

Imaging Ultrafast Dynamical Diffraction Wave Fronts in Strained Si with Coherent X Rays

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Dynamical diffraction effects in thin single crystals produce highly monochromatic parallel x-ray beams with a mutual separation of a few microns and a time delay of a few femtoseconds—the so-called echoes. This ultrafast diffraction effect is used at X-Ray Free Electron Lasers in self-seeding schemes to improve beam monochromaticity. Here, we present a coherent x-ray imaging measurement of echoes from Si crystals and demonstrate that a small surface strain can be used to tune their temporal delay. These results represent a first step toward the ambitious goal of strain tailoring new x-ray optics and, conversely, open up the possibility of using ultrafast dynamical diffraction effects to study strain in materials.

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The emergence of fourth generation synchrotron sources based on low-emittance rings [1] and of spatially coherent x-ray free electron lasers (XFELs) [2,3] open a new era in the study of high-coherence materials, such as perfect crystals, using high-coherence probes [4]. Recent years have seen an increase in the development and use of inverse microscopy techniques based on coherent x rays [5–11] and other types of speckle analysis [12,13] for the study of crystalline materials, as well as for the characterization of coherent x-ray beams [5,14–17]. These methods are based on the assumption of a Fourier transform relation between the sample's electron density and the x-ray scattered field, derived from the so-called kinematical approximation [18]. On the other end, large single crystals are one of the primary optical elements for x-ray experiments. For these, more complex interactions including multiple diffraction, absorption, and refraction effects must be considered, which are well described by the dynamical diffraction theory [18–20]. Dynamical diffraction effects have already proven very useful for applications in ultrafast x-ray optics [21]. Nevertheless, dynamical diffraction can arise already from samples of modest dimensions (~500 nm) and, in conjunction with the use of coherent nanobeams, have already been shown to be not only measurable [22,23] but also a hindrance to standard inversion algorithms for digital microscopies [24] and requiring adaptive solutions [25].

Here, we show that the exceptional coherence properties of the x-ray beam produced at MAX IV Laboratory can be exploited to understand dynamical diffraction effects in crystalline samples and how the wave front changes in the presence of strain and propose an approach to analyze them. The effect under study, presented theoretically in

Ref. [26], consists of the appearance of multiple monochromatic beams in the diffracted and forward directions, which present a transverse displacement of a few microns between each other. These beams, also known as echoes, are delayed between each other by few femtoseconds depending of the thickness of the crystal and energy of the x-ray beam. This ultrafast effect is used in x-ray optics at XFELs for self-seeding radiation production [21]. We have previously demonstrated the transverse displacement of the echoes in diamond plates [27] and Si wafers [28] using a 2 μm beam focused onto an optically coupled x-ray detector with submicron resolution. In this Letter, we improve the resolution of our previous works and demonstrate that localized surface strain in a Si wafer can produce a spatial modulation of the echoes that translates into the tuning of their time delay.

A nanobeam is used to select regions of a Si sample where a residual strain field was created by nanoindentation. The coherence of the x rays is exploited to measure the finely structured echoes, strongly dependent on the local strain field, with a sub-100 nm resolution only achievable with inverse microscopy. The technique used, known as x-ray teleptychography [29], is capable of measuring the x-ray diffracted field, using as a probe a small pinhole placed *after* the sample [30]. In this way, no assumption is made on the interaction of x rays with the sample and this approach can be extended to study dynamical diffraction effects. A similar configuration, using a large coherent beam, with the sample in the diffraction condition for strain sensitivity, has been recently shown in [31,32]. The results presented are corroborated by simulations based on dynamical diffraction theory. This Letter provides a very

the refinement of the pinhole is independent for the eight scans [41]. In the reconstruction, we use the pinhole obtained with a test specimen as an initial guess, see Fig. S4 of the Supplemental Material [34].

