PHYSICAL REVIEW A
ATOMIC, MOLECULAR, AND OPTICAL PHYSICS

THIRD SERIES, VOLUME 58, NUMBER 5 NOVEMBER 1998

RAPID COMMUNICATIONS

The Rapid Communications section is intended for the accelerated publication of important new results. Since manuscripts submitted to this section are given priority treatment both in the editorial office and in production, authors should explain in their submittal letter why the work justifies this special handling. A Rapid Communication should be no longer than 4 printed pages and must be accompanied by an abstract. Page proofs are sent to authors.

A complete photoionization experiment with polarized atoms using magnetic dichroism and phase tilt measurements

K. Godehusen and P. Zimmermann
Institut für Atomare und Analytische Physik, Technische Universität Berlin, Hardenbergstrasse 36, D-10623 Berlin, Germany

A. Verweyen, A. von dem Borne, Ph. Wernet, and B. Sonntag
II. Institut für Experimentalphysik, Universität Hamburg, Luruper Chaussee 149, D-22761 Hamburg, Germany
(Received 17 February 1998)

The study of the $5p$ photoemission from laser polarized (aligned or oriented) Eu atoms was used to deduce the ratio of the magnitude of the electric dipole amplitudes and the phase difference between the two outgoing ϵs and ϵd electron waves. The combination of magnetic dichroism experiments, the measurement of the difference spectra for two atomic polarizations, and phase tilt experiments, the observation of the modulation of the photoelectron signal on the angle η of linear laser polarization, is a very promising method for performing a ''complete'' photoionization experiment.
[S1050-2947(98)50111-4]

PACS number(s): 32.80.Fb, 32.80.Hd

Atomic photoionization $A+h\nu\rightarrow A^{+}+e^{-}$ is one of the fundamental processes in the interaction of electromagnetic radiation with matter. In spite of this, there is a lack of experimental data for a complete quantum-mechanical description of this process. This is due to the fact that, even in the approximation of nonrelativistic electric dipole transitions, neglecting spin-orbit interactions in the continuum and the term dependence of core wave functions, the degeneracy of the continuum states with respect to the angular momentum requires the knowledge of two matrix elements $\langle nl|D|\epsilon l\pm 1\rangle$. Hence three parameters, two absolute amplitudes and the relative phase between the two outgoing photoelectron waves, are needed. In most cases, however, only two experimental parameters, the partial cross section $\sigma(nl)$ and the asymmetry parameter $\beta(nl)$ for the angular distribution of the photoelectrons, are available. Additional information for a more complete analysis can be obtained by measurements of the spin polarization of the photoelectrons [1] or the alignment/orientation of the photoions [2,3]. Complementarily to these experiments, which concentrate on the final products of the photoionization process, one can prepare the initial states by using polarized target atoms. For polarized atoms the angular distribution of the photoelectrons is characterized by more than just one asymmetry parameter, as in the case of randomly oriented atoms, thus yielding the additional information [4].

The preparation of polarized atoms for experiments with synchrotron radiation may be achieved with inhomogeneous fields [5] or by laser pumping [6,7]. Here, we report on inner-shell photoionization experiments with laser pumped polarized (aligned or oriented) Eu atoms. Special emphasis is laid on the combined use of dichroism and phase tilt measurements of the angular distribution of the $5p$ photoelectrons for the determination of the relative magnitude of the dipole amplitudes and the phase difference between the outgoing ϵs and ϵd waves. In the nonrelativistic dipole approximation these parameters are sufficient for a ''complete'' quantum-mechanical description of the photoionization process.

The experiments were performed at the undulator beamline U1-TGM6 of the electron storage ring BESSY in Berlin. The Eu atoms, which were produced in a resistively heated crucible, were prepared in an aligned or oriented ground state by optical pumping with the linearly or circularly polarized radiation of a cw ring dye laser (≈100 mW, single mode) which was tuned to the transition Eu $4f^7 6s^2\ ^8S_{7/2} - 4f^7 6s6p\ ^8P_{5/2}$ at λ=466.3 nm. The optical pumping effect of this transition $J=7/2 \rightarrow J'=5/2$ for both polarization modes strongly enhances the population of the Zeeman sublevels with the highest projection numbers $M_J=\pm 7/2$ of the ground state: linearly polarized light produces an alignment with an equal population of both sublevels $M_J=\pm 7/2$; circularly polarized light yields an orientation with the dominant population of $M_J=+7/2$ or $M_J=-7/2$. For a quantitative analysis of the pumping process one has to take into account the actual experimental conditions by solving the corresponding rate equations of the absorption and emission processes. The hyperfine splitting and the isotope shift of both isotopes ^{151}Eu and ^{153}Eu has to be considered also.

