Using the photoinduced $L_3$ resonance shift in Fe and Ni as time reference for ultrafast experiments at low flux soft x-ray sources

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Somnath Jana,1,a) Shreyas Muralidhar,2 Johan Åkerman,2 Christian Schüßler-Langeheine,1 and Niko Pontius1

AFFILIATIONS
1Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, 12489 Berlin, Germany
2Department of Physics, University of Gothenburg, 412 96 Gothenburg, Sweden
a)Author to whom correspondence should be addressed: sj.phys@gmail.com

ABSTRACT
We study the optical-pump induced ultrafast transient change of x-ray absorption at $L_3$ absorption resonances of the transition metals Ni and Fe in the Fe$_{0.5}$Ni$_{0.5}$ alloy. We find the effect for both elements to occur simultaneously on a femtosecond timescale. This effect may hence be used as a handy cross correlation scheme, providing a time-zero reference for ultrafast optical-pump soft x-ray-probe measurement. The method benefits from a relatively simple experimental setup as the sample itself acts as time-reference tool. In particular, this technique works with low flux ultrafast soft x-ray sources. The measurements are compared to the cross correlation method introduced in an earlier publication.

INTRODUCTION
Availability of femtosecond ultrashort x-ray pulses leads to exciting insights into a plethora of ultrafast processes in solid state, molecular, or atomic physics. Most common for time-resolved x-ray experiments is the combination of an ultrashort x-ray probing pulse with a synchronized laser pump pulse in the infrared, visible, or ultraviolet range. Mandatory for properly conducted time-resolved experiments is a reliable reference for the time delay between the pump and the probe pulse, i.e., a time normal to which all measured data can be referred to. Ideally, such a time reference even provides an independent determination of time-zero, i.e., the time of simultaneous arrival of laser and x-ray pulse at the sample. The independent experimental determination of time-zero removes uncertainties of deriving a per se unknown dynamic response of the sample under investigation.

Different cross correlation schemes have been developed for this purpose. Cross correlation schemes usually provide an indirect measurement of time zero. In a sequential linear cross correlation process, photons of one pulse create excited states in a suitably sampled state that change its response, $R(t)$. The other pulse probes this altered response as a function of delay. To derive time zero from this measurement with high reliability, the response $R(t)$ has to be known precisely and involve timescales shorter than or similar to the experimental resolution. Finally, the probed response $R(t)$ is experimentally blurred by the temporal duration of the exciting and probing pulse. Effectively, the experimentally probed response corresponds to the convolution of the time dependent response $R(t)$ with the experimental temporal resolution allowing one to experimentally deduce time zero from this measurement.

For ultrashort x-ray sources without any natural intrinsic synchronization, e.g., Self Amplified Spontaneous Emission free electron lasers, an experimental shot-by-shot analysis of the relative time relation of x-ray and laser pulses is crucial to achieve a sub-100 fs synchronization and the corresponding experimental time-resolution. To this end, several x-ray-optical cross correlation schemes on a shot-to-shot basis have been put forward and are routinely used. However, even for ultrafast x-ray sources with an inherent, natural synchronization between x-ray and laser pulse (e.g., high-order harmonic generation, laser-driven plasma, or storage-ring based slicing sources), an independent absolute time reference is generally essential to put the measured transients onto a proper timescale. This is particularly important when the dynamics of different atomic species or that of separate subsystems probed through different observables shall
be compared. So do, for example, the recent results of delays between the electronic and magnetic responses in metallic magnets\cite{28,29} and delayed response in Ni demagnetization with respect to Fe in the FeNi alloy\cite{30,31} measured at extreme ultraviolet energies emphasize the need for an independent time-zero reference at soft x-ray energies. Beyond these physical requirements, a regular referencing of the timescale during extended measurements may be essential for correction of slow experimental drifts of time-zero, e.g., caused by thermal changes of the optical path lengths of the laser or x-ray branches.

