Particle scattering, particularly of photons, is one of the most widely used tools in science. The measured quantity is usually the particle flux or the intensity. The phase of the particle field, or more precisely the phase change of the field during the scattering process, goes undetected by the intensity measurement. The related loss of information is known as the “phase problem” in various areas of research.

Methods of phase determination are commonly based on creating two or more indistinguishable paths for the particle where at least one path does not include the sample. The resulting interference pattern is then accessible by intensity measurements. Interferometric techniques with x rays, light, neutrons, or atoms have been developed in this context. Results usually depend on details in the detected interference pattern, and measurements become exceedingly difficult for short wavelengths. In the x-ray regime, thermal and mechanical stability of the interferometer (IFM) are crucial, and the spatial limitations on a sample in close proximity to sensitive components of the IFM can be problematic for experimental work. Here we discuss an x-ray interferometric method that avoids this problem.

The typical x-ray IFM works with sample and reference placed in spatially separate paths inside the IFM. This allows the comparison of phase changes to the photon field caused by sample and reference. Besides potential stability problems, interference is not achieved if the direction of the x rays is changed by a scattering process in the sample. The spatial separation of x-ray IFM and sample for phase measurements would be highly desirable but to our knowledge has not been demonstrated previously. In this paper, we present measurements of phase changes caused by a sample placed outside the IFM, i.e., after recombination of the beam paths. We use the term “exo-interferometry” to emphasize this fact. Exo-interferometry permits us to analyze phase changes of x rays that were transmitted, reflected, or diffracted by a sample material. Our approach is based on recent theoretical work on time-dependent phase determination using an x-ray IFM. For experimental verification, we used a coherent nuclear resonant scattering process, which produces a signal that is sufficiently delayed to be observable by x-ray detectors. Previous experiments on nuclear resonant interferometry placed samples inside the IFM and did not attempt to obtain the time-dependent phase. The principles of exo-interferometric phase determination are elucidated next.

The addition of field amplitudes in the IFM allows the production of a time-dependent field with a phase that can be manipulated in a controlled way. Consider a very short x-ray pulse described by $\delta(t)$ that is incident on a triple-Laue IFM similar to the schematic shown in Fig. 1. If we place a reference material with response function $R(t)$ in one of the paths of the IFM and apply an additional phase shift $\alpha$ between the two paths, the exit field will be roughly proportional to $\exp[i\alpha] \delta(t) + R(t)$. It will be useful to introduce the delayed response explicitly. Let $R(t) = r_0 \delta(t) + R'(t)$, where $R'(t)$ is the delayed response that vanishes for “early” times $t < \tau$ after arrival of the x-ray pulse. The field amplitude after the IFM is then given by

$$r_0 + e^{i \alpha} \delta(t) + R'(t).$$ (1)

At early times, modulus and phase of the field are controlled by the phase shift $\alpha$, whereas at later times the components of the field are unaffected. The basic idea of exo-interferometric phase determination is now to mix field components with different time coordinates simply by scattering the x-ray field described by Eq. (1) off a sample. The degree of mixing will depend on the properties of the sample and on the adjustable value of $\alpha$. If the response of the sample material is prompt, i.e., proportional to $\delta(t)$, mixing does not occur – a delayed response is required. Assume the transmission through a sample with response function $S(t) = s_0 \delta(t) + S'(t)$ with delayed part $S'(t)$. The expression for the field amplitude after the sample contains three terms

$$s_0 (r_0 + e^{i \alpha} \delta(t) + \{s_0 R'(t) + (r_0 + e^{i \alpha}) S'(t)\} + R' S'(t)).$$ (2)

The first term describes conventional x-ray interferometry. The second term is reminiscent of the coherent su-
perposition of delayed reference and sample response that is observed when placing the sample inside the IFM. The last term is a convolution integral that also gives a delayed contribution. A similar expression, but all terms with $\alpha$ being absent, has been used to describe “time-domain interferometry” [11, 12].

The previous arguments lead to the following conclusions about the feasibility of exo-interferometric experiments. Reference and sample must produce time-delayed responses, and the convolution of the responses should be negligible, i.e., either the spectra of $R'$ and $S'$ have a small overlap or the response is sufficiently weak. There must be a time scale $\tau$ that clearly separates “early” and “delayed.” However, the time scale does not enter quantitatively – it is mostly determined by experimental circumstances. The response time of an x-ray IFM using single crystals with low-order Bragg or Laue reflections is of the order $10^{-15}$ s. In the present experiment, the x-ray pulses incident on the IFM had a longitudinal coherence time of about 0.3 ps corresponding to an energy bandwidth of 2 meV. In addition, the average length of the synchrotron radiation (SR) pulse, 70 ps, and the time resolution of the x-ray detector, 1 ns, have to be considered. Mainly caused by detector resolution, the time scale $\tau$ is placed in the nanosecond regime. The time scale for the nuclear-resonant response of sample and reference is determined by the lifetime of the nuclear excited state, which is 141 ns for the 14.4125 keV nuclear level of the $^{57}$Fe isotope used here.