The polarized Eu atoms are ionized by monochromatized synchrotron radiation. The geometry of the experimental setup is the same as in earlier experiments [8]: The polarization axis $\vec{E}$ of the synchrotron radiation is fixed within the horizontal plane; the polarization axis $\vec{A}$ of the laser radiation can be rotated by the angle η from 0° to 360° with respect to the horizontal plane. Both radiation beams propagate antiparallel to each other and intersect the vertical atomic beam in front of a 180° cylindrical mirror analyzer that detects the photoelectrons under the magic angle $\theta=54.7°$ relative to the polarization axis $\vec{E}$ of the synchrotron radiation.

Dichroism experiments in photoionization studies are characterized by the variation of the photoelectron intensity for two polarization directions with respect to the ionizing photons or the atomic polarization. For the two different atomic polarizations, alignment and orientation, using linearly polarized synchrotron radiation, the notations LDAD (linear dichroism in the angular distribution) and LMDAD (linear magnetic dichroism in the angular distribution) are used. The LDAD depends on the atomic alignment A_{20}, whereas the LMDAD is sensitive to the atomic orientation A_{10}.

In our case the atomic polarization is changed by the polarization of the laser radiation. For linearly polarized laser radiation we measure the difference of the photoelectron signals for two mutually perpendicular directions; for circularly polarized laser radiation the difference for right- and left-handed polarization $\sigma^{\pm}$ is detected. In the first case, the atomic alignment A_{20} is changed; in the second case, the atomic orientation A_{10} is changed.

For the analysis of the dichroism spectra it is convenient to normalize the difference spectra to the partial cross section [9] introducing the asymmetry parameters β^{η}_{LDAD} and β_{LMDAD}:

$$\beta^{\eta}_{LDAD} A_{20} = \frac{I(\eta) - I(\eta - 90°)}{I(0°) + 2I(90°)}, \tag{1}$$

$$\beta_{LMDAD} A_{10} = \frac{I(\sigma^+) - I(\sigma^-)}{I(\sigma^+) + I(\sigma^-)}. \tag{2}$$

Note that Eqs. (1) and (2) are only valid for completely linearly polarized synchrotron radiation ($P=100\%$); the actual value for the U1-TGM6 beamline is $P=98\%$ [9]. It is also important to note that for the $5p$ ionization from the ground-state Eu $4f^7 6s^2\ ^8S_{7/2}$ atomic multipole moments $A_{k_0 0}$ with $k_0>2$ do not contribute to LDAD and LMDAD.

There are only two arbitrary values for η for which the asymmetry parameters β^{η}_{LDAD} are linearly independent. We have chosen $\eta=0°$ and $\eta=45°$ for convenience in the theoretical description.

The asymmetry parameters β^{η}_{LDAD} and β_{LMDAD} can be expressed in terms of the corresponding dipole matrix elements and of the relative phase between the two outgoing electron waves. Using the general formalism for the angular distribution of photoelectrons from polarized atoms in Ref. [10] the asymmetry parameters for the $5p$ photoionization of polarized Eu atoms can be written in LSJ coupling as

$$\beta^{45°}_{LDAD} = C(L_f, S_f, J_f) \frac{x}{x^2+1} \cos(\delta_s - \delta_d), \tag{3}$$

$$\beta_{LMDAD} = C'(L_f, S_f, J_f) \frac{x}{x^2+1} \sin(\delta_s - \delta_d), \tag{4}$$

where $x = |D_s|/|D_d|$ denotes the ratio of the reduced dipole matrix elements for the transition of the $5p$ electron into the ϵs and ϵd continuum states and $(\delta_s - \delta_d)$ is the phase difference of the two outgoing electron waves. The constants C and C' depend on the total orbital momentum L_f, the total spin S_f, and the total angular momentum J_f of the final ionic states.

The following figures show the experimental results. A photoelectron spectrum of the Eu $5p^{-1}$ lines taken at the photon energy $h\nu=53$ eV is depicted in Fig. 1. Several lines can be seen that are separated into one group of sharper lines between 24 V and 29 eV and a second group of broader lines between 29 and 34 eV. The sharper lines can be assigned as ^9L_f lines and the broader lines as ^7L_f lines. Considering the sharper lines the labeled ones are assigned to the $5p^5 4f^7 6s^2$ configuration, whereas the remaining three large lines may be assigned to the configuration $5p^5 4f^7 5d6s$. The difference in the linewidth of the ^9L_f and ^7L_f is due to Auger transitions $5p^{-1} - 4f4f$, which are spin-forbidden for the ^9L_f levels [11].