Several common cross correlation schemes are based on dynamic effects induced by high power x-ray pulses,\cite{32,33,34,35} leading to a transiently altered material response, which is then probed by the optical laser pulse. These are well suited for intense ultrashort x-ray sources like Free Electron Lasers, but do not work for low x-ray fluxes like at High Harmonic Generation or slicing sources. In Refs. 28 and 29, a soft x-ray-optical cross-correlator scheme has been put forward, which is capable of providing a defined time reference for low flux soft x-ray sources with tunable photon-energy. It utilizes the laser excited displacive coherent phonon oscillation (DCPO) in a Molybdenum–Silicon (Mo/Si) multilayer\cite{36} as the pump induced response. The DCPO is probed through intensity variations of the first, second, or third order x-ray Bragg diffraction peak of the Mo/Si layers superstructure. The phase of this coherent oscillation provides a defined time reference for the time-resolved laser-pump and x-ray probe experiment. The phase shift of the DCPO in MoSi, with respect to the effective time zero of the pump-probe experiment, however, was experimentally not determined. Therefore, this cross correlator so far only supplies a relative time reference.

In this report, we study another soft x-ray-optical cross correlation scheme suited for low-flux x-ray sources. It is based on a pump-laser induced transient change of the L₃ resonance x-ray absorption spectrum (XAS) observed in different 3d transition-metal systems.\cite{37,38,39} This transient response reflects the excitation of the 3d valence electron system occurring within a few femtoseconds after photoexcitation by the ultrashort laser pulse. We study this effect for Ni and Fe in a Fe₀.₅Ni₀.₅ alloy sample, utilizing linearly polarized x-ray pulses. The transient absorption changes are compared to the phase determined from the measured DCPO cross correlation,\cite{36} which is measured concurrently. We find that fitting the transient change of the x-ray absorption by modeling the laser induced electronic excitation and relaxation process allows for an estimation of the time of photoexcitation, i.e., time zero with a precision of a few tens of femtoseconds. The advantage of the proposed cross correlation measurement is a relatively small physical requirements, a regular referencing of the timescale during extended measurements may be essential for correction of slow experimental drifts of time-zero, e.g., caused by thermal changes of the optical path lengths of the laser or x-ray branches.

As shown in the inset of Fig. 1(a) (at Ni L₃ edge), this transient modification is most pronounced at the low energy slope of the L₃ resonance, where it appears as a transient increase in absorption. Overall, the effect looks like a redshift of the absorption resonance. The absorption at the low energy slope increases; afterwards, it was found to partially recover within the subsequent picoseconds.\cite{25,32,33,35,39} Similar ultrafast modifications of the L₃ absorption spectrum have been reported for cobalt in a Co/Pd-multilayer\cite{34} as a non-linear effect when probing the sample absorption by single, intense ultrashort x-ray pulses.

These transient absorption changes have been assigned to pump-laser induced 3d valence electron excitations even though the descriptions of the process vary in their details. References 3, 32, and 37 relate the ultrafast L₃ modification to an increase in the 3d electron localization resulting from a transient reduction of the nearest neighbor 3d-4p hybridization in Ni as a consequence of the electronic excitations. Similar spectral changes in the Ni L₃ absorption have been calculated by theory and attributed to state blocking, i.e., a redistribution of 3d valence electrons from below to above the Fermi level through the optical excitation. The subsequent recovery of the absorption on a picosecond timescale has been assigned to the cooling and relaxation of the valence electron system through thermal coupling to the lattice.

Even if the exact physical mechanism behind the absorption changes at the 3d metals L₃ resonances upon laser excitation is still under discussion, a common base of all interpretations is that it is associated with the pump-induced excitation of the 3d valence electron system. We hence would expect the magnitude of the effect to depend on the number of valence electrons excited from below to above the Fermi edge. The maximum number of excited electrons is reached when the photoabsorbed energy has spread in the valence electron system through inelastic electron–electron scattering cascades. This occurs within a few tens of femtoseconds after photoexcitation\cite{40,41} and leads to a Fermi–Dirac-like hot electron distribution. Subsequently, the number of excited electrons reduces when the electron system cools down, leading to the recovery effect seen in the L₃ absorption change.

