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Establishing nonlinearity thresholds with ultraintense X-ray pulses

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X-rays have long been used to explore the electronic and structural properties of all forms of matter, using sources as varied as X-ray tubes to accelerator-based storage rings. X-ray methods have evolved over decades to become specialized tools for a broad range of investigations, with techniques ranging from X-ray scattering through X-ray spectroscopy to X-ray tomography. In general these methods all rely on X-ray measurements that depend linearly on the number of incident X-ray photons. With the advent of X-ray free electron lasers (XFELs), the ability to reach extremely high photon numbers in ultrashort pulse durations has resulted in a paradigm shift in our ability to observe nonlinear X-ray signals. This enormous increase in peak power (pulse energy/pulse duration) has been a doubleedged sword, with new and exciting techniques being developed but at the same time well-established techniques proving unreliable [1-3]. This requires a fundamental change in our approach to X-ray science at FELs, since this nonlinear regime is a largely unexplored area, making it hard to predict not only when to expect nonlinear contributions to a measurement, but also to understand the very nature of this response [4, 5].

We report an X-ray spectroscopic study that reveals important details on the thresholds for nonlinear X-ray interactions. By varying both the incident X-ray intensity and photon energy, we establish the regimes at which the simplest nonlinear process, two-photon X-ray absorption (TPA), can be observed. From these measurements we can extract the probability of this process as a function of photon energy as well as the sub-femtosecond lifetimes of the intermediate electronic states, allowing us to develop an analytical equation that predicts the efficiency of the TPA process on the basis of straightforward linear X-ray measurements. This result is the first step towards allowing scientists to perform XFEL measurements with some degree of certainty as to whether they are in the linear or nonlinear X-ray regime.

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