Femtosecond Time-Delay Holography

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One of the pressing questions about the high-resolution XFEL imaging and characterization of non-periodic or weakly-scattering objects is the effect of the intense FEL pulse on the object, during the interaction with that pulse. The method of single-particle diffraction imaging [1] requires a stream of reproducible particles (e.g. a protein complex or virus) inserted into the beam, whereby a coherent X-ray diffraction pattern is recorded. The pulse will completely destroy the object, but if the pulse is short enough the diffraction pattern will represent the undamaged object. Single-pulse diffractive imaging at FLASH shows no evidence of damage during the 10–30 fs pulses at spatial resolutions of 10's of nanometres [2]. We wished to dramatically increase our sensitivity to the particles' explosions, to be able to increase the understanding of the dynamics of particles irradiated by FEL pulses. This was done in two ways in a single experiment: by holographically measuring the time evolution of the particle at times after the pulse had pass through the object; and by making an interferometric measurement of the change in the optical path through the object. The experimental technique, time-delay holography [3], achieved a time resolution better than 3 fs, and a phase sensitivity of better than 3°, or a sensitivity of < 3 nm of the expansion of the particles.

The idea behind time-delay holography is to use the same pulse that initiated the interaction with an object to probe that object at a later time. This was achieved simply by placing a mirror behind the object to send the pulse back on the object a second time. The time delay is easily set by the distance between the mirror and the object. This geometry is in fact the same as the `dusty-mirror' experiment that was first carried out by Newton. Our experimental geometry is shown in Fig. 1. Key to this experiment is the multilayer backing mirror. Both the direct beam and the wide-angle scattered wave from the object reflect from this mirror, which must have high reflectivity over a large angular range. The reflected direct beam, on hitting the object for a second time, creates another scattered wave. This wave, however, carries structural information about the object at a later time than the initial interaction. If the object had expanded in this time, for example, the second diffracted wave would be more forward-peaked. The two scattered waves (the first from the

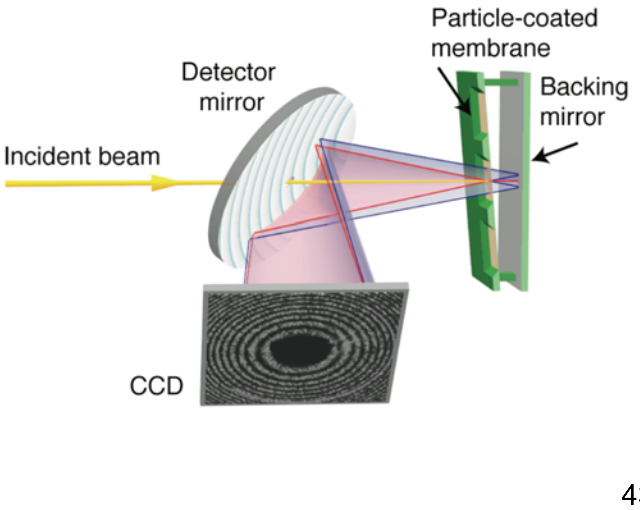

Fig. 1: X-rays from the FLASH FEL are focused on to a 'dusty mirror' consisting of particles on a membrane sandwiched closely to a multilayer backing mirror. An ultrashort pulse hits the particles twice: on the way in and after reflecting from the mirror. The two scattered waves interfere on the CCD to form a hologram that encodes the change in the particle in the brief time that the light was reflected back.

undamaged object, the second a well-defined time later) propagate together and interfere at a CCD. This interference is dominated by a distinct ring pattern, which is the interference of two spherical waves that are longitudinally displaced. With many objects placed in the beam, each one generates an interference pattern which then coherent adds with all others, modulating the ring pattern with a speckle pattern. The ring pattern accurately encodes the distance between the spherical waves, which can be determined to high accuracy. From this distance the delay between the events can be determined to better than 3 fs. We consider the recorded diffraction pattern as a hologram, as it consists of the interference of a known reference wave (the undamaged object) with an unknown object wave (the object undergoing an explosion).

The sample in our experiments consisted of an array of hundreds of silicon nitride membrane windows on which 140-nm diameter polystyrene spheres were placed. This was sandwiched against the multilayer backing mirror, positioned with a slight wedge to be able to vary the delay simply by moving to different membrane windows. The smallest gap was about 50 micron, to give a delay of about 300 fs, and the longest gap gave a delay of 8 ps. Each window gave the opportunity for several separate exposures. Even though the 20-micron-wide focused FEL pulse ablated a crater in the backing mirror and melted a hole in the silicon nitride, the entire window did not shatter so we could simply move to a new spot. The time-delay holograms of these samples were initially analyzed by considering the intensity of the interference between these waves as a function of scattering angle, or momentum transfer q. This was compared with calculated patterns, using a hydrodynamic model [4]. We see that, as predicted, the patterns become narrower and more forward peaked as the time delay is increased beyond 3.8 ps. This suggests that in real space the objects are indeed expanding following the interaction. Furthermore, the evolution of the structure factor of the polystyrene sphere is in good agreement with our calculations to the longest-measured delays of 8 ps where the sphere has approximately doubled in size.

At delays shorter than 3 ps the expansion of the sphere was less than the transverse spatial resolution of 60 nm and hence could not be observed. However, changes in the optical properties of the sphere could be observed for delays even shorter than 1 ps. This is due to the interferometric nature of the measurement. The wave scattering from a polystyrene sphere is phase shifted by an amount that depends on the sphere's refractive index and (at low q) its thickness. If these properties change by the time the pulse returns to the sphere then the phase shift on scattering the second time will be different to the first. This relative phase shift will cause a change in the ring fringe pattern of the hologram. For example, a change in the phase shift by p (half a wave) would reverse the contrast of the fringes by causing constructive interference where there would have been destructive interference. The relative phase can thus be determined from the positions of the ring maxima and minima. We observed an increase in the phase shift with time delay and for increasing pulse fluence. At a delay of 350 fs the change in phase shift was equivalent to an increase of the optical path by less than 1/50th of a wavelength. Since the refractive index is negative (and close to unity) this corresponds to a reduction of material projected through the ball as would occur if the sphere expanded in all directions by 3 nm.

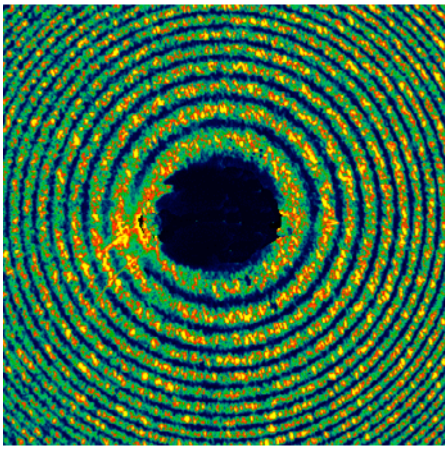

Fig. 2: A time-delay hologram recorded for a delay of 348 fs for 140-nm diameter polystyrene spheres. The ring pattern encodes both the delay and the change in phase of the pulse as it travels through the exploding object. Holograms recorded at different delays reveal the FEL-induced explosion of the spheres.

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