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Research Article

Moving the Frontier of Quantum Control into the Soft X-Ray Spectrum

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The femtosecond nature of X-ray free electron laser (FEL) pulses opens up exciting research possibilities in time-resolved studies including femtosecond photoemission and diffraction. The recent developments of seeding X-ray FELs extend their capabilities by creating stable, temporally coherent, and repeatable pulses. This in turn opens the possibility of spectral engineering soft X-ray pulses to use as a probe for the control of quantum dynamics. We propose a method for extending coherent control pulse-shaping techniques to the soft X-ray spectral range by using a reflective geometry 4f pulse shaper. This method is based on recent developments in asymmetrically cut multilayer optic technology and piezoelectric substrates.

The use of coherent control, or the selection of electronic dynamics by shaping the amplitude and phase of incident femtosecond laser fields, is well established in the infrared [1], visible [2], and ultraviolet [3] ranges of the electromagnetic spectrum. Visible laser light can be dynamically adjusted, with high precision, to enhance or enable specific chemical and physical processes. Currently quantum control experiments focus on the control of molecular motion, vibrational excitations [4], and to a limited degree, electron dynamics in the valence band [5]. This has been an enabling technology for many scientific studies from the selectivity of chemical reaction pathways [6, 7] to the manipulation of quantum dots [8].

With seeded X-ray FELs, the possibility now exists to extend the concept of coherent control to the shorter wavelengths of the soft X-ray spectrum. These shorter wavelengths correspond to core level electron transitions and allow for the probing of inner shell electron dynamics of complex systems.

As intense sources of coherent ionizing radiation are only just becoming available, little work has been performed, so far, in the area of coherent control for the specific pumping of core states or electron wave packets. However, developments in coherent control of the generation of high harmonics in the extreme ultraviolet (EUV) have demonstrated the ability to selectively break bonds and control the products of gas phase reactions [9].

The limits of existing soft X-ray pulse-shaping techniques motivate the need for the implementation a more robust pulse shaper, similar to those of existing 4f pulse shaper designs used in visible laser technology. The standard 4f design [2] consists of two gratings and two converging lenses of the same focal distance. The gratings are separated by 4 focal lengths, and the lenses are located in such a way that they create 3 focal planes: one at each grating and one located directly between the gratings. The focal plane (Fourier plane) between the lenses is a location where each frequency in the pulse is mapped into a spatial position. This allows the phase and amplitude of each frequency components to be modified independently, often by either a spatial light modulator or an acoustooptic modulator. The incoming pulse is dispersed off the first grating, and the dispersed light propagates to the first lens. The first lens then maps the frequency components of the pulse to a spatial location in the back focal plane where the components are modified in amplitude, phase, or both. The second, symmetric lens/grating pair is used to invert the process and reconstruct the pulse.

A direct extension of the optical components from visible techniques into the soft X-ray spectrum is not possible. Refractive lenses for soft X-rays do not exist; spatial light modulators and acoustooptic modulations would be exceedingly absorbing, and mechanically ruled X-ray gratings are inefficient and have very small dispersion angles producing long optical systems.

With recent developments in novel multilayer-based optics [10] and nanometer precision deformable substrates [11], it is possible to develop a soft X-ray 4f geometry pulse shaper.

Multilayer optics are based on a well-established technology in the EUV and soft X-ray communities. They are synthetic Bragg structures consisting of many layers of alternating materials in a bilayer configuration where the reflection from each interface adds coherently to the reflection from the interface above and below. They have been successfully used in experiments at the FLASH (Free electron LASer at Hamburg) [12] facility [13–17] where the energy density is large enough to destroy the multilayers [14, 18] as well as in experiments where the multilayer survives numerous pulses [16, 17, 19]. Multilayer optics are also used to chirp and shape high harmonic generated attosecond pulses [20–22]. However, chirped multilayers have limits in their abilities that exclude them for use in a coherent control of soft X-ray scheme. Multilayers are static structures and cannot dynamically change their chirp, limiting their capability to shape the pulses. More importantly a single EUV/soft X-ray multilayer mirror is limited in its ability to significantly lengthen or compress pulses larger than 10 femtoseconds. This is due to the fact that all materials in the EUV/Soft X-ray are absorbing and have attenuation lengths in the micron and submicron ranges producing a fundamental limit on the thickness of a multilayer structure. The maximum amount of lengthening or compression of a pulse that can be achieved upon a single reflection of a multilayer is approximately

$$\tau = \frac{Nd}{c}, \quad (1)$$

where τ is the number of femtoseconds in which a pulse is lengthened or compressed, N is the number of bilayers in the multilayer, d is the mean period thickness of the multilayer and c is the speed of light.