**EXPERIMENT**

As in Ref. 28, the experiments were performed at the BESSY II Femtoseconds beamline (UE6/1-ZPM).\cite{42} Vertically polarized x-rays were used to measure both the XAS in transmission and the superlattice Bragg peak in reflection (see Fig. 1). We used a Fe₀.₅Ni₀.₅ alloy thin film to measure the time dependent L₃ resonance XAS and a [Mo(1.86 nm)/Si(2.07 nm)]₄₀ multilayer to probe the oscillation of the first order superlattice peak (SL1) at both the Fe and the Ni L₃ edge energies. The Fe₀.₅Ni₀.₅ (25 nm) sample has been deposited on a Ta(3 nm)/Al(0.3 μm)/Si₃N₄(0.2 μm) substrate, where the Al layer acts as a heat sink and the 3 nm Ta as a seed layer. Another 3 nm Ta capping protects the sample from oxidation when exposed to air. An infrared laser of 800 nm wavelength, 3 kHz repetition rate, and approximately 50 fs pulse duration was used to excite the samples. The pump laser beam cross section was with 0.35 × 0.25 mm² larger than the corresponding x-ray probe size of 0.15 × 0.04 mm². The XAS measurements were performed at 45° incident angle to increase the effective thickness and with that the absorption of the Ni₀.₅Fe₀.₅ sample.

For the detection of the superlattice (SL) Bragg peaks, the sample surface relative incident angle for the measurement was 13.5° at the Fe.  

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edge energy (~707 eV) and 11.2° at the Ni energy (~858 eV) (compare Fig. 1). The pump fluence was set to about 80% of the damage threshold for both the samples in order to maximize the pump-induced change without risking sample degradation. The data acquisition was performed by alternatingly measuring the x-ray absorption signal with and without pump pulse (referred to in the following as pumped and unpumped signal, respectively) on a shot-by-shot basis. Here, the slicing source was running at 6 kHz and the pump laser at 3 kHz, allowing for the elimination of intensity fluctuations slower than the repetition rate.42 To further eliminate slow drifts of time zero, our measurement protocol implemented alternating data collection between the alloy and the Mo/Si multilayer sample on an hourly timescale. The details of the correction procedure are given in the Appendix.

Figures 2(a) and 2(c) show the transient relative absorption change measured at the low energy \( L_3 \) resonance slope of Fe and Ni [compare Fig. 1(a), inset for Ni], respectively. The symbols (circles and squares for Fe and Ni, respectively) represent the experimental data, and the dashed lines are the least squares fits to the experimental data (see below). The measured transients are obtained from the measured data by calculating the relative change of the pumped XAS signal with respect to the unpumped. Both XAS transients show a sharp absorption increase up to about 0.4%, which peaks at about 100 fs delay. A slower gradual drop follows the initial quick rise. While the Ni transient levels off at about 0.25% after 1 ps, the Fe transient changes sign after 0.5 ps and decreases to ~0.15%.

We model the XAS cross correlation function, \( C_{\text{XAS}}(t) \), as the convolution of the experimental resolution, \( P(t) \), and the transient response from the sample \( R_{\text{XAS}}(t) \),

\[
C_{\text{XAS}}(t) = P(t) * R_{\text{XAS}}(t),
\]

\( P(t) \) is a normalized Gaussian function with FWHM = 130 fs (Ref. 35) modeling the experimental temporal resolution; \( R_{\text{XAS}}(t) \) is assumed as

\[
R_{\text{XAS}}(t) = H(t - t_0) \times \{ a - b \times (1 - \exp(- (t - t_0)/t_1)) \}.
\]

Here, the first term is a Heaviside step function scaled by \( a \). This approach assumes the photoinduced XAS change to be quasi-instantaneous on the timescale of the experimental temporal resolution. The second term takes care of the relaxation of the signal modeled through a single exponential decay. The fits are shown in Figs. 2(a) and 2(c).
Figures 2(b) and 2(d) display the measured DCPO of the Mo/Si-multilayer each probed on the corresponding SL1-Bragg peak at identical photon energies as used in (a) and (c), respectively. Both traces show the first three DCPO oscillation periods as known from Ref. 28. As described in the experimental section, the XAS and the DCPO transients of each photon energy have been measured in repeated alternation, which means that they are plotted on an identical delay scale (see the Appendix).

