 degree Bragg reflection.

EBS would allow significantly increased efficiency as the crossed polarizers require good (urad) collimation in both directions

Limited to optically active materials (e.g. with magnetic splitting)

What about the 50 μeV?

Much better than what presently can be done using IXS, but not so different than what is available using INS (though NIS DOS is superior to INS DOS)

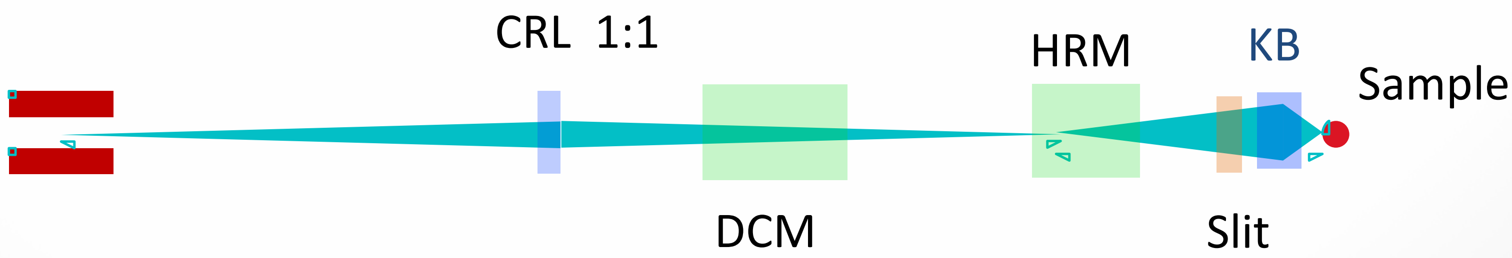
Experience with momentum resolved scattering-> for many cases of interest, good energy resolution must be matched by good Q resolution, which is not possible with NIS.