The implementation of a soft X-ray pulse shaper could employ an all-reflective folded geometry [23] using novel multilayered optics designed for the soft X-rays (Figure 1). It has been demonstrated that asymmetrically cut multilayers efficiently disperse X-rays [10] so they could be used as the dispersive element. A multilayer-coated normal incidence cylinder could be used as the focusing element. In the Fourier plane, a normal incidence multilayer with a variable thickness attenuation coating would allow for amplitude control and a piezoelectric substrate would allow for phase control.

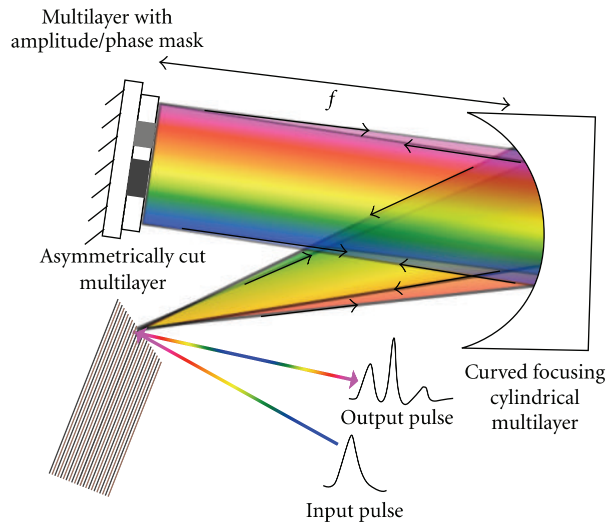

Figure 1: This is the proposed X-ray equivalent of the folded 4f geometry already demonstrated in the visible regime. It consists of a thick asymmetrically cut multilayer acting as a high-efficiency grating, a focusing multilayer mirror, a movable mask to shape the pulse and, a normal incidence multilayer to enable the folded 4f geometry.

In what follows each of these optics will be described in detail, excluding the focusing multilayer mirror as reflective multilayer optic coatings are a well-developed field [24].

Asymmetrically cut multilayers are based on thick multilayer structures that are cut along a bias to the multilayer stack (Figure 2). The first-order Bragg diffraction occurs in the direction of the central wavelength for the multilayer. These structures have similar dispersive properties as asymmetrically cut Bragg crystal monochromators [25]. The first-order grating efficiency of asymmetrically cut multilayers, which can be described by dynamical diffraction theory [10], is almost 100%. Another advantage of these devices is that the dispersion properties are controlled by the multilayer's d-spacing (period thickness) and the cut angle. Using d-spacing in the nanometer range produces a highly dispersive system creating reasonable-size optical elements for the optical system.

For dynamic pulse shaping control in the Fourier plane, a substitute for a spatial light modulator needs to be found. Amplitude control is relatively simple to achieve. One just needs to add structured absorbers of varying thickness to the top surface of the multilayer. However, this approach also has two drawbacks: the absorber layer adds a slight phase shift to the specific spectral component undergoing amplitude modulation and the structured surface is no longer dynamic. To overcome the second drawback, a moveable Fourier plane optic with numerous preprogrammed possible combinations of different patterned absorption amplitude masks is required. Phase control can be implemented by using a phase mask nanostructure underneath the multilayer. The height of the nanostructure changes the phase delay of spectral frequency component. This type of design has been implemented before to produce a π phase shift in

