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Non-linear optical techniques and optical properties of condensed molecular systems

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Structure, dynamics, and optical properties of molecular systems can be largely modified by the applied pressure, with remarkable consequences on their chemical stability. Several examples of selective reactions yielding technologically attractive products can be cited, which are particularly efficient when photochemical effects are exploited in conjunction with the structural conditions attained at high density. Non-linear optical techniques are a basic tool to unveil key aspects of the chemical reactivity and dynamic properties of molecules. Their application to high-pressure samples is experimentally challenging, mainly because of the small sample dimensions and of the non-linear effects generated in the anvil materials. In this talk I will present results on the electronic spectra of several aromatic crystals obtained through two-photon induced fluorescence and two-photon excitation profiles measured as a function of pressure (typically up to about 25 GPa), and discuss the relationship between the pressure-induced modifications of the electronic structure and the chemical reactivity at high pressure. I will also present the first successful pump-probe infrared measurement performed as a function of pressure on a condensed molecular system. The system under examination is liquid water, in a sapphire anvil cell, up to 1 GPa along isotherms at 298 and 363 K. These measurements give a new enlightening insight into the dynamical properties of low- and high-density water allowing a definition of the two structures.