Localized modes? Molecular modes? Diffusion?
Low energy contributions to DOS

Optical Layout

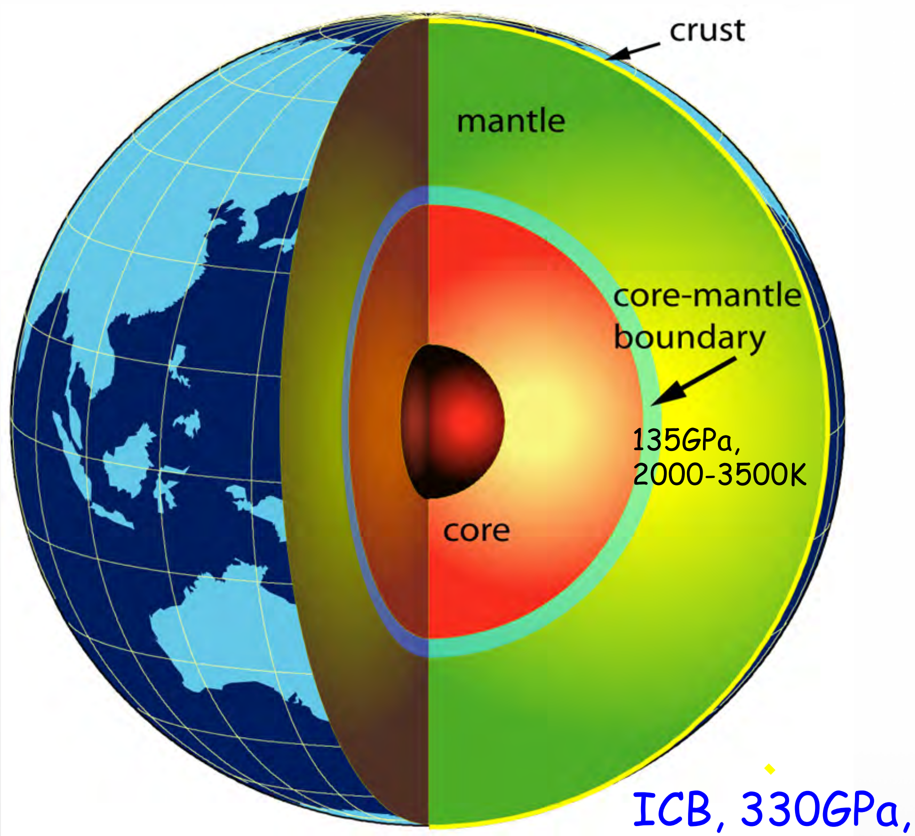
NSLS-II IXS Beamline

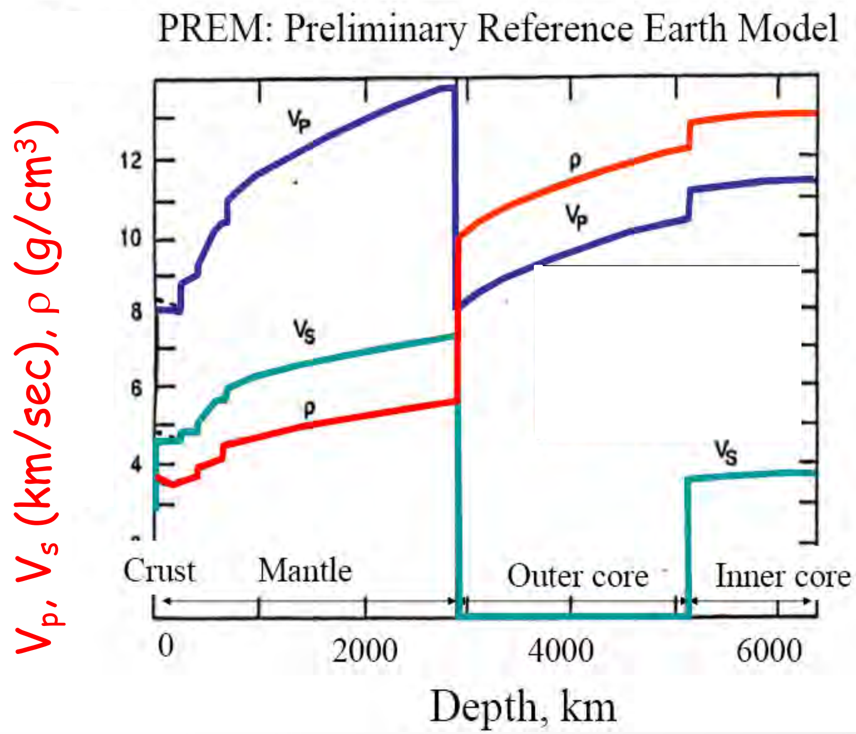
How to get a small spot with a ***dispersive*** high-resolution monochromator…?
…put the HRM at a virtual source point.



+ Small spot possible at sample
+ Small beam size at HRM -> smaller crystals
- High power density on 1st HRM crystal (Reduce incident BW?)
- Probably some residual impact of HRM refraction over extended length

Geoscience





ICB, 330GPa,
4000+ K

Earth's Center, 365 GPa
6000+ K

Velocity distribution (and ρ) well know from seismic measurements
Needed: Lab measurements relating T, Density & Composition to V

A catalogue of sound velocities **in extreme conditions...**
& More General Material Properties

