Two-color photoionization of atoms in low and high radiation fields

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Abstract. Atomic photoionization was studied by means of electron spectroscopy using two photon sources of different wavelengths for the excitation. In one part of the experiments, alkaline atoms were excited by an optical laser of high spectral resolution prior to their photoionization by short wavelength radiation from the synchrotron radiation source BESSY. These low field studies concentrated on measurements of the linear alignment dichroism in the angular distribution (LADAD) and of the linear magnetic dichroism (LMDAD) in the $2p$ ionization of laser-polarized sodium atoms. In another type of experiments, the high intensity of a femtosecond optical laser was used for the investigation of dressed atoms, i.e. of the photoionization dynamics in strong external fields. In particular, the above threshold ionization (ATI) of rare gases was studied at the free electron laser facility FLASH in Hamburg, Germany. Measurements of the linear dichroism, here in the direct two-photon ionization of the $1s$-shell in He, were performed in order to determine the partial photoionization cross sections related to the emission of electrons with $s$- and $d$-symmetry, respectively.

1. Introduction
Two-color pump-probe experiments have demonstrated in the last decades their particular importance for the understanding of photoionization process and underlying dynamics [1, 2]. In many of these studies, an optical laser of high spectral resolution is used to change in a controlled way, via resonant excitation of an outer electron, the initial state for the photoionization, which is induced for example by synchrotron radiation (SR). The atoms are exposed to relatively low radiation fields created by the continuous-wave optical laser and the SR. Therefore, excitation and ionization processes can be treated independently. In addition to high-resolution spectroscopic studies (e.g. [3, 4]), special advantage is taken from the well-defined and easily changeable polarization of the laser. Due to the dipole selection rules for excitation with linearly and circularly polarized light, which induces only transitions of the type $\Delta m = 0$ and $\Delta m = \pm 1$, respectively, it is possible to prepare aligned or oriented atoms, i.e. to obtain an non-isotropic population of the sub-levels $m_l$ in the electronic state with total angular momentum $J$. Subsequent resonant photoexcitation or direct photoionization with polarized SR has provided many valuable and very detailed information on the photoionization dynamics in polarized excited (e.g. [5, 6]) as well as ground state atoms polarized by optical pumping (e.g. [7, 8]). In general, the corresponding measurements of the linear (circular) dichroism provide ideal research grounds to access the dynamics of the photoionization process (e.g. [9] and references therein). As example for this type of studies, recent high-resolution experiments on laser-polarized sodium atoms and the corresponding theoretical treatment are described below.
In contrast to these studies, the interaction of an intense femtosecond optical laser with the atom enables the investigation of dressed atoms, i.e. of the photoionization dynamics in strong external fields [10-12]. When applying an optical field of $10^{10}$ to $10^{13}$ W/cm² to the atom during the ionization by a XUV radiation pulse, the electron spectrum is strongly affected and the simultaneous action of both pulses has to be considered for the adequate description of the process (e.g. [13]). These experiments have attracted more and more interest in the last years due to the performances and availability of free electron laser (FEL) sources for the VUV and XUV wavelength regime, such as FLASH in Hamburg (Germany) [14]. In most cases, the experimental spectra of the two-color two-photon ionization process are well described by theoretical models based on solving the time-dependent Schrödinger equation or applying the so-called "soft-photon" approximation [15]. A typical experimental study undertaken at FLASH will be given for illustration.

