Working Group I Subgroup Summary Report:
X-ray Photon Correlation Spectroscopy at LCLS/G4

S. Mochrie and L. Sorensen, MIT

(1) Initial experiments at the APS reveal that it is now understood in quite some detail what to expect from XPCS experiments and therefore what is required for such experiments.

(2) XPCS requires both partial transverse (spatial) coherence and partial longitudinal (temporal) coherence. Specifically, the sample should be illuminated by no more than a few transverse coherence lengths. In addition, for experiments carried out in a transmission geometry, the requirement of partial longitudinal coherence requires (a) that $\Delta\lambda/\lambda QL\sqrt{1-Q^2/k}$ be a few or less, and (b) that $\Delta\lambda/\lambda Q^2 W/k$ be a few or less, where $\Delta\lambda/\lambda$ is the relative bandwidth, W is the sample thickness, L is the dimension across the beam, Q is the scattering vector, and k is the wavevector of the incident x-rays. Here, we assume that $k = 40$ nm^{-1}, that $L = 100$ μm, equal to the transverse coherence length of the beam, and that $W = 0.3$ mm. Evidently, the required bandwidth depends on the scale of the phenonmena to be investigated. Suppose that $Q = 5$ nm^{-1} – corresponding to a length scale of 1.2 nm – then these criteria require $\Delta\lambda/\lambda \cong 10^{-6}$. (The criteria for a reflection geometry would be different and have not yet been fully explored.)

(3) In any case, if we further assume a pixelated detector with 20 μm pixels, then the detector, or preferably detector array, should be located 20 m or more from the sample. This is because the angular extent of speckles from a sample illuminated by a beam $L \cong 100$ μm across is on the order of λ/L, and because the detector should be able to resolve speckle. The scattering angle corresponding to a Q of 5 nm^{-1} with $k = 40$ nm^{-1} is on the order of 6 degrees. For $Q = 10$ nm^{-1}, it would be 12 degrees. The hutch footprint of an XPCS station should allow for scattering measurements with a 20 m sample-to-detector distance with these sorts of scattering angles. Even larger scattering angles will be required if fluctuations near a Bragg peak are to be studied in a reflection geometry.

(4) The scheme for carrying out XPCS measurements at LCLS/G4 that we will describe depends on being able to determine the contrast of a speckle pattern obtained in a single pulse. Theory and experiments at the APS have shown that the standard deviation of the speckle

contrast is given by $1/(\bar{n}N_p)$, where $\bar{n}$ is the mean number of photons per pixel and N_p is the number of pixel pairs used to calculate the intensity-intensity correlation function. To calculate $\bar{n}$, we will assume a scattering cross-section per unit volume of 3×10^{-2} cm^{-1}, a solid angle per pixel of 10^{-12} steradians, and that there are $n_0 = 10^{12}$ photons per bunch in a bandwidth of 10^{-6}. Then, $\bar{n} = n_0 \Sigma W \Delta\Omega = 10^{-4}$ x-rays per pixel per pulse, assuming the detection efficiency in the CCD is 100%. Further taking $N_p = 10^6$, we find that the standard deviation of the speckle contrast from a single pulse is 1. We estimate that the speckle contrast itself is 0.5, so that the signal-to-noise (*S/N*) from a one pulse experiment is 0.5. Running at 120 Hz for 10 minutes yields *S/N* ≅ 100. If one can collect from 10^7 or 10^8 pixels, the advantage is clear and mandates a major detector development effort. In fact, improved detectors would be of major utility at third generation sources.

(5) In order to measure time correlations, it is essential to probe the sample with two pulses separated by some time delay τ. The range over which τ can be varied is the range over which the sample dynamics can be probed. Ideally, τ would be varied from 10^{-12} s to 10^{-6} s or longer. If two pulses with a variable separation, or a single pulse with a variable pulse length cannot be produced we do not see how XPCS can be realized at LCLS/G4. In our scheme, we suppose there are two pulses and the x-ray scattered by both pulses are detected in one CCD exposure. Then, for small τ, both pulses sample the same configuration of the sample, so that the observed contrast in the CCD is the contrast of a single speckle pattern. For large τ, by contrast, each pulse will sample two different configurations of the sample, so that the observed contrast in the CCD is the contrast of two independent speckle patterns. This is one half the contrast of a single speckle pattern. Thus, as one varies τ the observed speckle contrast will vary from one value to one-half of that value. The time at which the crossover occurs is the characteristic relaxation time of the sample under study at the Q in question.