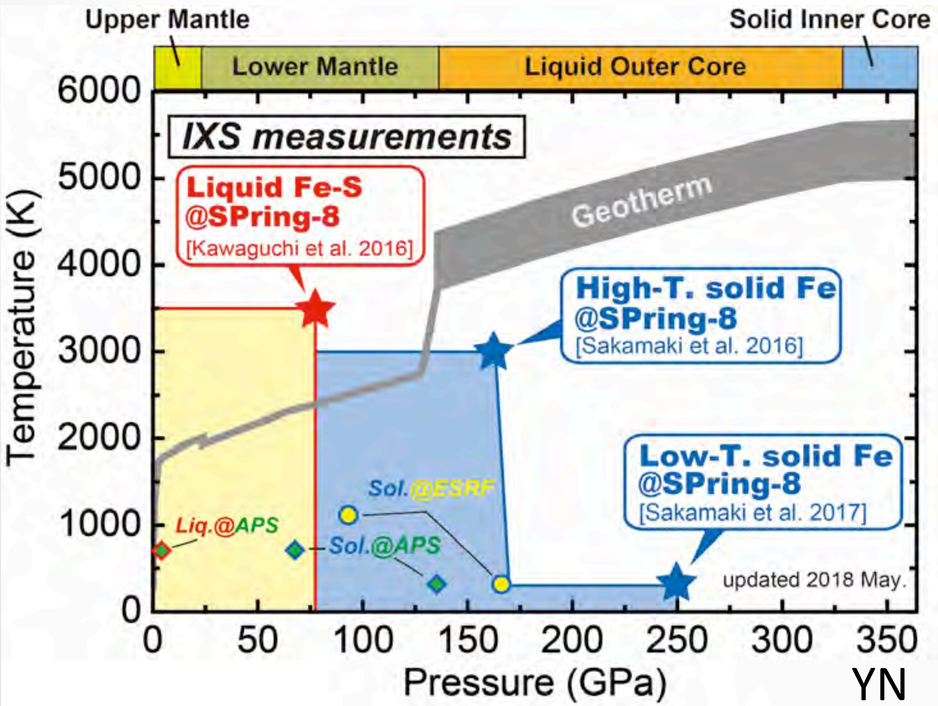
IXS & NIS

Complementary Techniques:

IXS gives direct access to acoustic mode dispersion (mostly LA, sometimes TA) in solids and liquids

NIS Gives access to the Debye velocity (some combination of LA and TA) in iron-containing solids

Each method has issues/complications – better to have both.



IXS at SPring-8 over the last 15 Years
Mostly at low-Q (powders&liquids)

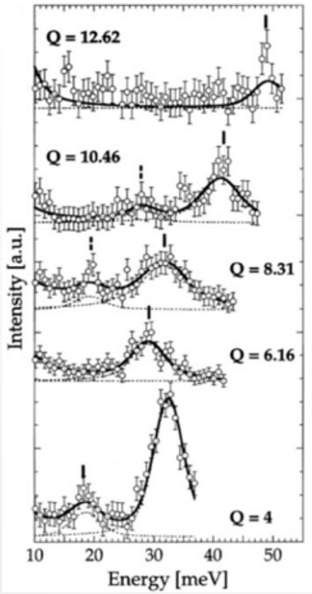
Fukui, Nakajima
(MDL -> External User)

Ohtani-lab (inc. Sakamaki)
Hirose-lab

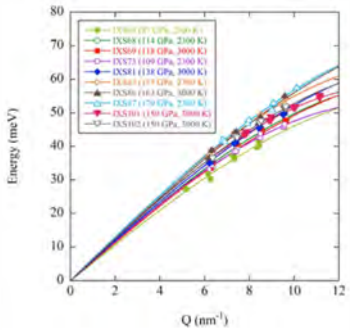
Best for nearly isotropic materials

Sound Velocity by IXS

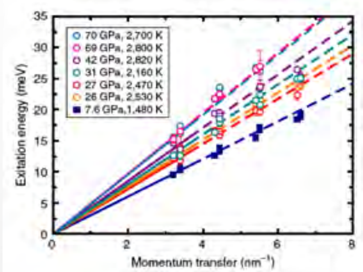
V_p from Q->0 limit of acoustic dispersion
Identify Fe acoustic mode, plot E vs Q, get limit…