2. Experimental

The experiments on laser-polarized atoms combining laser and SR were performed at the SR source BESSY II in Berlin (Germany). Details of the experiments are presented elsewhere [16]. In brief, the counter-propagating laser and synchrotron radiation intersect a beam of sodium vapor, which is produced by a radiatively heated oven, in the source volume of a high-resolution electron energy analyzer (Scienta SES-2002) mounted at the magic angle (54°44') with respect to the linear SR polarization vector. For the present study, electron spectra were recorded at a photon energy of $h\nu_{(\text{SR})} = 54$ eV with an total spectral resolution of 27 meV. For the excitation of the sodium atoms, two continuous-wave single-mode ring dye lasers (Rhodamine 6G) were used to excite efficiently both hyperfine components of the Na$^{3p\ 3s\ 2S_{1/2}}$ ground state when pumping the transitions to the Na$^{*\ 3p\ 3p\ 3P_{1/2}}$ or the Na$^{*\ 3p\ 3p\ 3P_{3/2}}$ excited state. Up to 40% of the sodium atoms could be brought to the excited state in this way. Under these conditions the atoms were exposed to a field of not more than about $10^{4}$ to $10^{5}$ W/cm² when the laser was operating at maximum average power of 1W and was focused to a diameter of about 50 μm in the interaction volume.

The experiments in the high field regime were performed using the two-color pump-probe set-up at FLASH [17]. Both the FEL and the optical laser beam were introduced into the experimental chamber in a collinear geometry and intersected an effusive gas jet within the acceptance volume of a magnetic bottle electron spectrometer (MBES). In order to ensure perfect time overlap between the pulses and to eliminate effects arising from the inherent time jitter of the FEL of about 500 fs [18], the experiments were performed using a picosecond optical laser synchronized to the 10-20 fs FEL pulses [14]. The relative orientation between the linear polarization vectors (purity better than 99%) was changed by means of a rotatable half-wave plate in the optical path. The FEL was operated in single-bunch mode at 13.7 nm (90.5 eV) with 5 Hz repetition rate and mean pulse energy of about 20 μJ. The optical field was produced by an intense (up to 2 mJ, 800 nm, 4 ps) Ti:Sapphire laser. The beam diameters of the FEL and the optical laser in the interaction volume were measured to be about 30 and 50 μm, respectively, which produces optical dressing fields of more than $10^{11}$ W/cm². The energy resolution in the electron spectra was mainly determined by the bandwidth of the FEL (0.5-1%) and the MBES (at best 1-2% of the electron kinetic energies) [17].

3. Results and discussion

3.1. Photoionization of laser-excited atoms

In many studies of laser-excited atoms sodium was chosen as primary example in order to highlight fundamental aspects of the photoionization process or to demonstrate the feasibility of a particular type of investigation [1]. Recently, it was therefore also used to investigate the effect of light polarization for the intensity distribution between the fine-structure states of the Na$^{*\ 2p\ 3n\ 1}P$ and Na$^{*\ 2p\ 3p\ 1}S, P, D$ multiplets for ionization from the ground and the excited state, respectively. Photoionization from the Na$^{*\ 2p\ 3p\ 1}P_{3/2}$
excited state (Figure 1) results in a complex structure of 10 fine-structure components, which are, with the exception of the very close lying (1.5 meV) \(^3\)P\(_0\) and \(^3\)P\(_1\) lines, all resolved in the experiment. Assignment and energy positions of the states are given with respect to tabulated reference data [20]. The displayed spectrum was recorded with a relative angle \(\theta = 0^\circ\) between the linear polarization of the SR and the laser, i.e. parallel orientation of both vectors. The overall intensity distribution is in agreement with earlier experiments [3] showing in particular the strong intensity of the \(^1\)D\(_2\) line upon photoionization from the \(^3\)P\(_{3/2}\) excited state. This line is completely absent upon ionization from the \(^3\)P\(_{1/2}\) state. In the lower part of the figure, the linear alignment dichroism in the angular distribution (LADAD) is displayed. The spectrum has been obtained by subtraction two spectra recorded at \(\theta = 90^\circ\) and \(\theta = 0^\circ\). It is obvious that the individual lines behave quite differently, showing different amplitudes for the relative change of the intensity as well as different signs of the LADAD. For the interpretation of the results we have compared the experimental spectra with theoretical results obtained in the multi-configurational Dirac-Fock approximation as well as in the generalized geometrical model (GGM) [3, 19]. Different from the standard treatment of photoionization based on a geometrical model [21], the GGM takes into account intermediate coupling in the initial atomic and final ionic states, but neglects configuration mixing and energy dependence of the electron wavefunction within the energy range of the multiplet. The LADAD spectrum obtained from the GGM is included in the figure showing that almost all variations are perfectly reproduced. The main achievement of the GGM with respect to the geometrical model relies in the inclusion of intermediate coupling. When pure LSJ coupling is applied the agreement between and theory is less satisfactory [19], in particular in the energy region of the \(^3\)P\(_{0,1,2}\) and \(^1\)P\(_1\) lines, where the LSJ coupling gives very different signs and intensity ratios for the LADAD of the individual lines.