The dichroism spectra of the $5p^{-1}\ ^9P$ levels of *oriented* Eu atoms taken at the photon energy $h\nu=43.5$ eV are shown in Fig. 2. The upper part gives the photoelectron intensity $I(\sigma^+)$ and $I(\sigma^-)$ in the region between 25 and 29 eV for

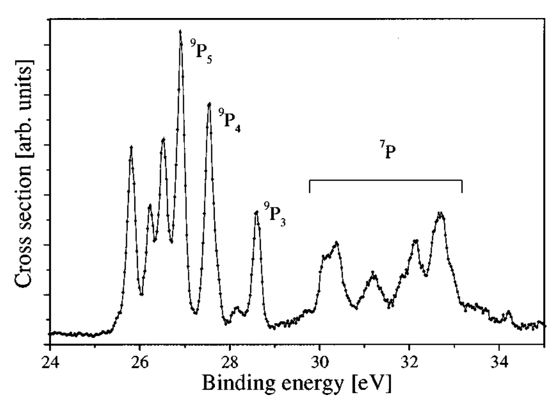

FIG. 1. Photoelectron spectrum of Eu taken at the photon energy $h\nu=53$ eV.

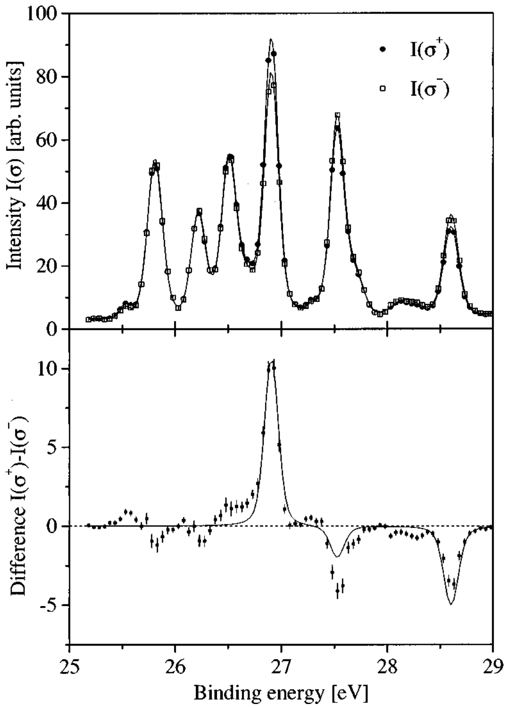

FIG. 2. Dichroism spectra of the $5p^{-1}\ ^9P$ levels for oriented Eu atoms taken at the photon energy $h\nu=43.5$ eV. Upper part: photoelectron intensity I for right-handed (σ^+) and left-handed (σ^-) circularly polarized laser radiation. Lower part: difference spectrum $I(\sigma^+)-I(\sigma^-)$ with the theoretical curve (solid line) using Eq. (4).

right- and left-handed circularly polarized laser radiation, respectively. The lower part shows the difference spectrum together with the theoretical curve. This curve was obtained from the factors C' of Eq. (4) multiplied by the partial experimental cross section [see Eq. (2)] for the three lines $5p^{-1}\ ^9P_5$, $^9P_4$, and $^9P_3$. By these means the relative difference spectrum was obtained, which was fitted to the experimental spectrum by multiplying with the optimal value for the parameter $x/(x^2+1)\sin(\delta_s-\delta_d)$.

There is good agreement for the $^9P_5$ and $^9P_3$ lines regarding the magnitude and sign of the dichroism effect. The discrepancies in the $^9P_4$ line may be due to deviations from the LSJ coupling scheme or the influence of configuration mixing with $(5d,6s)^2$ configurations.

Similar dichroism experiments with *aligned* Eu atoms were performed with linearly polarized laser radiation. From Eqs. (3) and (4) one can see that both methods yield the same dependence $x/(x^2+1)$ on the relative magnitude of the dipole matrix elements $x=|D_s|/|D_d|$, whereas the sine dependence of the phase difference $(\delta_s-\delta_d)$ in our case $[(\delta_s-\delta_d)\approx 180°]$ favors the use of circularly polarized laser radiation for the determination of the sign of the phase difference. In both cases, however, the experimental uncertainties in the alignment or orientation parameters A_{20} or A_{10} limit the accuracy in the determination of the dipole matrix elements. It is, as we shall see, very favorable to combine the dichroism experiment with phase tilt measurements. In this method, which was introduced with photoionization experiments of laser-aligned Ca atoms [9], the photoelectron intensity as a function of the angle η of laser polarization is detected (see Fig. 3).