Solid Fe, 28 GPa, RT
Fiquet, Science 2001



Solid Fe, HP,T
Sakamaki, et al



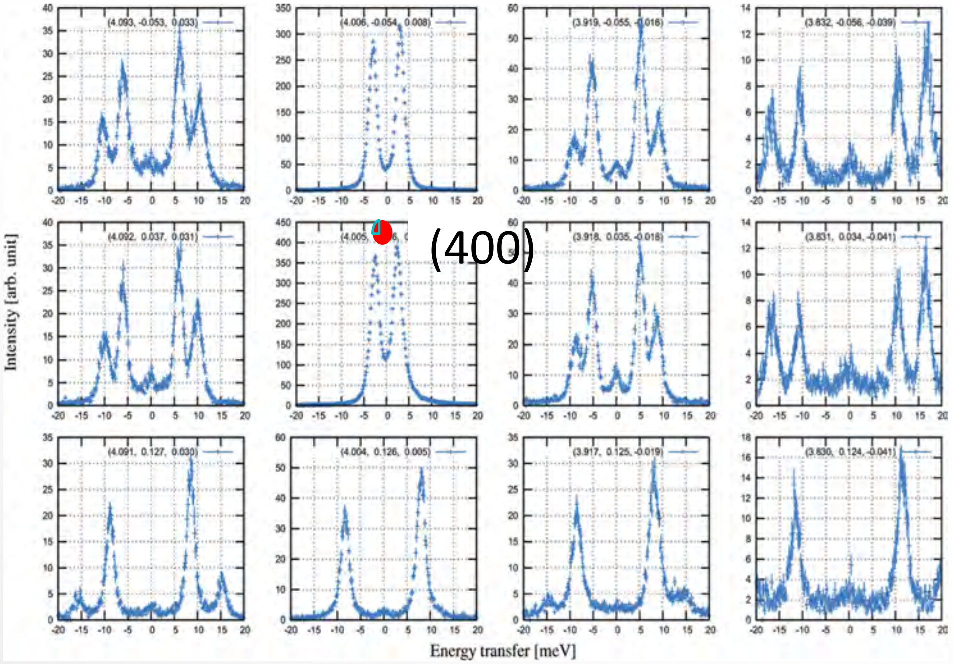
Liquid FeC, HP,HT
Nakajima, et al

Many Caveats:
- Non-Linearity
- Preferred Orientation
- Temperature Uniformity
- Fast Sound (liquid)

Not discussed here…
Years & many pages
of discussion with referees.

Single Crystal Technique

Fukui et al, JSR 2008



MgO About (400) Bragg Point using 12-Analyzer Array
Easily track LA,TA dispersion
Fit at arbitrary Q's using Christoffel's eqn.

Starting to do this under pressure.

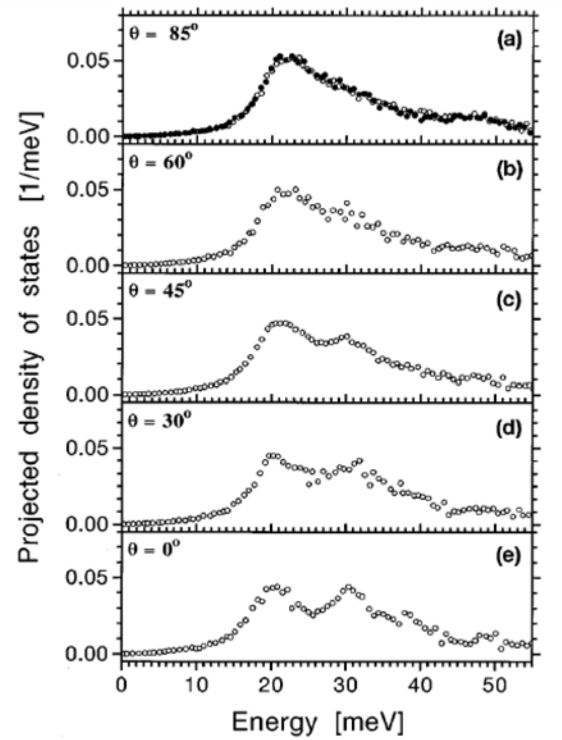
Interest in anisotropic materials
(e.g. post-pervoiskites)

As may be associated
with regions of anomalous sound
velocity as caused by preferred
orientation due to
pressure/growth

NIS: DOS -> Really PPDOS

Carefully Speaking
NIS gives the PPDOS =

Partial (Isotope only)
Projected (phonon polarization in beam direction)
Density of States



Iron Borate
Chumakov et al, prb 1997
Kohn et al, prb 1998
See $\mathbf{k} \cdot \mathbf{e}$

An opportunity with anisotropic materials:

Use a small (<~1 um beam) to measure small crystals or pick out single grains.
Investigate dependence of the DOS on the relative orientation of the beam and the crystal axes.

EBS helps get down to the <~um grain size

NIS not very angle sensitive so strain under pressure may be less of an issue

Vortex Beams:
X-Rays with Orbital Angular Momentum

Beams that (in cylindrical cords) have an azimuthal phase dependence scaling as

$e^{il\varphi}$ (l an integer)