![Figure 1](image1.png)

**Figure 1.** (Top) Photoelectron spectrum of sodium laser-aligned in the Na\(^+\) 2p\(^6\)3p \(^2\)P\(_{3/2}\) excited state, recorded at \(h\nu(SR) = 54\text{eV}\) and with parallel orientation between the linear polarization vectors of SR and laser. (Bottom) Experimental results (red dots) and theoretical simulation within the GGM (green solid line) for the LADAD.

![Figure 2](image2.png)

**Figure 2.** (Top) Photoelectron spectrum of sodium laser-oriented in the Na 2p\(^6\)3s \(^2\)S\(_{1/2}\) ground state, recorded using linearly polarized SR at \(h\nu(SR) = 54\text{eV}\) and circularly polarized laser light. (Bottom) Experimental results (red dots) and theoretical simulation within the GGM (green solid line) for the LMDAD.

In a second part of the experiments, circularly polarized laser light was used enabling us to orient the excited state as well as the ground state. When probing the orientation of the Na 2p\(^6\)3s \(^2\)S\(_{1/2}\) ground
with linearly polarized synchrotron radiation, the individual components of the Na\(^{2p^53s}\) multiplet (Figure 2) show a strong variation with respect to the helicity of the circular laser polarization. The resulting spectra of the linear magnetic dichroism in the angular distribution (LMDAD), i.e. the difference between two spectra recorded with left-handed and right-handed circularly polarized laser, is given in the lower part. As for the LADAD, the experimental results are well described by the GGM. In this case, the LSJ coupling scheme fails also to describe the dichroism correctly. Especially for the \(^1P_1\) line, the LSJ model does not produce any dichroism, although experiment and GGM show a clear effect. In the GGM, the dichroism of the \(^1P_1\) line has its origin in the mixing with \(^3P_1\).

In general, the dichroism measurements can serve to extract the complete information on the photoionization process, i.e. to determine the partial photoionization amplitudes and the relative phases of the outgoing waves. In the present case of 2p photoionization of atomic Na, the relative importance of the emission of a continuum electron with s or d symmetry can be extracted. Theory predicts a value of \(z = 3.4\) for the ratio between the d and s channel amplitudes and a phase difference of \(\Delta\) between 1.2 and 1.4. The experimental results show the same tendencies, i.e. a ratio \(z = 3.2 \pm 2.6\) and a positive sign of \(\cos \Delta\), but the quite large error bar underlines the high sensitivity of the experiment to small fluctuations as well as the extremely high precision necessary in future studies in order to improve the present results.

3.2. Above Threshold Ionization of rare gas atoms

For the experiments in the high field regime, the two-color Above Threshold Ionization (ATI) of rare gases was investigated. ATI is the direct observation of a process where two photons interact simultaneously with the atom. When during the ionization process by the XUV photon an additional Visible/NIR field is present in the interaction volume, the outgoing electron is also exposed to this strong dressing field and can absorb or emit a second (or more) photon (Figure 3). In the photoelectron spectrum, this effect is observed through the appearance of so-called sidebands, which show up, when the radiation field is strong enough (>10\(^{10}\) W/cm\(^2\)). Due to the high sensitivity to the strength of the applied dressing field, the method is ideally suited to measure precisely the temporal width of the XUV pulses and/or to determine the relative jitter between two femtosecond pulses [12, 18]. It was first observed and studied with high harmonic generation sources [10-12], but the monochromaticity of the Free Electron Laser sources such as FLASH enable us now to perform experiments unperturbed from interference effects between neighboring harmonics, which complicates often the detailed interpretation of the experiments at harmonic sources.