The transient response for the DCPO cross correlation of the MoSi sample is described correspondingly by

$$
C_{\text{DCPO}}(t) = p(t) * R_{\text{DCPO}}(t).
$$

Here, the response function $R_{\text{DCPO}}(t)$ of the DCPO is assumed as

$$
R_{\text{DCPO}}(t) = 0 \quad \text{for } t \leq t_0,
$$

$$
R_{\text{DCPO}}(t) = A \times (O(t) - 1) + B \times (t - t_0) \quad \text{for } t > t_0.
$$

Here, $O(t)$ describes the damped cosine like DCPO by

$$
O(t) = \cos(2\pi(t - t_0)/p) \times \exp(-(t - t_0)/T_r).
$$

with $T_r$ the damping time of the oscillation amplitude, $p$ the oscillation period, and $(t_0 - t_0)$ is the phase offset of the cosine function with respect to time zero $(t_0)$. The latter entails the delay of the oscillation onset, which is related to the electron-phonon interaction time in Mo. The constant $A$ represents the amplitude of the displacement, leading to an intensity drop of the SL peak intensity upon laser excitation. The linear term $B \times (t - t_0)$ models a slow linear change of the oscillation offset with time. The fit results of $C_{\text{DCPO}}(t)$ to the experimental data are shown as dashed lines in Figs. 2(b) and 2(d).

The most relevant individual fitting parameters for the experimental data are summarized in Tables I and II with their one-sigma uncertainties. The mechanism for DCPO does not depend on the probe x-ray photon energy and should yield, apart from statistical errors, the same time dependence for the Ni and Fe photon energies. We therefore relate the time axes of both Fe and Ni transient XAS change to the DCPO measurement tentatively using $t_0 = 0$ as time-zero reference.

The obtained onset $(t_0)$ from the fitting of the XAS transients reveals almost simultaneous response for Ni and Fe $(t_0 = -3 \pm 8$ fs; compare Table II) within our experimental error bar. The transient

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**FIG. 2.** (a) and (b) [(c) and (d)] show the dynamics of XAS signal at the rising edge of Fe (Ni) L3 absorption resonance and the oscillation of the superlattice peak intensity in MoSi-multilayer at the same energy, respectively. The circles (squares) are the experimental data, and the dashed line corresponds to the fit. The red (blue) shades are filled between the fit and $\pm 1$ standard deviation of the data points estimated from the scatter of data before the arrival of the pump pulse. The black vertical lines of panel (a) and (c) indicate the time-zero $(t_0)$, and the shades are filled between $t_0$ and $\pm 1$ standard deviation of the $t_0$ parameter as derived from the fitting. The delay axis is aligned with respect to the phase $(t_0)$ of the corresponding displacive oscillation. See text for the details of the fitting.
XAS change for Fe and Ni can hence be used as time reference for ultrafast dynamics experiments with a precision of approximately ten fs using the rather simple fit model presented above. The different recovery behavior of the two ions with the sign change found only in Fe suggests that a more elaborate model may further improve the precision as would a better statistics of the data. Furthermore both the XAS transients start almost simultaneously with a DCPO oscillation with phase \( t_0 = 0 \), which hints toward a quasi-instantaneous response of the DCPO upon laser excitation for our experimental conditions. Relative to \( t_0 = 0 \), the obtained \( t_0 \) from the XAS transients are \(-10 \pm 7 \) and \(-7 \pm 6 \) fs (see Tables I and II) for the Fe and Ni L\(_3\) edges, respectively, which implies an onset of the DCPO delayed by \( 9 \pm 9 \) fs.

A \( t_0 \) value close to zero appears reasonable, in particular, since we performed the DCPO measurement at a rather high pump fluence. While in general the phase lag can be fluence dependent, it has been shown in Ref. 43 that the phase lag converges to zero for the high fluence in the case of SrRuO\(_3\)/SrTiO\(_3\) multilayer system.

We notice that the fit to the Ni XAS rising slope does not describe the experimental data as well as for the Fe. For Ni, we get a better agreement assuming a temporal resolution of \( \sim 220 \) fs instead of 130 fs. We assign this to a possible incomplete drift correction for Ni. The fits with different temporal resolutions give the same values for \( t_0 \) within our error bars (\(-8 \pm 10 \) fs for temporal resolution of 220 fs).