The teleptychography reconstructions return the amplitude and phase of the forward diffracted wave field and the transmitted beam at the pinhole position (cf. Fig. S5 of the Supplemental Material [34]). Figures 2(a) and 2(a') show the forward diffracted intensity (the square of the retrieved amplitude) from the strain-free sample out and in diffraction conditions, respectively. Echoes are indeed only seen in diffraction. These measurements reproduce previous results obtained with direct measurements [27], validating our approach. The divergence of the focused beam is not propagated through the echoes in the diffraction plane, but is only visible in the direction perpendicular to it. In the

horizontal plane, indeed, the crystal acts as an x-ray beam filter diffracting only the photons with a divergence comparable with the Darwin width of the selected diffraction peak, which in our case is of 100 nrad [18]. In the vertical plane, this filter effect does not apply [cf. Fig. 2(a')]. Nevertheless, the transmitted beam preserves the full divergence of the incoming beam, overlapping with the echoes at large distance from the sample. Figures 2(b) and 2(b') show the diffracted intensities of Figs. 2(a) and 2(a') at the sample plane, obtained by the propagation of the retrieved wave field [29,32]. The sample plane coincides with the location of the x-ray beam focus and, therefore, provides the best possible resolution of the forward diffracted field. Here, we note that there is no strict correspondence, in real space, between the position of the sample in the focus of the beam, its thickness, unintentionally

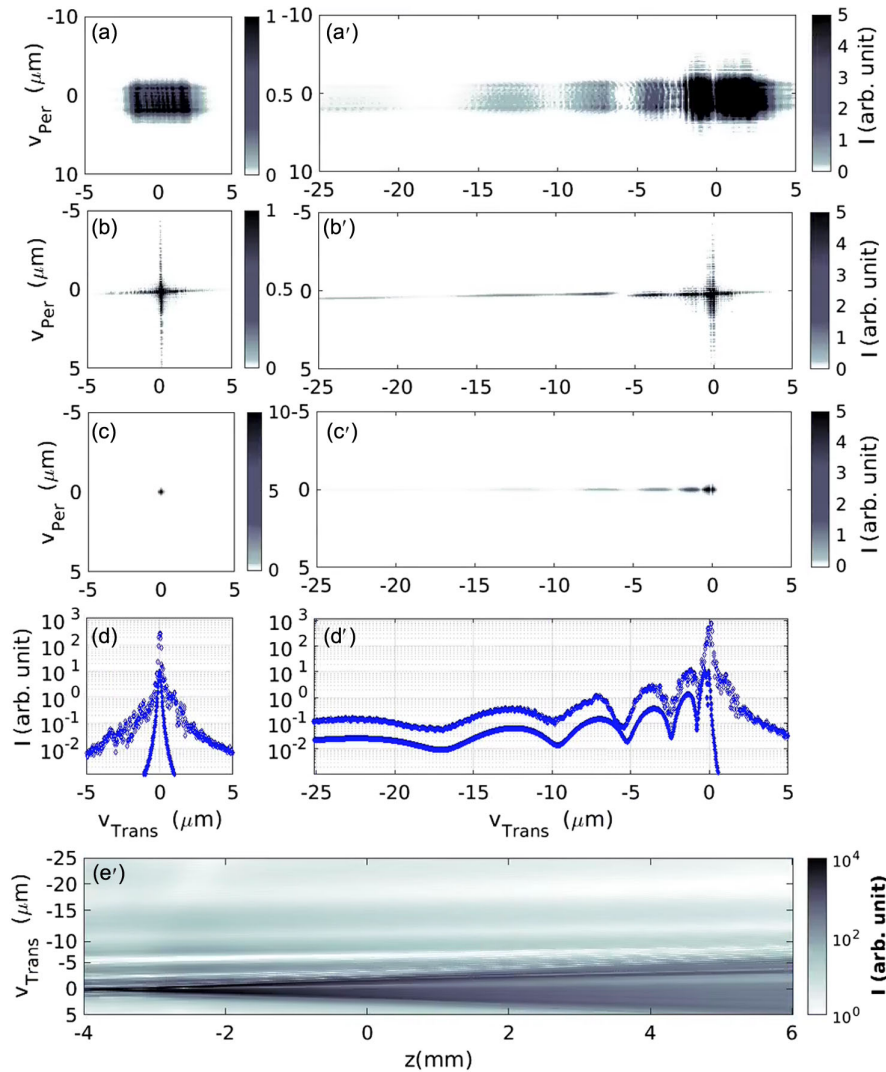


FIG. 2. Intensity of the FD wave front from strain-free Si wafer at two angles: (left) 1° away—echoes are absent—and (right) at the diffraction angle. (a),(a') Reconstruction at the pinhole plane. (b),(b') Reconstruction propagated to the KB focus. (c),(c') Simulation at the focus. (d),(d') Line cuts through (a),(a') and (b),(b'). The simulated signal, lower line, is offset for clarity. (e') Propagation of (a') along the beam direction z up to the sample position, illustrating the divergence of the transmitted beam and the parallel echoes.

