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X-ray and optical wave mixing

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Light-matter interactions have advanced our understanding of atoms, molecules, and materials while also being central to a number of applied areas. Though optical interactions have been heavily studied, their microscopic details are often poorly understood. To date it has not been possible to directly probe the microscopic details of light-matter interactions. X-ray and optical wave mixing, specifically sum frequency generation, was proposed nearly a half century ago as an atomic-scale probe of light-matter interactions. The process is, in essence, optically modulated x-ray diffraction : x-rays inelastically scatter from optically induced charge oscillations and probe optically polarized charge in direct analogy to how conventional x-ray diffraction probes ground-state charge. Here we use an x-ray free electron laser to demonstrate x-ray/optical sum frequency generation through nonlinear interaction of the two fields in single crystal diamond. Optically modulated x-ray diffraction from the (111) planes generates a sum (x-ray + optical) frequency pulse. The measured conversion efficiency ($\sim 10^{-7}$) determines the (111) Fourier components of the optically induced charge and associated microscopic field that arise in the illuminated sample. To within experimental error bars the measured charge density is consistent with first principles calculations of microscopic optical polarizability in diamond. The measurements and calculations indicate that light predominantly perturbs chemical bonds in the diamond lattice. This finding should be generally applicable to covalent semiconductors and closely related materials such as graphene. A simple bond charge model reproduces the measured charge density to within $\sim 50\%$, suggesting that these models can provide reasonably accurate estimates of microscopic optical polarizability in, for instance, photonic and photovoltaic devices based on silicon. The ability to measure atomic-scale charges and fields induced by light should contribute to a better understanding of materials while also creating new ways to study phototriggered dynamics.