I-T Nonlinear Processes Induced by Ultrafast and Intense Extreme Ultraviolet (XUV) Pulses

Recent technological progress on the generation of intense XUV pulses has opened up a new field on ultrafast and nonlinear optics. At such short wavelength only a gas can be a nonlinear medium. Nonlinear optics in XUV regions is interesting for two reasons. The first one is that the attosecond pulse generation is possible only in the XUV region, simply because an optical cycle of the visible-UV photon is in the time scale of femtosecond. The second one is that nonlinear response of the medium is not yet known at the XUV region. In this project, we have carried out investigation from those two aspects.

I-T-1 Time-Frequency Analysis of High-Order Harmonic Generation

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Recently it has been demonstrated by the group of Krausz (Austria) that it is possible to generate a single attosecond XUV pulse through high-order harmonic generation. Still, there are many things to be understood and clarified on the mechanism and conditions for the attosecond pulse generation. By solving time-dependent Schrödinger equations, we carry out the time-frequency analysis to obtain the temporal profile for each order of harmonic pulses for hydrogen (with a single-active electron) and also for helium (with two-active electrons) at various conditions. When the intensity is low, the temporal peak of harmonics has been found to coincide with the peak of the fundamental pulses. At higher intensity, however, we have found that the peak of the harmonic temporally shifts toward the leading edge of the pulse, and the amount of the shift is smaller for higher order harmonics.

I-T-2 Two-Photon Above-Threshold Ionization Cross Sections of Rare Gases by XUV Photons

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[*Phys. Rev. Lett.* **93**, 083903 (2004)] [*Phys. Rev. A* **70**, 043412 (2004)] Above-threshold ionization (ATI) is the successive absorption of an additional number of photons more than the minimum required to ionize an atom. For rare gas atoms, ATI by infrared ~ visible photons has been very well studied theoretically as well as experimentally. Due to the recent technological progress in highorder harmonic generation and free-electron lasers, a bright light source is becoming available in the extreme ultraviolet (XUV) wavelength region. Provided with such progress, the time is matured to investigate, theoretically as well as experimentally, ATI in the XUV regime. Furthermore, two-photon ATI has a practical importance since it can be used for the pulse characterization (*i.e.*, autocorrelation) of XUV pulses.

In this work we have developed a theory to calculate two-photon above-threshold ionization cross sections of rare gases by XUV photons in the extended framework of multichannel quantum defect theory (MQDT). The advantage of the use of MQDT is that it automatically incorporates not only the spin-orbit interactions but also configuration mixing to some extent. On the other hand, the limitation of the present approach is that we have included only two lowest core states (two-core model), $p^{5}[^{2}P_{3/2}]$ and $p^{5}[^{2}P_{1/2}]$. Since free-free dipole moments do not converge in the length gauge, we have made the combined use of the length gauge for small r and the acceleration gauge for large r. The two-photon ATI cross sections for Xe and Ar associated with the $p^{5}[^{2}P_{3/2}]$ and $p^{5}[^{2}P_{1/2}]$ ionic core states are found to be 1.0×10^{-51} cm⁴ s and 2.0×10^{-51} cm⁴ s, respectively, for 25 eV photons.

I-U Control of Photoionization Processes Using Lasers

Optical control of various photoabsorption processes are of great interest in recent years. In this project, we have theoretically explored the possibility to control spin degree of freedom of photoelectrons and ejection angle of photoelectrons.

I-U-1 Control of the Spin-Polarization of Photoelectrons/photoions Using Short Laser Pulses

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To establish a method for the control of spin degree of freedom is one of the most important issues in modern technology and science, since highly spinpolarized species such as electrons, ions, and nucleus, *etc.*, are very useful not only to develop a new technology using semiconductors but also to study various spin-dependent dynamics. We theoretically propose a generic pump-probe scheme to control spin-polarization of photoelectrons/photoions by short laser pulses. By coherently exciting fine structure manifolds of a multivalence-electron system by the pump laser, a superposition of fine structure states is created. Since each fine structure state can be further decomposed into a superposition of various spin states of valence electrons, each spin component evolves differently in time. This means that varying the time delay between the pump and probe lasers leads to the control of spin states. Specific theoretical results are presented for twovalence-electron atoms, in particular for Mg, which demonstrate that not only the degree of spin-polarization but also its sign can be manipulated through time delay. Since the underline physics is rather general and transparent, the presented idea may be potentially applied to nanostructures such as quantum wells and quantum dots.

I-U-2 Control of Photoelectron Angular Distributions Using a Dressing Laser

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Strong dressing laser field can induce various interesting modification in laser-matter interactions. Among them, an interesting modification is observed in the photoionization spectra in the wavelength region at which two-photon near-resonance is satisfied for the initially occupied state by a probe laser and initially unoccupied state by a dressing laser. This is known as laser-induced continuum structure. Related to this, if the ionization processes consists of several channels, it should be possible, in principle, to alter the branching ratio of photoionization into several channels, and if the branching ratio is successfully altered, we may also see the alteration of photoelectron angular distribution. In this work we have theoretically studied the modification of photoelectron angular distributions using a dressing laser. We have found a significant change of photoelectron angular distributions for the K atom, as the probe laser frequency is scanned across two-photon resonance under the presence of dressing laser.