comparable to the beam focal depth, and the propagation of the echoes in the focal plane. Indeed, the diffracted field measured is produced at the sample exit surface, and the backpropagation of the wave field beyond this point does not reflect any physical reality. The pixel size obtained for the reconstruction is 32 nm, while the resolution is estimated to be 55 nm, using Fourier ring correlation [42] for two different scans of the forward beam out of diffraction conditions (see Fig. S6 in the Supplemental Material [34]). Figures 2(c) and 2(c') show the simulation of the intensity of the forward diffracted field at the sample position for the configuration used, i.e., a 100 μm thick Si crystal at the asymmetric (111) reflection, using a horizontal diffraction geometry and a strain-free model, as described in [27]. The agreement with simulations is more clearly seen in the line cuts shown in Figs. 2(d) and 2(d'), where the data are offset for clarity. Figure 2(e') shows a map of the intensity of the echoes along the beam propagation direction. Because of the beam divergence, already at a distance of 2 mm from the sample surface, the echoes are overlapping with the divergent transmitted beam. Moreover, the echoes are parallel to the beam propagation direction, as expected.

From the indented sample, data are collected at different positions to compare the FD wave fronts produced by different strain fields. The dynamical diffraction goes far beyond the volume affected by the indent-induced strain. As a consequence, the echoes are still produced with a large contribution from the strain-free part of the Si crystal. Simulations of the expected echoes distribution are performed with our code [27,28] that we have adapted to model strained crystals following the work in Refs. [43–45]. The model assumes that the indent produces an isotropic strain field of amplitude proportional to the load, which decreases with distance from the indent following an inverse exponential law. Numerically this is achieved by slicing the crystal in 20 nm layers parallel to the surface, each with a different value of lattice parameter following the exponential decay. This numerical step, necessary to reduce the complexity of the calculations, approximates the radial to a linear decay. Despite the fact that the approximation introduces a variation of a factor between 1 and 2 of the effective decay length x rays experience along all possible paths defined by the dynamical diffraction process, it still provides good correspondence between data and simulations. We expect that improving the model to reflect a true radial decay will increase this correspondence. Finally, our model also disregards the strain anisotropy expected along different crystallographic directions, due to different elastic constants. This should be included in a future model.

Figures 3(a)–3(c) show the results from the inversion of the data collected, respectively, at 1 mm from the indented area and close to two nanoindents with loads 25 and 75 mN. The FD signal retrieved shows an increase of the number of echoes and a decrease of their mutual distance

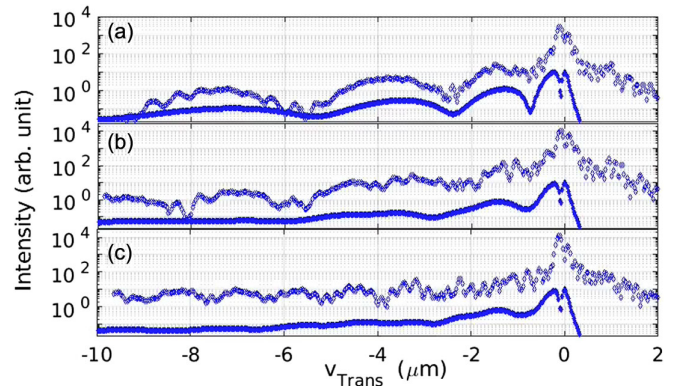


FIG. 3. Line cuts through the retrieved (empty dots) and simulated (filled dots) echoes from the indented Si sample in regions with different strain state, cf. text for details. Intensities are offset for clarity.