A series of typical spectra recorded on atomic Xe with the photon energy of the FEL at 90.5 eV is presented in Figure 4. The region of the 5p ionization (binding energy of 12.1 and 13.4 eV for the \(^3P_{3/2}\) and \(^3P_{1/2}\) spin-orbit component, respectively) is shown together with the sidebands belonging to an additional absorption of the optical photon. As a function of the relative orientation of the linear polarization vectors, indicated by the angle \(\theta\), the intensity redistribution between the main line and the sideband changes in a characteristic way. For parallel orientation (\(\theta = 0^\circ\)), the intensity transfer from the main line to the sidebands is the strongest, for perpendicular orientation (\(\theta = 90^\circ\)) the smallest. The observed variations are well described within the soft-photon approximation [15] and in principle it is possible to extract from the presented data the relative partial cross section for the two-photon ionization of atomic Xe. However, for the case of ionization in the 5p shell of Xenon the detailed analysis is rather difficult, since the one-photon process leads already to the emission of electrons with two different (s and d) symmetries (see Figure 3). The two-photon process is then described by outgoing \(p\)- and \(f\)-electrons, which are produced via different pathways and are therefore subject to interference effects, which are difficult to treat in detail. An additional complication of the analysis arises from the fact it is difficult to separate the contributions from each 5p component. The energy separation of the two 5p spin-orbit components (1.3 eV) is close to the distance between the main photoline and the sidebands (1.5 eV), given by the photon energy of the dressing laser (800 nm).
For these reasons, the first measurements of polarization effects in the two-color ATI were performed for ionization of the 1s-electron in atomic He [22]. In these spectra, at (relatively) low dressing fields (about 8x10^{10} W/cm²) only one sideband is seen, which directly reflects the two-photon ionization of the 1s-electron into the s- and d-continua. By measuring the polarization dependence of the sideband intensity, a slightly higher (factor of 1.5) emission probability for s than for d electrons was determined from the experimental results. This observation is in contrast to the general propensity rules predicting that, upon absorption of one photon, transitions of the type (\ell \to \ell + 1) are favored with respect to those described by a decrease of the angular momentum (\ell \to \ell - 1). Detailed calculation using the solution of the time-resolved Schrödinger equation are in good agreement with the experiment and explain the behavior by the overlap of the oscillatory asymptotes of the coupled continuum states in the particular case of He 1s two-photon ionization [22].

4. Conclusion
Recent results of two-color photoionization studies combining a XUV and a Visible/NIR photon sources are presented. For the experiments in the low field regime, the action of both photons can be treated separately and spectroscopic investigation with high spectral resolution are possible. The linear dichroism (LADAD and LMDAD) in the 2p photoionization of laser-polarized sodium atoms was chosen as example to illustrate the present state-of-the-art for this type of experiments, with respect to the experimental performances as well as to the theoretical models describing the dichroism. Very detailed information of the photoionization process can be obtained, up to the complete determination of all parameters describing the photoionization process ("complete experiment"). For the future the extension to more complex atomic systems such as the open-shell 3d transition metals is envisaged, necessitating the inclusion of configuration mixing in the theoretical models. For the high-field regime, the combined action of both photons has to be taken into account. A recent example for the study of two-color ATI in rare gases was discussed, where for the first time the dichroic effect in the direct two-photon ionization was investigated. With the advent of the XUV Free Electron Laser sources providing monochromatic, intense, ultra-short pulses for the photoionization process, these
studies of non-linear effects will attract more and more interest. In combination with an intense femtosecond laser in the Visible/NIR wavelength regime, multi-photon processes as well as resonant excitation and corresponding electronic relaxation processes in strong dressing fields will be in the center of future investigations.

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