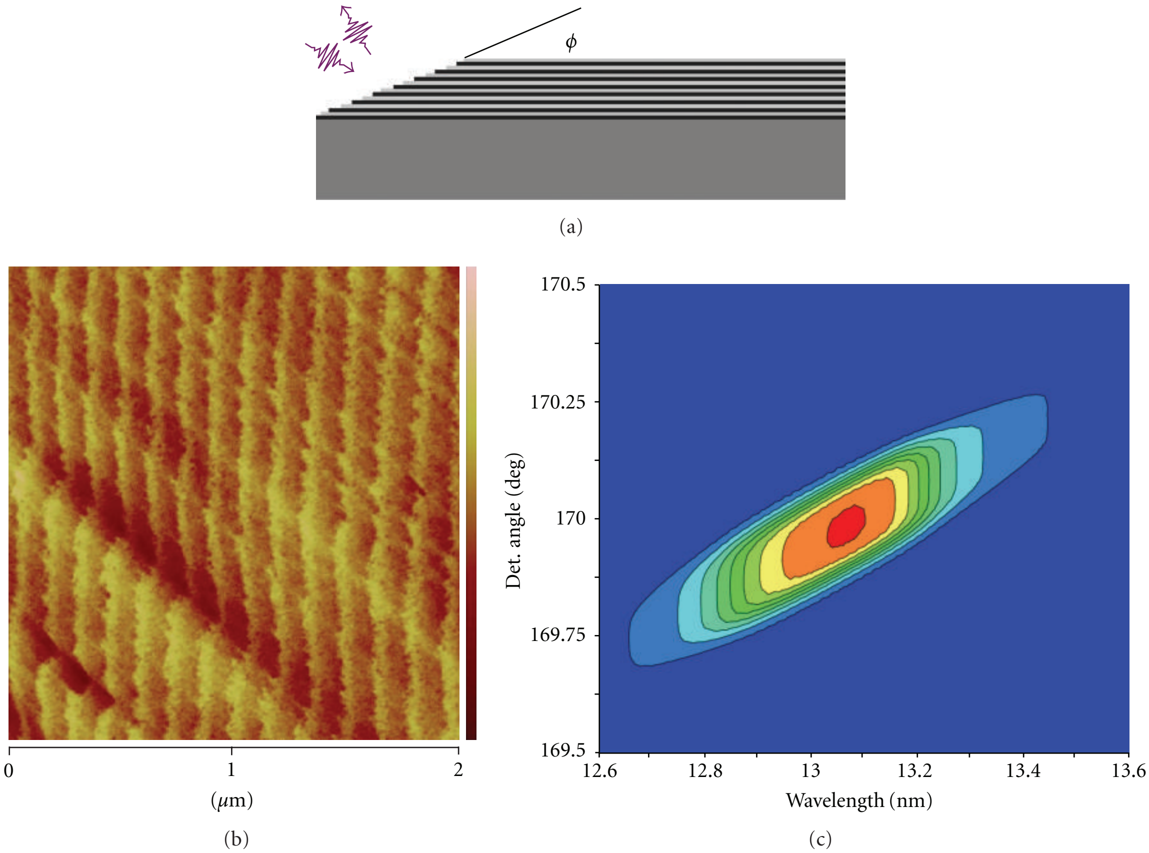

Figure 2: (a) A pictorial diagram of an asymmetrically cut multilayer with cut angle ϕ. (b) An atomic force micrograph of the surface of the asymmetrically cut multilayer. The periodic structure observed on the surface is due to the individual layers in the multilayer. These bilayers act as a high period grating. Also seen are striations left after polishing. As the asymmetrically cut multilayer is a volumetric grating, the surface imperfections have little effect on the overall efficiency and dispersion. (c) Reflectivity measurements of the dispersion from an asymmetrically cut multilayer conducted at beamline 6.3.2 at the Advanced Light Source. The multilayer cut angle ϕ was 5.4 degrees, and the measurement was conducted 5 degrees from normal of the top surface of the mirror. This particular asymmetrically cut multilayer corresponds to a grating d-spacing of ~70 nm and had >95% efficiency into the 1st order.

the use of a binary phase grating at EUV wavelength [26]. Recent developments in dynamically controlled piezoelectric substrates for hard X-ray nanofocusing optics [11] can be implemented for full dynamic control of the phase.

Seeded X-ray FELs offer new capabilities but also requirements for further development of X-ray optics. We have conceived the concept and design of a 4f pulse shaper for the quantum control of core level electrons. "Proof-of-principle" measurements of the design at soft X-ray wavelengths are currently underway.

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