for increasing indentation strength. At higher strain, the echoes tend to merge into a continuum. Simulations of the dynamical diffraction signal using models with different strain fields provide a rather good agreement with data. Teleptychography reconstructions from this sample and the respective simulations are shown in Figs. S7–S10 of the Supplemental Material [34]. More accurate fitting routines will be implemented in the future. However, the position of the echoes are easily extracted via algorithms to find local maxima, as shown in Fig. S11. New echoes with shorter time delays arise in the strained areas, confirming this experimental approach to observe the strain-tailored FD maxima involved in this ultrafast process. Their time delay, visualized in Fig. S12 of the Supplemental Material, can be calculated using Eq. (1) and is shown in Fig. 4. The strain amplitude (expressed as a fractional variation of the lattice parameter) and the decay length of the exponential that best reproduces the data for the three regions (see Fig. S9 in Supplemental Material at [34]) are (i) 5×10^{-5} over 2 μm , Fig. 4(a), (ii) 10^{-4} over 7 μm , Fig. 4(b), and (iii) 10^{-4} over

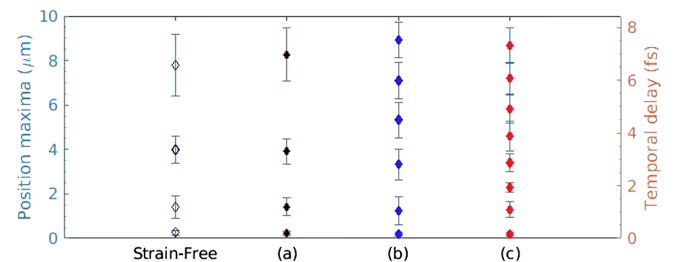


FIG. 4. (Left) Transverse displacement of the echoes along the first 8 μm of the reconstruction with respect to the transmitted beam as extracted from Fig. S11 in the Supplemental Material at [34] and (right) calculated delays from Eq. (1). For the strain-free sample and the indented sample at the positions (a), (b), and (c) as presented in Fig. 3. The bars correspond to the half width at half maximum of the respective echoes.

10 μm , Fig. 4(c). These findings are in agreement with results from literature [46].

These pioneering results demonstrate the extreme sensitivity of dynamical diffraction effects to localized strain fields of small amplitude. Echoes are produced from a volume extending throughout the full sample thickness, of which the strained area represents a mere 10%. Further improvement of the model will include the implementation of a radial decay of the strain field and eventually the crystal elastic anisotropy. A natural progression of this research is measuring echoes in the diffraction direction, where the spatiotemporal signal does not suffer from the absorption effects, as in FD. As presented in Fig. S13 in the Supplemental Material [34], the echoes probe the crystal in a more homogeneous manner, which allows one to retrieve more clearly the information from lattice deformations.

We have presented an experimental study of the dynamical diffraction produced by a Si wafer in transmission geometry, in the presence of a localized strain. We have shown that strain can be used to finely tune time delay of the echoes, in a way we are able to model. We achieve a high resolution in both sample and detection space with the use of a focused beam combined with a teleptychography approach. With the use of a nanobeam, we effectively increase the sensitivity of our measurement to strain gradients and sample heterogeneity by reducing the illuminated volume. Our approach provides a sensitive and efficient way to analyze the impact of strain of different amplitudes onto dynamical diffraction effects from perfect crystals. Conversely, understanding how dynamical diffraction is affected by the presence of strain in otherwise perfectly ordered crystals offers a new and visibly more sensitive tool for the characterization of strain and defects in crystalline materials. This Letter provides insights on the dynamical effects arising in coherent imaging experiments involving high-atomic-number crystals, such as Ni, Au, InSb, or CdTe, for which the extinction length reduces to the micron and submicron level, and contributes to the landscape of emerging experimental and analysis approaches developed to model them. It is important to mention that the presence from the echoes in the diffracted signal could be distorting the results in temporal ultrafast studies.

Finally, being able to predict and control the behavior of dynamical diffraction in the presence of strain opens the fascinating possibility of strain-tailoring ultrafast optics. This might include the generation of multibunch sources from single ultrafast pulses or the production of tailored crystals for split-and-delay lines with full control of the timing and width of the generated pulses with respect to the incoming pulse.

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