

X-ray Echo Spectroscopy

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Inelastic X-ray Scattering (IXS) requires improvements in:

- Spectral resolution $\Delta \varepsilon$.
- Momentum-transfer resolution ΔQ .
- IXS signal strength.

IXS is at an Impasse over Spectral Resolution



IXS is at an Impasse over Spectral Resolution



IXS is at an Impasse. How to Overcome?



Decoupling the spectral resolution from the bandwidth of the x-rays



Spin Echo

Spin echo is the refocusing in the time domain of the defocused spin magnetization by time reversal.



Soft X-Ray RIXS - Energy-Compensation



Figure 2

Illustration of the energy-compensation principle of grating dispersion applied to a RIXS measurement. Both gratings have an identical central groove density and the same distance to the sample. After passing through the entrance slit, X-rays of varied energy $\hbar\omega \pm \varepsilon$ are dispersed onto the sample and experience inelastic scattering. Scattered X-rays of identical energy loss (Δ_1 or Δ_2) are dispersed and focused to the same point on the CCD.

H. S. Fung et al., AIP Conf.Proc. 705, 655 (2004).

C. H. Lai, ..., D.J. Huang, J. Synchrotron Radiat. 21, 325 (2014)

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- X-ray echo spectroscopy principles.
- X-ray echo spectrometer design & performance.
- Comparison with HERIX.
- Feasibility.
- Conclusions and outlook.

- X-ray echo spectroscopy, is a space-domain analog of the spin echo spectroscopy.
- An image of an x-ray source is defocused by a focusing-dispersing system $\hat{O}_{\rm D}$.
- The defocused image is refocused in a time-reversal focusing-dispersing system $\hat{O}_{\rm R}$ (echo).



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Refocusing (echo) takes place if the linear dispersion $G_{\rm D}$ in $\hat{O}_{\rm D}$ is compensated (time-reversed) by linear dispersion $G_{\rm D}$ in $\hat{O}_{\rm D}$: $G_{\rm D} + G_{\rm R}/A_{\rm R} = 0$



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- If the defocused beam is inelastically scattered from a sample with an energy transfer ε , the echo signal acquires a lateral shift $G_{\rm R}\varepsilon$.

• An inelastic scattering spectrum is measured by mapping ε on the pixel detector.

• Spectral resolution:

 $\Delta arepsilon = rac{\Delta x_2}{G_{
m P}}$

does not rely on the monochromaticity of x-rays!



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X-ray echo spectrometer design & performance



X-ray echo spectrometer design & performance



X-ray Echo Spectrometer - Elastic Scattering



X-ray Echo Spectrometer - Inelastic Scattering



X-ray Echo Spectrometer - Inelastic Scattering



X-ray Echo Spectrometer - Inelastic Scattering



X-ray echo spectrometer design & performance



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X-ray echo spectrometer design & performance



X-ray Echo Spectrometer Tolerances



X-ray echo spectrometer design & performance



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Bragg-Diffracting Crystals as Diffraction Gratings Angular Dispersion

An asymmetrically cut crystal behaves like a diffraction grating dispersing photons with different photon energies: effect of angular dispersion.



Yu. Shvyd'ko X-Ray Optics, Springer-Verlag (2004)

$$\mathcal{D} = \delta \theta' / \delta E$$
 = dispersion rate

$$\mathcal{D} = rac{2\sin\theta\sin\eta}{E\sin(\theta-\eta)}$$

Bragg-Diffracting Crystals as Diffraction Gratings Angular Dispersion

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Angular dispersion rate
$$\mathcal{D}$$
 is biggest for:

$$\mathcal{D} = \delta \theta' / \delta E =$$
 dispersion rate

$$\mathcal{D} = rac{2\sin heta\sin\eta}{E\sin(heta-\eta)} \Longrightarrow rac{2\tan\eta}{E}$$

1. $\theta' - \eta \rightarrow 0^{\circ}$ and $\theta \rightarrow 90^{\circ}$. 2. lower photon energies *E*; E=9 keV is chosen as a compromise

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Yet often insufficient for practical applications



APS, Sept 09, 2016

Multi-Crystal Optic as a Diffraction Grating with Enhanced Angular Dispersion Rate

The angular dispersion rate in a multicrystal optic can be dramatically enhanced by almost two orders of magnitude compared to that of a single crystal:

$$\mathcal{D}_{\cup_n} = b_n \mathcal{D}_{\cup_{n-1}} + \mathcal{D}_n$$

The multi-crystal dispersing element ("diffraction grating") is characterized by a cumulative angular dispersion rate \mathcal{D}_{\cup} , a cumulative asymmetry parameter b_{\cup} , and a spectral window of imaging ΔE_{\cup} .



