Introduction

Traditionally, the interaction of matter with light is exploited to gain a detailed understanding of the quantum mechanical foundations of atomic, molecular, ionic and nuclear systems. While in particular at the most fundamental level this continues to be a challenge, by now it is nevertheless possible both in theory and in experiment to deliberately control the quantum dynamics of many systems of interest. This on the one hand opens perspectives for a multitude of fascinating applications. On the other hand, it provides a handle to probe the validity range of theoretical models and intuitive interpretations, and to explore systems far away from their equilibrium properties.

In this overview, we outline some of our recent approaches towards the active control of quantum dynamics and material properties. The target systems range from visible frequencies with atoms near their ground state up to the hard X-ray domain of nuclear excitations. Interestingly, the fundamental mechanisms based on coherence, non-linear interactions and quantum effects underlying the control of the dynamics are often shared across the different platforms. In this sense, the various approaches complement each other, which together with the fruitful interplay of theory and experiment enable us to extract the fundamental principles of laser control.

Fano Control of Light-Matter Interaction

Spectroscopy is a primary tool to investigate structure and dynamics of physical systems. The archetype spectroscopic signature is a symmetric Lorentzian line shape, which is observed if the probing radiation is near-resonant to an isolated bound-state resonance in the system. In contrast, excitation of a continuum of resonances leads to a structure-less spectral response. More generally, a broad class of asymmetric line shapes arises, if an isolated resonance couples to a continuum of resonances. These Fano line shapes were first characterized by Ugo Fano to theoretically model the ionization of Helium by electron impact, which can either proceed directly into a continuum of final states, or via an auto-ionizing isolated bound-state resonance, that subsequently decays to the same final state.

Fano resonances, the consequence of a fundamental coupling process, play an important role in a multitude of quantum processes and applications across the natural sciences.

In a joint collaboration of experimental and theoretical teams at the institute [1], we have developed a new interpretation of Fano resonance absorption in the time domain, which opens a direct route to the external manipulation of the spectral response. In this picture, the detected signal arises from the interference between the incident probing radiation, and the radiation emitted by the sample’s time-dependent oscillating dipole moment excited by the probing field. A spectral continuum translates into a short peak in the temporal response, while an isolated resonance leads to a slower exponential decay of the oscillating dipole moment. The key result of the new interpretation is the observation that the coupling between continuum and isolated bound state resonance responsible for the asymmetric Fano line shapes leads to a relative phase shift between the dipole response and the incident radiation. This phase shift changes the interference between the two contributions, and thereby modifies the observed line shape. In particular, an intuitive analytic one-to-one
Control of Cooperative Dynamics in Atoms and Nuclei

The success story of optical lasers is founded on coherence, non-linearities and quantum effects. Recent advances in existing and upcoming light sources provide access to laser-like light in the X-ray and gamma-ray frequency domain, with a multitude of novel applications across all the natural sciences. However, it remains a challenge to exploit quantum optical phenomena in the X-ray domain, which will be required to unleash the full potential of the new light sources. Motivated by this, we develop quantum optical methods in the hard X-ray domain. A particularly promising platform is X-ray cavity quantum electrodynamics (QED) with Mössbauer nuclei embedded in thin-film cavities probed by near-resonant X-ray light, as used in a number of recent experiments. We develop a quantum optical framework for the description of this setting. It allows identifying and separating all physical processes contributing to the recorded signal, and encompasses nonlinear and quantum effects. Based on this model, the light polarization and an external magnetic field can be used to control the nuclear level structure.

An important step towards X-ray quantum optics is the generation of coherent superpositions of quantum states. These were crucial in the development of quantum mechanics, as exemplified by the famous thought experiment about the fate of Schrödinger’s cat. Nowadays, such coherent superpositions are key resources for quantum technologies. But in experiments, usually inevitable decoherence destroys the superposition states, restricting many applications. An important source of decoherence is spontaneous emission, induced by the interaction with the surrounding vacuum field. We have developed a method to manipulate the interaction with the vacuum in such a way that it produces the desired superpositions instead of destroying them [2]. These spontaneously generated coherences (SGCs) are realized based on two mechanisms. First, the cooperative acceleration of the nuclear dynamics is exploited. Second, the vacuum cavity field experienced by the nuclei is tailored suitably via the external magnetic field.

As a result of this phase control, the experiments clearly demonstrated the conversion of asymmetric Fano line shapes naturally occurring in doubly-excited helium into symmetric Fano line shapes. The broader impact of these results also includes a complementary interpretation of related phenomena such as electromagnetically induced transparency or laser without inversion. Future experiments could exploit that the change in absorption is directly linked to the phase shift. This gives access to phase information, which together with the possibility to manipulate the coupling between different quantum mechanical states provides a handle to control and interpret correlations and dynamics in more complicated settings. Since the time-domain analysis and the phase-control of Fano line shapes are fundamental concepts, the developed methods can be generalized to other target systems and spectral regions, including the hard X-ray or gamma-ray domain.

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gap is formed, and certain frequencies cannot propagate through the medium. In collaboration with the group around S. Y. Zhu (Beijing Computational Science Research Center, China), we showed that a detuning of the two light fields forming the standing wave leads to a motion of the light-induced photonic crystal-like structure [4]. Due to the Doppler shift, light propagating along this movement or in reverse direction through the atoms experiences different optical properties. This breaks the time reversal symmetry, such that light is emitted from one side to the other, but not in reverse direction. This optical dive was demonstrated in a proof-of-principle experiment in the group of J.-X. Zhang (State Key Laboratory of Quantum Optics and Quantum Optics Devices, Taiyuan, China).