Implying a singularity/vortex on axis.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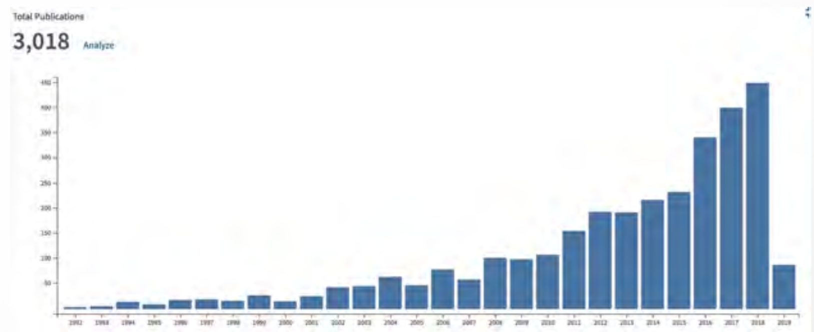
Orbital angular momentum of light and the transformation of Laguerre-Gaussian laser modes

L. Allen, M. W. Beijersbergen, R. J. C. Spreeuw, and J. P. Woerdman
Phys. Rev. A **45**, 8185 – Published 1 June 1992

Article References Citing Articles (2,133) PDF Export Citation

ABSTRACT

Laser light with a Laguerre-Gaussian amplitude distribution is found to have a well-defined orbital angular momentum. An astigmatic optical system may be used to transform a high-order Laguerre-Gaussian mode into a high-order Hermite-Gaussian mode reversibly. An experiment is proposed to measure the mechanical torque induced by the transfer of orbital angular momentum associated with such a transformation.



Light that can couple to molecules angular momentum and induce torque
e.g. Babiker et al, prl 1994, Picon et al, njp 2010, Ribic et al, prl 2014

Also possible strong dichroism (VanVeenendaal & Mcnulty, prl 2007)

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What does it look like?

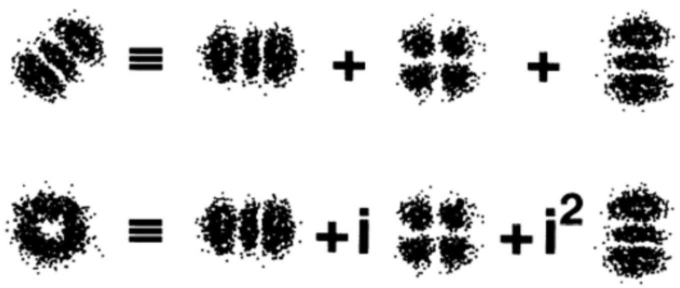


FIG. 3. The decomposition of a TEM_{02} Hermite-Gaussian mode at 45° into a set of Hermite-Gaussian modes and the decomposition of a Laguerre-Gaussian mode into the same, re-phased, set.

Allen et al, pra 1992

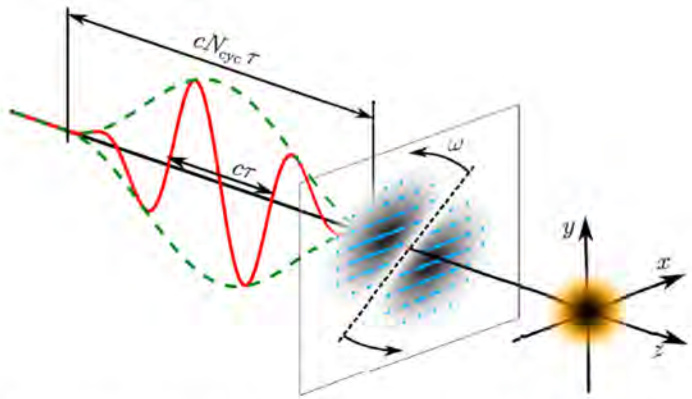


Figure 1. Sketch of the physical system consisting of a light pulse interacting with a hydrogen atom at the origin, where the time pulse dependence and its associated initial parameters are introduced. As an example, we plot the transverse profile of the vector potential (4) for a linearly polarized LG mode with topological charge $\ell = 1$.