In the present study, we utilized linearly polarized x-ray pulses. The XAS transient change will be detectable with circular polarized x-rays as well. Then, however, a separation of the electronic response from magnetic dynamics is required, achieved, e.g., by averaging the signals measured for opposite circular helicities or magnetic field orientations, respectively.

In summary, we characterized a new cross correlation experiment for an ultrafast optical-pump soft x-ray-probe experiment suited for low intensity x-ray sources. The technique is based on probing the transient absorption change at the L\(_3\) resonances of the 3\(d\) transition metals Ni and Fe. Time zero is estimated from the experimental traces by associating the transients with the quasi-instantaneous excitation of the 3\(d\) valence electron system by the optical pump laser and the following relaxation. Within the measurements of this study, we are able to determine time zero by the proposed XAS cross correlation method within an experimental error of 7 fs.

**TABLE I.** Results obtained from the fitting of the oscillation of SL1 peak intensity in the Mo/Si sample at Fe and Ni L\(_3\) edges presented in Figs. 2(b) and 2(d). The errors correspond to the one sigma values.

|          | Fe      | Ni      |
|----------|---------|---------|
| \( A(\%) \) | 0.8 ± 0.02 | 0.67 ± 0.02 |
| \( p(\text{fs}) \) | 582 ± 6 | 585 ± 6 |
| \( t_0(\text{fs}) \) | 0 ± 5 | 0 ± 4 |

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**APPENDIX: TIME-ZERO DRIFT CORRECTION**

We have collected the data on the Fe\(_{0.5}\)Ni\(_{0.5}\) alloy and the Mo/Si sample alternatively on an hourly timescale to correct for slow drifts of time-zero throughout the experiment performed at each resonance energy. Figures 3(a) and 3(b) show the variation of the time-zero vs the measurement time obtained at Fe and Ni L\(_3\) energies, respectively. In this context, time-zero \( t_0 \) refers to the onset of the DCPO of the Mo/Si multilayer assuming the phase offset to be zero (see the main manuscript for the detail of the fitting function). As any individual DCPO scan is not suitable for refinement of all the parameters due to large noise, the average of all DCPO scans is fitted to obtain first guess values of different parameters, which are then used as fixed parameters to refine the onset \( (t_0) \) of any individual DCPO scan. Such obtained \( t_0 \) is then used to shift the delay-axis of the individual DCPO scan in order to calculate the

**TABLE II.** \( t_0 \) obtained from the fitting of the \( C_{\text{FeSi}} \) data at Fe and Ni L\(_3\) edges presented in Figs. 2(a) and 2(c). The errors correspond to the one sigma values.

|          | Fe      | Ni      |
|----------|---------|---------|
| \( t_0(\text{fs}) \) | \(-10 \pm 5\) | \(-7 \pm 6\) |

**FIG. 3.** Panels (a) and (b) show the variation of time-zero as a function of time at the Fe and Ni L\(_3\) edges, respectively. The blue squares are obtained from the fitting of each individual DCPO scan. The dashed line is obtained by applying a Savitzky–Golay filter as described in the text. The error bars correspond to the one standard deviation of the time-zero parameter of the fit-function when all other parameters are kept fixed (see the text for the detail). The red circles are the time-zero-offset for the individual delay scans measured in the FeNi–alloy, obtained by assigning their times to the smoothed curve (dashed line).
average of all DCPO scans for the next iteration. The whole process is repeated interactively until the convergence of each \( t_0 \) parameter to within \( \pm 1 \) fs is achieved. Figures 1(a) and 1(b) show the drift of the \( t_0 \) with time. A smoothed variation of the drift (dashed curve) is obtained by applying a Savitzky–Golay filter (with size 7 and order 3) three times consecutively on the obtained time zero variation (blue squares). The delay-axes of the transient XAS scans measured between the DCPO measurements are corrected by assigning their times to the smoothed curve.

**DATA AVAILABILITY**

The data that support the findings of this study are openly available in “HZB Data Service” at http://doi.org/10.5442/ND000007, Ref. 44.

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