Experiments were performed at sector 3 of the Advanced Photon Source. The storage ring was filled with 23 electron bunches producing about $6.2\times10^6$ x-ray pulses per second. The SR was monochromatized to about 2 meV bandwidth to avoid overload of the detector system while maintaining the spectral brightness of the x rays around the nuclear transition energy. After reduction of the beam size to $0.3\times0.3$ mm$^2$, the photon flux was $10^8$ ph/s and about $3\times10^7$ ph/s after the triple-Laue IFM. A schematic of the experimental setup is shown in Fig. 1. Three situations were realized: the sample is placed outside the IFM and magnetized collinear with the linear polarization of the SR, as before but magnetized perpendicular to polarization and propagation direction of the SR, the sample is mounted inside the IFM and magnetized parallel to the polarization direction of the SR. In all cases, the reference was a 1.1-µm-thick foil of stainless steel (Fe$_{0.55}$Cr$_{0.25}$Ni$_{0.25}$) that was 95 % enriched in the $^{57}$Fe isotope. The sample outside the IFM was a 2.5-µm-thick iron foil that was 53 % enriched in the $^{57}$Fe isotope. The control measurements with sample inside the IFM used a 2.2-µm-thick iron foil that was 95 % enriched in the $^{57}$Fe isotope. Very good knowledge of the time response of magnetized iron and stainless steel motivated this choice.

For each experimental situation, we measured six time

![FIG. 1: Experimental setup. The incident SR is split at the first face of the triple-Laue IFM (LLL). The reference, R, is placed in the lower beam path. A rotatable 0.5-mm-thick, polished Be platelet works as a phase retarder, P. The sample, S, is kept spatially separate from the IFM, except in the control experiment, where it is placed next to the reference but in the upper beam path. The avalanche photodiode detector, D, provides time-resolved spectra.](image1)

![FIG. 2: Time spectra (a),(b) and phase angle (c) for the control case, i.e., the sample mounted inside the IFM. The time spectra (a) correspond to phase-retarder settings of $\pi$, $-\pi/2$, 0, and $\pi/2$ (top to bottom). The time spectrum (b) is the response of the sample only, i.e., the reference-beam path was blocked. The solid line is a fit to the data. The time-dependent phase angle was either extracted directly from spectra (a) using Eq. (3), (symbols), or calculated with parameters obtained from the fit of (b) and an isomeric shift of 1Γ, (line).](image2)
FIG. 3: Time-dependent intensities (a) and phase angles (b) for the sample mounted outside the IFM and magnetized perpendicular, A, or parallel, B, to the polarization of the SR. The solid lines are fits to the intensity data. The phase angles were either extracted directly from spectra using Eq. (3) as explained in the text, (symbols), or calculated with parameters obtained from the fits of (a) and an isomeric shift of $1\Gamma$, (line).

spectra: the phase retarder adjusted to $\alpha = 0, \pi, \pm \pi/2$, and either IFM path blocked. The spectra were collected in back-to-back sequences of 10 min/spectrum to minimize systematical errors caused by potential beam instabilities and temperature drifts. Temperature variations during the experiment were less than 150 mK. For the control case, the relative phase change of the x-rays by the sample is calculated from the four interference spectra $I(\alpha)$ without specific knowledge about the IFM imperfections. With $\xi = I(0) - I(\pi)$ and $\eta = I(\pi/2) - I(-\pi/2)$, we obtain from ref. [7] for the time-dependent phase angle

$$e^{i\phi} = \frac{\xi + i\eta}{\sqrt{\xi^2 + \eta^2}}.$$  \hspace{1cm} (3)

The time spectra for the different phase-retarder settings, the time spectrum of the sample alone, and the derived phase angle, $\phi$, are shown in Fig. 4. A standard evaluation code, CONUSS [13], was used to fit the spectrum of the sample. The excellent agreement supports the notion of well-defined hyperfine interaction parameters of the chosen iron foil.