Phoebe Evers

Phase Control in High-Frequency Nonlinear Optics

The control of matter by phase-shaped electromagnetic radiation is now routinely achieved from radio frequencies up into the visible domain, enabling access to quantum dynamics from nuclear spins via rotations of large and small molecules to vibrational dynamics of individual molecular bonds and some electronic states. However, to control matter on a fundamental and comprehensive level — involving all electrons in low-lying bound states or excitations within atomic nuclei — high-frequency radiation needs to be shaped in amplitude and phase.

In our experiments, we study the elementary building blocks of phase control in high-frequency light. We focus on the highly-nonlinear optical process of high-harmonic generation converting visible laser light and its coherence properties into the extreme ultraviolet (XUV) regime. One experimental challenge is the stabilization of the driving visible laser field to enable complete transfer of the femtosecond driver pulse through the strongly-nonlinear recollision process into the spectral phase of the produced high-order attosecond-pulsed harmonic light.

The phase of the XUV harmonic light depends on the exact shape of the driver field, which creates essentially two non-local components separated by the split mirror. An intuitive time-domain interpretation was found for this phenomenon: The time-domain beating of the two-color subfields resulted in a double pulse with a temporal spacing given by the inverse frequency difference of the subfields. Spectral interference between the corresponding two temporally separated attosecond pulse trains resulted in the sub-harmonic modulation.

Controlling the CEP from 0 to $\pi$ allowed us to fully spectrally tune the sub-harmonic modulation from peak to peak. This provides evidence of spectral phase control between two attosecond pulse trains all the way from 0 to $2\pi$, thus achieving a substantial goal of attosecond high-frequency (XUV) control of coherent light.

Philipp Raith, Thomas Pfeifer

Storing and Manipulating X-ray Photons

Optical lasers have been at the heart of quantum physics for the last decades. The commissioning of the first X-ray free electron laser in the atomic, crystal first X-ray optics elements bring into play higher photon frequencies and promote the emerging field of X-ray quantum optics. Compared to optical photons, X-rays are not plagued by the diffraction limit and can be much better focused. Thus, focusing optics and quantum information to shorter wavelengths in the X-ray domain opens the potential of shrinking computing elements in future photonic devices using photons as the information carriers.

Prior to the realization of short-wavelength quantum photonic circuits, mastery of X-ray optics and powerful control tools of single-photon wave packet amplitude, frequency, polarization, and phase are required. In particular, efficient phase-sensitive photon storage and phase modulation, preferably for single-photon wave packets, are desirable but so far very challenging. A first successful scheme using nuclei as the "cage" to store single X-ray photons was designed theoretically in Ref. [7] exploiting a nuclear forward scattering setup. Employing nuclei to tame X-rays has here a two-fold advantage. First, due to their suitable transition frequencies, nuclei rise as promising candidates for X-ray quantum optics studies. Second, when using a solid-state sample with Mössbauer nuclei, the delocalized nature of the excitation leads to decohererent effects. These in turn open the possibility to control the re-emission of the single photon by external magnetic fields responsible for the nuclear hyperfine splitting.

The typical nuclear forward scattering setup involves a solid-state target containing $^{57}$Fe Mössbauer nuclei. A synchrotron radiation pulse tuned on the 144 keV nuclear transition from the ground state to the first excited state shines perpendicularly to the nuclear sample (see Fig. 5). Typically, given the small coupling constant, at most one nucleus is excited per pulse. In coherent scattering, the absence of nuclear recoil or spin flip does not allow identification of this nucleus, leading to a delocalized excitation often denoted as nuclear excitation. The key to control the excitation decay relies on an externally applied magnetic field inducing hyperfine splitting of the nuclear states. Due to the broadband nature of the incident pulse, several nuclear transitions are addressed simultaneously. The scattering process and resulting hyperfine transitions caused by inducing interference effects between these transitions. Switching off the hyperfine magnetic field at specific times will render the transition currents interfere destructively, strongly suppressing the nuclear decay and leading to coherent storage of the excitation for time intervals of 10-100 ns. Storing on the magnetic field will release the stored single photon and by using a releasing hyperfine magnetic field oriented in the opposite direction to the initial one, a phase modulation of $\pi$ can be achieved for the X-ray [7]. With coherent storage and phase modulation as powerful means to control single X-ray photons, an important missing link between nuclei, photonic devices and photon qubits is established.

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Fig. 5: (a) Relevant energy level scheme of $^{57}$Fe Mössbauer nuclei. A synchrotron radiation pulse tuned on the 144 keV nuclear transition from the ground state to the first excited state shines perpendicularly to the nuclear sample (see Fig. 5). Typically, given the small coupling constant, at most one nucleus is excited per pulse. In coherent scattering, the absence of nuclear recoil or spin flip does not allow identification of this nucleus, leading to a delocalized excitation often denoted as nuclear excitation. The key to control the excitation decay relies on an externally applied magnetic field inducing hyperfine splitting of the nuclear states. Due to the broadband nature of the incident pulse, several nuclear transitions are addressed simultaneously. The scattering process and resulting hyperfine transitions caused by inducing interference effects between these transitions. Switching off the hyperfine magnetic field at specific times will render the transition currents interfere destructively, strongly suppressing the nuclear decay and leading to coherent storage of the excitation for time intervals of 10-100 ns. Storing on the magnetic field will release the stored single photon and by using a releasing hyperfine magnetic field oriented in the opposite direction to the initial one, a phase modulation of $\pi$ can be achieved for the X-ray [7]. With coherent storage and phase modulation as powerful means to control single X-ray photons, an important missing link between nuclei, photonic devices and photon qubits is established.

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