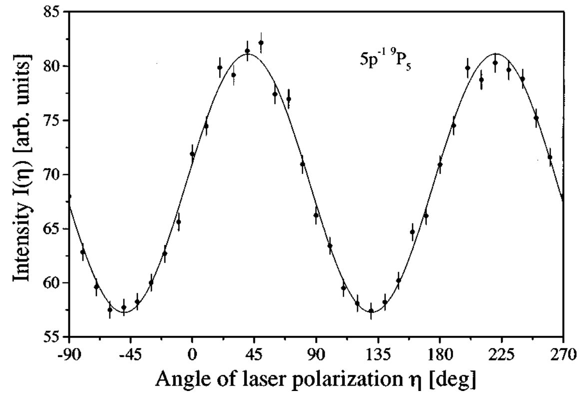

FIG. 3. Phase tilt measurement for the $5p^{-1}\ ^9P_5$ level showing the modulation of the photoelectron signal as a function of the angle η of the laser polarization direction. From Eq. (5) the phase tilt $\delta=2\eta_{max}=87(3)°$ was obtained.

For fixed detector position and variable laser polarization the intensity of the photoelectrons is modulated by [9]

$$I(\eta)=a+b\cos(2\eta-\delta). \tag{5}$$

The coefficients a and b and the phase tilt δ are functions of the dipole amplitude and the detector configuration. In contrast to the coefficients a and b, the phase tilt δ is independent of the alignment A_{20}. For the detector configuration of a cylindrical mirror analyzer at the angle $\theta=54.7°$ the phase tilt δ is given by

$$\tan\delta=\frac{\beta_{LDAD}^{45°}}{\beta_{LDAD}^{0°}}=-\frac{4}{\pi}\frac{x}{x^2-0.5}\cos(\delta_s-\delta_d). \tag{6}$$

The derivation of Eqs. (3), (4), and (6) from the general expression in Ref. [10] and a more detailed discussion are planned for a forthcoming paper. Figure 3 shows as an example the modulation of the photoelectron signal of the $5p^{-1}\ ^9P_5$ line as a function of the laser polarization angle η. From Eq. (5) the phase tilt $\delta=87(3)°$ was obtained.

Combining the dichroism and the phase tilt measurements, we derive the relative magnitude of the dipole amplitudes $x=|D_s|/|D_d|$ and the phase difference $(\delta_s-\delta_d)$ of the two outgoing ϵs and ϵd waves. The different dependencies on x and $(\delta_s-\delta_d)$ lead to very small error bars ($\approx 5\%$) in the final results. The main source of errors is the uncertainties ($\approx 30\%$) in the determination of A_{10} and A_{20}.

Figure 4 shows the combination of the results of dichroism and phase tilt measurements at the photon energy $h\nu=53$ eV. One can easily see that the area of possible values for x and $(\delta_s-\delta_d)$ is greatly reduced by the combination.

The final results for x and $(\delta_s-\delta_d)$ are shown in Fig. 5 for four different photon energies. The solid lines are the theoretical values that were obtained using the Cowan code [12]. The agreement between theoretical and experimental values is satisfactory, although the behavior of the phase difference with a minimum in the region of 51–53 eV is not reproduced by the calculations. This region is characterized by the $5s$ thresholds [13], which were not included in the calculations. These thresholds have no effect on the experi-

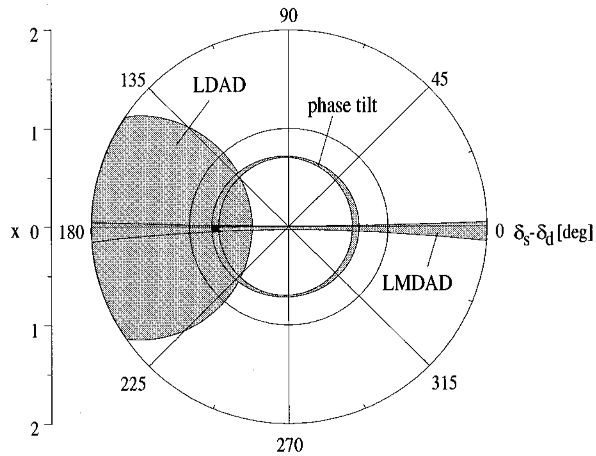

FIG. 4. Polar plot of the different sets of values for x and $(\delta_s - \delta_d)$ derived from the measurements of LDAD, LMDAD, and phase tilt at $h\nu = 53$ eV. The area where all three sets intersect is the black square.