Multi-Crystal "Diffraction Gratings" for 0.1-meV-Resolution Echo Spectrometers



Dispersing elements D_D (a) and D_R (c) of the defocusing \hat{O}_D and the refocusing system \hat{O}_R , and their spectral transmittance function (b) and (d), respectively. The dispersing elements are examples of an in-line four-crystal CDDW-type optic, comprised of collimating (C), dispersing (D_1, D_2) , and wavelength-selecting (W) crystals ensuring large cumulative dispersion rate \mathcal{D}_U , large bandwidth ΔE_U , and large angular acceptance $\Delta \theta_U$. Different functions of D_D (a) and D_R (c) dictate their different designs.

X-ray echo spectrometer design & performance



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Focusing, Collimating, and Imaging Optics





J.-H. Kim, et al, J. Synchrotron Rad., 23, 880-886, (2016)



refocusing $\hat{O}_{\rm R}$

1D imaging optics f_2 : graded multilayer mirror

X-ray echo spectrometer design & performance



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Momentum transfer resolution

The required momentum transfer resolution ΔQ deterines the needed angular acceptance $\Omega_x \times \Omega_y$ of the collimating optic $f_1: \Delta Q \simeq \Omega K$ ($\Omega = \Omega_x \simeq \Omega_y$)

 $\Delta Q = 0.1 \text{ nm}^{-1} \Rightarrow \Omega = 2.2 \text{ mrad } 09.1 \text{ keV}$ $\Delta Q = 0.45 \text{ nm}^{-1} \Rightarrow \Omega = 10 \text{ mrad } 09.1 \text{ keV}$ (proven Montel mirror acceptance)



X-ray echo spectrometer design & performance



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X-ray echo spectrometer design & performance



Vertical spot size on the sample $\Delta X_{ m D}$



Vertical spot size on the sample



Vertical spot size on the sample

defocusing $\hat{O}_{\rm D}$ refocusing $\hat{O}_{\rm R}$ $\Delta X_{\rm D} = G_{\rm D} \Delta E_{\cup \rm D}$ Det f_2 l_1 \mathcal{Z} lo Typically: $\Delta E_{\cup_{\mathrm{D}}} > \Delta E_{\cup_{\mathrm{R}}}$. Therefore, the spot size (a)accepted by the refocusing D_{R} f_1 D_{D} system $\Delta X_{\mathrm{R}} < \Delta X_{\mathrm{D}}$: Vertical scattering plane $\Delta X_R'$ $\Delta X_{\rm R} = G_{\rm D} \Delta E_{\cup_{\rm R}}$ If $\Delta X_{
m B}/f_1 > \Delta heta_{D_{
m B}},$ Ф the spot size and the spectral bandwidth accepted by the refocusing system is further reduced (b) $Q \simeq 2K\sin(\Phi/2)$ to: $\Delta X'_{\rm R} \simeq f_1 \Delta \theta_{D_{\rm R}}$ Horizontal scattering plane

 $\Delta heta_{D_{
m R}} \simeq 250~\mu$ rad is the

angular acceptance of $D_{\rm R}$.

 $\Delta X'_{
m p} \simeq 50~\mu{
m m}$, for $f_1=0.2~{
m m}$.

X-ray echo spectrometer design & performance



Scattering volume: absorption limited

Signal strength scales with the scattering volume and, therefore, with the absorption length, provided sample thickness is larger.

Sample	$L_{\rm a}^{ m XES}$	$L_{\rm a}^{\rm HERIX}$	$L_{\rm a}^{\rm HERIX}/L_{\rm a}^{\rm XES}$		
YBa ₂ Cu ₃ O ₇	10	300	30		
$HgBa_2Ca_2Cu_3O_8$	4	34	8.5		
$Bi_2Sr_2Ca_2Cu_3O_{10}$	7	31	4.5		
SmFeAs	6	58	10		
CeFeAs	8	70	9		
$NbSe_2$	16	32	2		
VO_2	18	260	14		
Fe_3O_4	8	140	18		
$SrTiO_3$	50	180	3.6		
Sr ₃ CuIrO ₆	36	120	3.3		
$\mathrm{URu}_2\mathrm{Si}_2$	3	9	3		
H_2O	1500	21000	14		

Table 3: Absorption/attenuation lengths L_{a} in selected high- T_{c} superconductors and other materials with diverse properties of fundamental and practical interest at the operating photon energies of the HERIX (23.74 keV) and XES (9.13 keV) spectrometers. Larger absorption length is advantageous for the IXS signal strength. Smaller absorption length is advantageous if the samples are inhomogeneous.

Scattering volume: sample thickness limited

Thin-film samples ($\lesssim 1~\mu$ m)

Same scattering volume for 9-keV (XES) and for 24-keV (HERIX) x-rays. Expected signal strength enhancement $\simeq 10^3$ with a 1-meV-resolution x-ray echo spectrometer compared to HERIX.

Samples in diamond anvil pressure cells $(3 - 40 \ \mu m)$

			- medium
pressure	sample diameter	sample thickness	Ruby
GPa	μ m	μ m	Light,
30	100-200	20-40	A-Tays
60	60-100	10-15	Diamon
100	50-70	<10	
200	30-40	$\simeq 3$	Sample

Sample size depends on pressure range. A few typical examples (Victor Struzhkin).