If the sample is placed outside the IFM, Eq. (2) is applied, and, if one neglects the convolution integral, the relative phase results from Eq. (3) with the replacement $\xi = I(0) - I(\pi) - 4f I_S$. The last term is composed of the time spectrum of the sample, $I_S$, and a factor $f$ that accounts for absorption in the reference and IFM imperfections. $f$ is the product of IFM visibility, 0.45 in this case, and intensity ratio of the individual IFM paths. We determined experimentally $f = 0.504$. Fig. 4 shows the time-dependent intensities and phases for the two directions of magnetization with the sample outside the IFM. In Figs. 2 and 3 one notices overshoots at phase jumps, i.e., at sign changes of the transmitted electric field associated with zero intensity. In those minima, the measurement is most sensitive to effects of the detector resolution and potentially other systematic errors. However, the resulting phase errors are associated with very small field amplitudes, which reduces their significance.

FIG. 4: Reconstructed energy spectra for the exo-interferometric case and sample magnetized perpendicular, A, or parallel, B, to the polarization of the SR, and the control case with sample inside IFM and magnetized parallel, C. The solid lines are fits to the average spectra (circles). Spectra from individual sequences are shown as shaded dots. The energy scale is given in units of the nuclear level width $\Gamma = 4.66$ neV.
The determination of the time-dependent phase as described in the previous section did not require specific knowledge of reference or sample. The phase angle together with the measured intensities of the sample alone, as displayed in Figs. 2(b) and 3(a), permits us to reconstruct the energy response $S(E)$ of the sample by Fourier transformation

$$\tilde{S}(E) = \int S_s(t) \exp(i \phi(t)) \exp(i E t / \hbar) dt.$$  \hspace{1cm} (4)

In this expression, $S_s(t)$ is the time spectrum of the sample, shown in Figs. 2(b) and 3(a), and $\phi(t)$ is the phase angle determined earlier and shown in Figs. 2(c) and 3(b). $\tilde{S}(E)$ was calculated for each sequence of data (about 1h collection time). In Fig. 4 we show the reconstructed energy spectra $|\tilde{S}(E)|^2$ for each data sequence, the average of all data sequences, and the calculated energy spectra. The oscillation pattern in the time spectra and the separation of the resonances in the energy spectra are dominated by the magnetic field at the $^{57}$Fe nuclei. The results for magnetic field values agree within 0.03% for the control case and within 0.2% and 0.7% for the exo-interferometric situations A and B. In addition, the reconstructed energy spectra contain a small but noticeable overall shift, the so-called isomeric shift. One obtains 1.037(6)$\Gamma$ for the control case, in agreement with the literature value of 0.9(2)$\Gamma$ \cite{14}. The exo-interferometric situations A and B give 1.05(2)$\Gamma$ and 0.67(2)$\Gamma$ for the same quantity. The values are given in units of $\Gamma=4.66$ neV, the width of the nuclear-excited state. In our measurement, we used an avalanche photodiode detector with about 1 ns resolution, which results in an accessible energy range in the reconstructed spectrum of about 140 $\Gamma=0.66 \mu$eV. The energy resolution is related to the observed duration of the time-delayed signal \cite{7}. Here the time duration of 116 ns was determined by the operating mode of the Advanced Photon Source and detector dead time. It leads to an energy resolution of 23 neV. The additional broadening of the resonances in Fig. 4 is caused by thickness-related self-absorption effects in the samples. The agreement of calculation and reconstructed spectra is generally very good even though Fig. 4 shows deviations, which appear to be stronger for the exo-interferometric cases. Also for the control case, a slight asymmetry in the spectrum can be observed. Possible explanations include inhomogeneity in sample or reference and non random imperfections of the IFM. At present, we have no conclusive explanation for these small but noticeable effects. In situation B, our neglect of the convolution term in Eq. 2, which amounts to an approximation in the process of phase determination, leads to larger deviations of, e.g., the measured isomeric shift. A glance at Fig. 5 shows that two resonances of the sample are rather close to the origin of the energy scale, which marks the position of the single resonance of the stainless-steel reference. The overlap of the energy spectra leads to a stronger contribution of the convolution term than in situation A, and its neglect causes a larger distortion in the reconstructed energy spectra.

Exo-interferometry has been introduced as spectroscopic use of an x-ray IFM with distinct advantages in sample utilization. The ideas of exo-interferometry are general, but potential applications need to use technically available time scales, which are essentially given by the best time resolution of x-ray detectors. At present, streak cameras provide ps resolution, which could make a meV-energy range accessible with $\mu$eV resolution. The presented data have unambiguously demonstrated that time-dependent phase changes in x-ray fields can be measured with samples positioned inside or outside an x-ray IFM. To demonstrate the principle, our studies were conducted in transmission geometry. But, in contrast to conventional x-ray interferometry, crystal diffraction or surface reflection could be investigated as well. For example, in pump-probe time-resolved x-ray diffraction experiments \cite{12}, exo-interferometric phase determination may become a very useful tool. It also becomes possible to manipulate the x-ray beam between IFM and sample, e.g., by focusing or polarization control.

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