Nonlinear x-ray physics and future x-ray metrology

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Nonlinear optics is the field of optical lasers since the past 50 years. Nonlinear effects arise when the optical properties of the medium are modified due to the presence of photons. The key quantity that determines these effects is the photon degeneracy, i.e., the number of photons per mode of the radiation field. This number can assume values of 10^9 for gas lasers and up to 10^{14} for mode-locked or Q-switched lasers. At such high photon numbers the light of frequency modulates the microscopic charge distribution, leading to phenomena like high-harmonic generation, frequency mixing, up- and down-conversion and more.

An interesting scenario arises if the sample is illuminated by two laser beams, the frequency difference of which corresponds to the frequency of an excitation within the system. In this case, stimulated emission at the difference frequency is observed (stimulated Raman scattering) that allows for an extremely high sensitivity in probing excitations in molecular systems or the identification of species particular vibrational modes that form transiently during a sequence of nonequilibrium processes. It appears to be extremely intriguing to extend these concepts into the x-ray regime [1].

Indeed, the advent of new radiation sources, especially free-electron lasers, opens the field of nonlinear optics at x-ray energies. While the photon degeneracy barely reaches 0.1 at existing third-generation synchrotron sources, it may reach up to 10^9 at x-ray laser sources. In combination with the outstanding time resolution of these laser sources, x-ray nonlinear spectroscopies might be applied to monitor fast transient processes and intermediate states that are encountered in environments that are not accessible to optical radiation. Examples are phase change dynamics in correlated electron materials or dynamical processes in tribological systems like bond formation and bond breaking between two interfaces in frictional contact.

Another nonlinear effect bears interesting applications in the field of x-ray quantum optics: The process of parametric down conversion (PDC) leads to the emission of a pair of photons in an entangled state, i.e., a state that cannot be written as a simple product of photon state and field mode. While PDC has been demonstrated at x-ray energies [2], entanglement of photons at x-ray energies remains to be shown [3]. Applications of such entangled states could be tests of the nonlocality of quantum mechanics via Bell's theorem. An interesting question to investigate is how pairs of entangled photons generated in sufficiently high intensity could be employed for a new type of x-ray absorption measurement and spectroscopy [4].

A central paradigm in the field of quantum optics is the two-level atom. In the optical regime the sharpest atomic resonances are realized in ultracold quantum gases. In the x-ray regime extremely narrow linewidths are observed for nuclear resonances. The hyperfine interaction of these two-level systems with their environment can be rather well controlled and almost the natural linewidth is observed in solid state samples. Resonant (Mössbauer) nuclei can be employed to probe fundamental quantum optical effects like single-photon superradiance and the collective Lamb shift [5] as recently has been demonstrated [6]. These effects result from the interaction of a large number of identical atoms with a common resonant radiation field. Cooperative effects at x-ray energies have so far been explored in the single-photon regime. It thus appears to be very appealing to investigate such effects in the multi-photon regime.

The interaction of many identical resonant atoms with a radiation field plays an important role for fast radiative energy transfer as it occurs in light-harvesting systems in photobiology. Systems of identical resonant atoms can thus serve as a model system to explore these effects in a systematic fashion, as they are crucially dependent on the geometric arrangement of the resonant atoms.

Nonlinear optics, however, is not the domain of extremely high photon numbers only. Requirement for nonlinear optics at low photon numbers are materials with optical properties that are significantly altered even by single photons. When this alteration persists long enough to be experienced by photons that subsequently interact with the material, large nonlinearities can be expected. Nuclear resonances with their lifetimes in the range between picoseconds and nanoseconds are interesting candidates in this field. Alternatively, nonlinear optical effects at low light levels can be reached with resonant atoms placed in a high-finesse, small-volume cavity. It has to be explored under which conditions this can be achieved at energies of hard x-rays.

Recently, laser based spectroscopy opened unique possibilities for ultrahigh precision measurements of physical quantities by application of the frequency comb technique [7]. One aim of this focus panel is to explore how these concepts can be pushed to shorter and shorter wavelengths. The ultimate goal would be the increase of the precision of the frequency comb technique into a regime where one could probe the temporal variation of fundamental physical constants as postulated within certain cosmological models.

An important requisite that is indispensable for future studies outlined above is multicolour x-ray laser radiation with a well-established control of longitudinal coherence, i.e., monochromaticity, that can be achieved by seeding [8] or XFEL-oscillator schemes [9]. Under such conditions a precise coherent control of atomic and nuclear transitions becomes possible that forms the basis of nonlinear and quantum-optical effects in the x-ray regime.

References

[1] B. D. Patterson, SLAC Technical Note, SLAC-TN-10-026
[2] P. Eisenberger and S. McCall, Phys. Rev. Lett. **26**, 684 (1971), Y. Yoda et al., J. Synchrotron Rad. **5**, 980 (1998), B. W. Adams et al., J. Synchrotron Rad. **7**, 81 (2000).
[3] A. Palffy, J. Evers and C. H. Keitel, Phys. Rev. Lett. **103**, 017401 (2009).
[4] Z. Zhao et al., Anal. Chem. **80**, 7635-7638 (2008), B. W. Adams, this conference
[5] M. O. Scully, Phys. Rev. Lett. **102**, 143601 (2009)
[6] R. Röhlsberger, K. Schlage, B. Sahoo, S. Couet and R. Rüffer, Science 328, 1248 (2010)
[7] S. T. Cundiff and J. Ye, Rev. Mod. Phys. 75, 325 (2003)
[8] G. Geloni, V. Kocharyan, and E. Saldin, DESY Internal Report DESY 10-133
[9] Kwang-Je Kim, Yuri Shvyd'ko, and Sven Reiche, Phys. Rev. Lett. **100**, 244802 (2008)