Same or similar scattering volume for 9 keV x-rays (XES) and 24 keV HERIX. Though, transmission of the 9-keV photons through a 2-2.4-mm thick diamond is 10-times smaller, the expected signal strength enhancement is still very high $\simeq 170 - 55$ with a 1-meV-resolution x-ray echo spectrometer compared to HERIX.

0.05/3 µm

0.5 to 1.5 µm

0.5 to 1.5 um

0.5 to 1.5 um

0.5 to 1.5 um

Metal gasket /

Ni/Al

Cu (In, Ga) sea

ZnO

CdS

Mo Kapton

X-ray echo spectrometer design & performance



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X-Ray Echo Spectrometer Efficiency





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X-Ray Echo Spectrometers compared with HERIX

Spectrometer:	XES1	XES05	XES02	XES01	HERIX
Parameter:					
Photon energy E [keV]	9.13	9.13	9.13	9.13	23.74
Photon momentum K [nm ⁻¹]	46.2	46.2	46.2	46.2	120.3
Spectral resolution $\Delta \varepsilon$ [meV]	1	0.5	0.2	0.1	1.5
Spectrometer bandwidth ΔE_{\cup} [meV]	14.2	14.2	5.5	5.5	0.9
Momentum transfer resolution ΔQ [nm ⁻¹]	0.4	0.2	0.12	0.02	1.2
Angular acceptance $\Omega_v imes \Omega_h$ [mrad ²]	10×10	5×5	2.5×10	0.43×5	10×10
Max. scattering angle $\Phi_{_{\rm M}}$	154°	154°	154°	154°	35°
Max. momentum transfer $Q_{\rm M}$ [nm ⁻¹]	90	90	90	90	70
Analyzer arm size [m]	2	3.5	3.0	3.5	9
Incident photon polarization	π	π	π	π	σ
Spotsize (V×H) on the sample $[\mu m^2]$	70×5	140×5	130×5	280×5	20×35
Required detector resolution [μ m]	14	10	13.5	6.8	-
Spectral flux F @APSU [ph/meV/s] $\times 10^{10}$	30	30	30	30	4.2
Rel. signal strength $S^{\text{XES}}/S^{\text{HERIX}}$	$1767/\xi$	$442/\xi$	$66/\xi$	$5.8/\xi$	1

Table 1: Operation parameters and performance characteristics of the proposed echo spectrometers XES1, XES05, XES02, and XES01 compared with the parameters of the IXS spectrometer HERIX at 30ID@APS. The signal strength $S \propto \Delta E_{\cup}^2 \times \Omega_v \times \Omega_h \times F \times L_s$, where L_s is the scattering length. The relative signal strength values in the bottom row have to be corrected for each particular sample by the ratio $\xi = L_s^{\text{HERIX}}/L_s^{\text{XES}}$. The scattering length L_s is related either to the absorption length L_a , or to the sample thickness L. Typically $\xi \simeq 2 - 30$. A $5 \times 5 \ \mu \text{m}^2$ *monochromatic* focal spot size on the sample is assumed in all cases.

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Ultra-high-resolution IXS spectrometer (UHRIX)



Ultra-high-resolution IXS spectrometer (UHRIX)

Phonons in Glycerol

(a)

50

 $Q = 1.4 \text{ nm}^{-1}$

0.06

0.04

0.02

• UHRIX: demonstrated at the APS in 2013

• Features high-contrast spectral resolution function with a 0.6-meV bandwidth.

• Enables IXS with a sub-meV spectral and a 0.25nm⁻¹ momentum transfer resolution.



X-Ray Echo Spectrometers vs UHRIX



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• The narrow-band angular dispersive multi-crystal CDDW monochromators and analyzers used in UHRIX, have to be converted into broadband CDDW dispersive elements.

• $2-\mu$ m-resolution charge integrating pixel detectors with single photon sensitivity are state of the art A. Schubert et al, J. of Synchrotron Rad. 19 (2012) 359

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• The spectral resolution of the x-ray echo spectrometers does not rely on the monochromaticity of the x-rays, ensuring strong signals along with a very high spectral resolution.

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• X-ray echo spectroscopy, a counterpart of neutron spin-echo, is being introduced to overcome limitations in spectral resolution and weak signals of the traditional inelastic x-ray scattering (IXS) probes.



• The spectral resolution of the x-ray echo spectrometers does not rely on the monochromaticity of the x-rays, ensuring strong signals along with a very high spectral resolution.

• The x-ray echo spectrometers will allow, either, up to $\simeq 1000$ -fold reduction in measurement time for experiments at presently available $\simeq 1 \text{ meV}/1 \text{ nm}^{-1}$ resolution, or an unprecedented $\simeq 0.1 \text{ meV}/0.1 \text{ nm}^{-1}$ resolution with a up to $\simeq 10$ -fold improved signal strength.



Time-Length Space of Excitations & Scattering Probes



Time-Length Space of Excitations & Scattering Probes



Time-Length Space of Excitations &
Scattering Probes λ [nm]