Picon et al, njp 2010

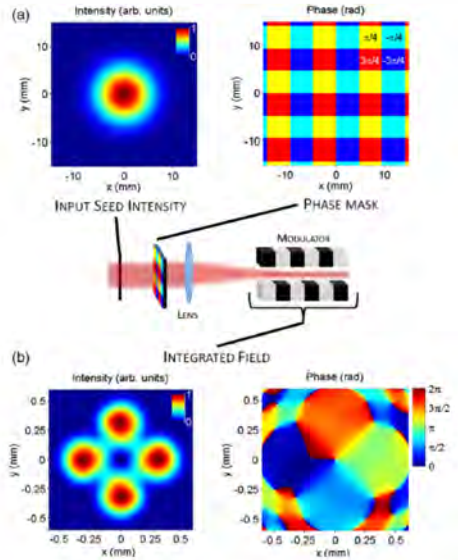
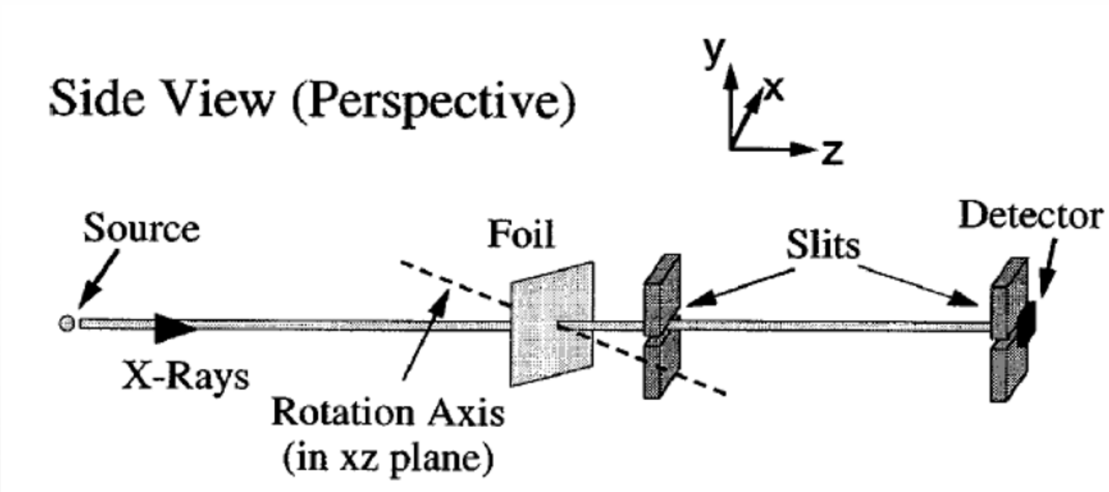


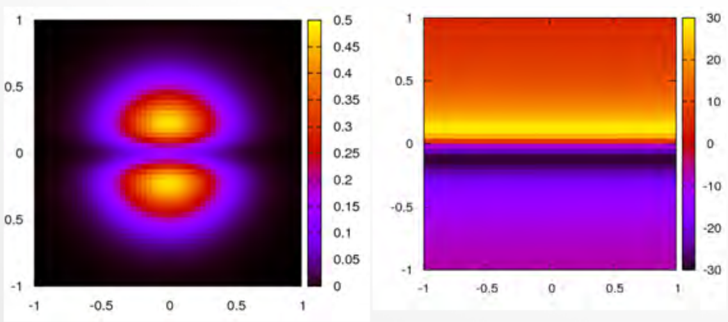
FIG. 2 (color online). (a) The optical phase mask (right) used to modulate the transverse intensity profile of an initially Gaussian seed laser (left). (b) Transverse intensity (left) and phase (right) distributions of the integrated seed laser field along the modulator.

Ribic et al, prl 2014

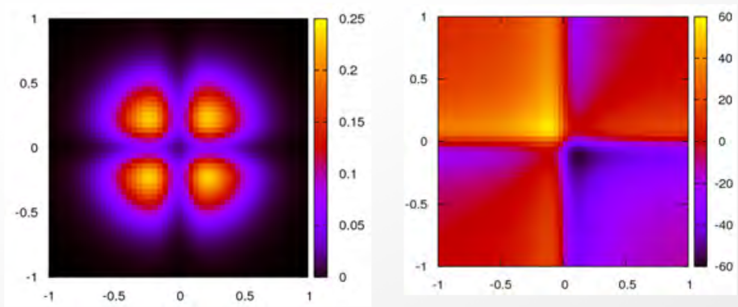
The nuclear analogue has already been made…



Baron et al, prl , 1996



Single Rotating Foil
At exact resonance



Dual (Crossed) Rotating Foils
At exact resonance

So what is it good for…?