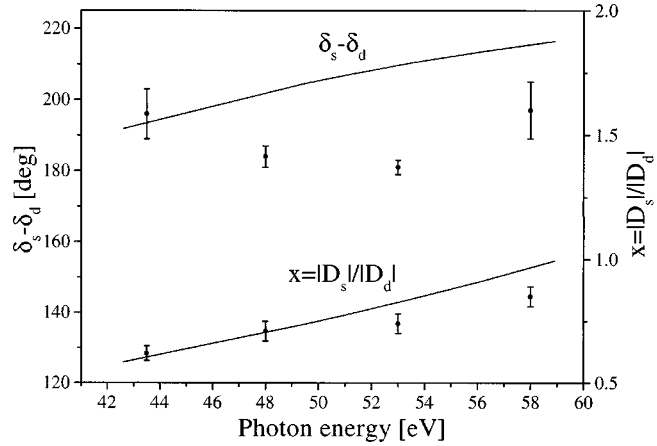

FIG. 5. Relative magnitude of the dipole amplitudes $x = |D_s|/|D_d|$ and phase difference $(\delta_s - \delta_d)$ for the outgoing electron waves ϵs, ϵd. The solid lines are the theoretical values obtained using the Cowan code.

mental $5p$ photoionization cross section and, therefore, x seems to be insensitive to them, but they might influence the phase difference.

In conclusion we have shown that the combination of dichroism and phase tilt measurements in the angular distribution of photoelectrons of polarized atoms is a very important step toward realizing a complete photoionization experiment. The phase tilt measurements are an essential addition to dichroism experiments. Because of their independence of the alignment A_{20}, they offer the opportunity to determine the ratio of the dipole matrix elements and their phase difference with much higher accuracy.

An important feature of our experimental setup is the use of laser pumping for the polarization of the atoms. This technique allows an easy change of the atomic multipole moments by means of rotating the direction of laser polarization or changing from linearly to circularly polarized light.

The authors would like to thank Dr. M. Martins for the calculations using the Cowan code and the BESSY staff for their assistance. The help of M. Grothe and I. Schubert is gratefully acknowledged. This work was supported by the Deutsche Forschungsgemeinschaft under Contract Nos. Zi 183/10-1 and So 176/2-1.

[1] U. Heinzmann, J. Phys. B **13**, 4353 (1980).

[2] A. Hausmann, B. Kämmerling, H. Kossmann, and V. Schmidt, Phys. Rev. Lett. **61**, 2669 (1988).

[3] J. B. West, K. Ueda, N. W. Kabachnik, K. J. Ross, H. E. Beyer, and H. Kleinpoppen, Phys. Rev. A **52**, R9 (1995).

[4] H. Klar and H. Kleinpoppen, J. Phys. B **15**, 933 (1982).

[5] O. Plotzke, G. Prümper, B. Zimmermann, U. Becker, and H. Kleinpoppen, Phys. Rev. Lett. **77**, 2642 (1996).

[6] M. Pahler, C. Lorenz, E. v. Raven, J. Rüder, B. Sonntag, S. Baier, B. R. Müller, M. Schulze, H. Staiger, P. Zimmermann, and N. M. Kabachnik, Phys. Rev. Lett. **68**, 2285 (1992).

[7] A. von dem Borne, Th. Dohrmann, A. Verweyen, B. Sonntag, K. Godehusen, and P. Zimmermann, Phys. Rev. Lett. **78**, 4019 (1997).

[8] Th. Dohrmann, A. von dem Borne, A. Verweyen, B. Sonntag, M. Wedowski, K. Godehusen, P. Zimmermann, and V. Dolmatov, J. Phys. B **29**, 4641 (1996).

[9] M. Wedowski, K. Godehusen, F. Weisbarth, P. Zimmermann, M. Martins, Th. Dohrmann, A. von dem Borne, B. Sonntag, and A. N. Grum-Grzhimailo, Phys. Rev. A **55**, 1922 (1997).

[10] S. Baier, A. N. Grum-Grzhimailo, and N. M. Kabachnik, J. Phys. B **27**, 3363 (1994).

[11] H. Ogasawara, A. Kotani, and B. T. Thole, Phys. Rev. B **50**, 12332 (1994).

[12] R. D. Cowan, *The Theory of Atomic Structure and Spectra* (University of California Press, Berkeley, 1981).

[13] M. Richter, M. Meyer, M. Pahler, T. Prescher, E. v. Raven, B. Sonntag, and H. E. Wetzel, Phys. Rev. A **40**, 7007 (1989).