Caveat: Plots for E=0, only, Other energies are shifted.
So full time evolution is complicated

Clearly interaction is only useful/interesting over nuclear bandwidth
-> can only couple usefully to other resonant nucleii

Need a case where there is rotation motion of nucleus/molecule without too large a diffusion broadening to keep sufficient interaction cross section – maybe tricky.

Why EBS: Interaction formulas appear to scale with inverse of transverse beam size so a small beam is desirable.

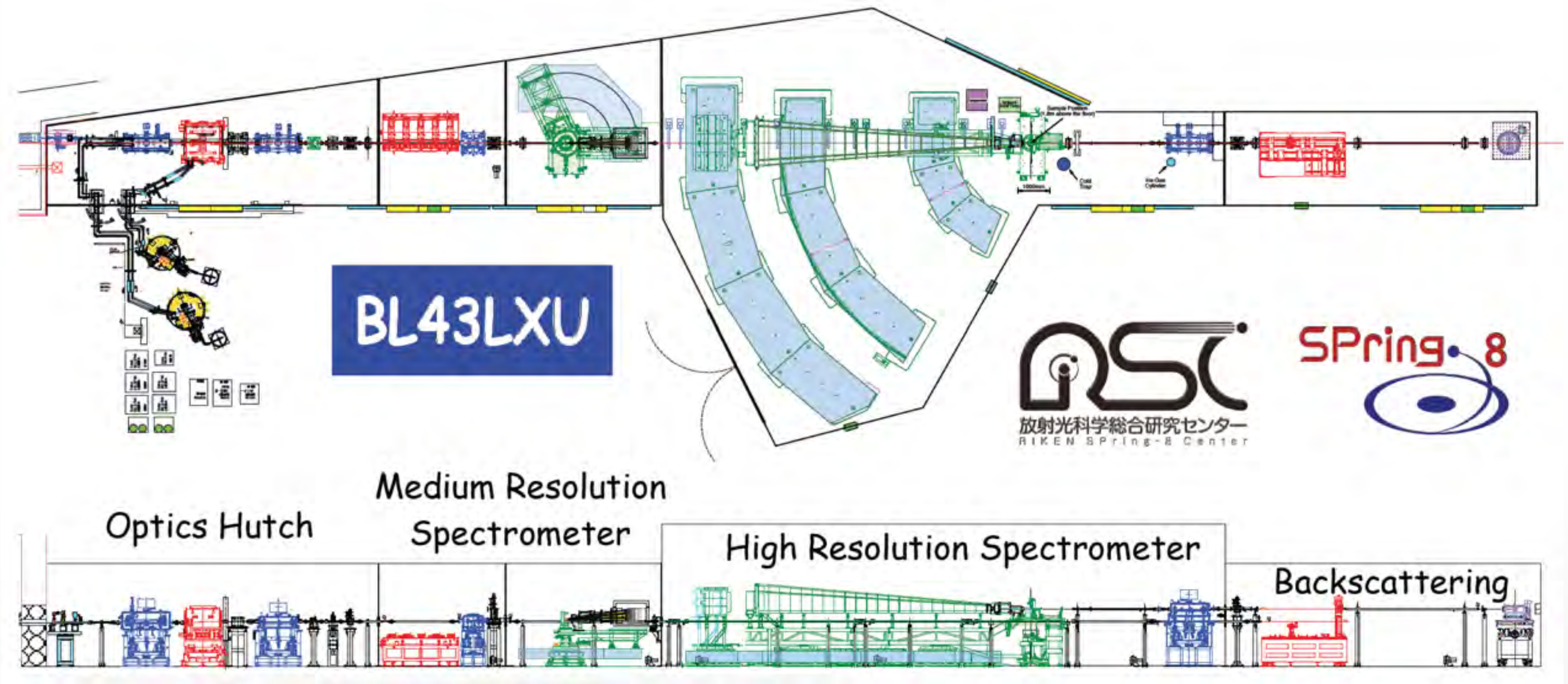
Concluding Comments

Largest potential may be in microscopy (either NFS or SMS)

Some push (still/again) for a time-resolving area detector
(Note: SMS does not require time resolution but is a low-rate expt)

Opportunities for DAC work with micron or sub-micron beams
Projected DOS for anisotropic materials (also NFS/SMS)

Speculation about vortex beams



Thank You!

"... its all about reality and making some noise ..." M. Shinoda