High-resolution experimental determination of the angular distribution and spin polarization of xenon $7d'$ and $9s'$ photoelectrons and comparison with theoretical results

M. Spieweck, M. Drescher, F. Gierschner, R. Irrgang, and U. Heinzmann
Fakultät für Physik, Universität Bielefeld, Postfach 100131, 33501 Bielefeld, Germany
(Received 23 January 1998)

Using narrowband tunable linearly or circularly polarized VUV radiation an examination of the dynamical parameters describing the autoionization process of atomic xenon was performed. With a resolving power of $\sim 10^5$ in the excitation step a fully resolved determination of the position, magnitude and shape of the angular distribution and integral spin-polarization resonances was possible for the low Rydberg orders $n=7$ and $n=9$ of the d and s series, respectively. Comparisons are drawn between the experimental data and some theoretical calculations published in recent years.
[S1050-2947(98)00108-5]

PACS number(s): 32.80.Dz, 42.62.Fi, 32.70.$-$n, 42.65.$-$k

I. INTRODUCTION

The rare gases are relatively easy to deal with experimentally and thus far have been the best candidates for a promising comparison between theoretical calculations and experimental measurements due to their simple electronic structure in the ground state with filled shells and a total angular momentum $J=0$. Therefore during the last decades the rare gases were used as a test case for a most detailed comparison between theory and experiment [1–5]. Special attention was paid to the region of autoionization resonances between two ^2P ionization thresholds. Comparison with the observed quantity and shape of the relative cross section, the angular distribution, and the transferred spin polarization served as a sensitive test of different theoretical approaches [6]. However, up to now even for the lowest Rydberg orders no experimental data exists fully resolving the narrow s resonances in the Xe spectrum [3,4]. With the narrow band VUV radiation and high wavelength resolution used in this work it is now possible to perform an experimental examination of the behavior of the β and A parameters of the Xe $9s'$ autoionization resonance.

II. EXPERIMENT

To generate photoelectrons from xenon $7d'$ and $9s'$ autoionization resonances a windowless nonlinear sum frequency mixing technique is utilized because the excitation energy in the range of 96.2–97.1 nm lies well below the LiF cutoff (105 nm). Furthermore, narrowband VUV light has to be circularly polarized for measurements of the Fano effect [7]. Both demands can be met with the experimental setup that was used in a series of previous high resolution experiments on molecular targets like HCl, DCl, and HBr [8–10]: Two pulsed laser beams with frequencies ω_R and ω_T are focused into a pulsed Xe jet serving also as the nonlinear optical medium for frequency conversion. The radiation with ω_R is tuned to the $6p'$ $[\frac{1}{2}]_0$ two-photon resonance at 222.57 nm leading to strong enhancement of the intensity at the sum frequency $\omega_{\mathrm{VUV}}=2\omega_R+\omega_T$ [10]. For the production of circularly polarized VUV light the tunable radiation with frequency ω_T in the range of 709–761 nm can be circularly polarized using a $\lambda/4$ retarding plate [11]. A normal-incidence monochromator with a transmission of about 2% is used to separate the generated sum frequency from the fundamental and other conversion products. The degree of circular polarization, intensity, and optical alignment of the light beam with a flux up to 10^{10} photons/s is monitored using a four-mirror analyzer and a quad-photodiode, respectively. Wavelength calibration of the tunable light is carried out by recording optogalvanic spectra of a Xe/Ne-filled hollow cathode lamp with an absolute accuracy of $\pm 2\ \mathrm{cm}^{-1}$. The VUV bandwidth was determined to be less than $1.2\ \mathrm{cm}^{-1}$. A detailed description of the VUV source is given in Ref. [11].

In the ionization region, the VUV light perpendicularly intersects the supersonic target beam. This beam is produced by a pulsed nozzle-skimmer combination. The generated photoelectrons are extracted by a static dc field of 300 V/cm and after acceleration to 100 keV they are guided into a Mott-polarimeter [12,13]. In addition to the two backward and two forward counters needed for spin-polarization detection the instrument contains a straightforward counter to measure the integral photoelectron yield. The whole setup is shown in Fig. 1.

In the case of angular distribution measurements of the photoelectrons the setup is much simpler because no circularly polarized light and no spin-polarization detection system are needed: The $\lambda/4$ retarding plate is replaced by a $\lambda/2$ Fresnel rhomb to rotate the plane of linear polarization of the light. The Mott-polarimeter and electrostatic lens system are replaced by a double multichannel plate for differential photoelectron yield detection with no extraction field applied.

III. RESULTS AND DISCUSSION

Due to the fact that the studied atomic target xenon is also used as a nonlinear optical medium for VUV light generation it is not possible to obtain a high photoelectron signal over a wide energy range, since photoelectron yield as well as phase matching and susceptibility are strongly influenced by autoionization resonances [14–16]. Only at resonance positions can angular distribution and, in particular, spin measure-

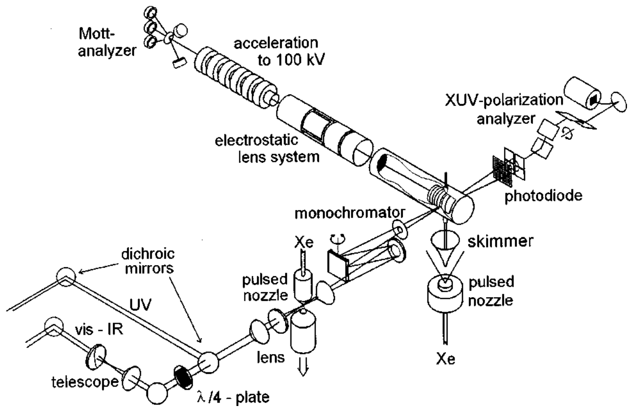

FIG. 1. Experimental setup for photoelectron spin-polarization measurements using VUV radiation, generated by sum-frequency mixing of two laser beams in xenon. In the case of angular distribution measurements the λ/4 retarding plate is replaced by a λ/2 Fresnel rhomb to rotate the plane of linear polarization of the light and the Mott polarimeter and electrostatic lens system is replaced by a double multichannel plate for photoelectron yield detection.

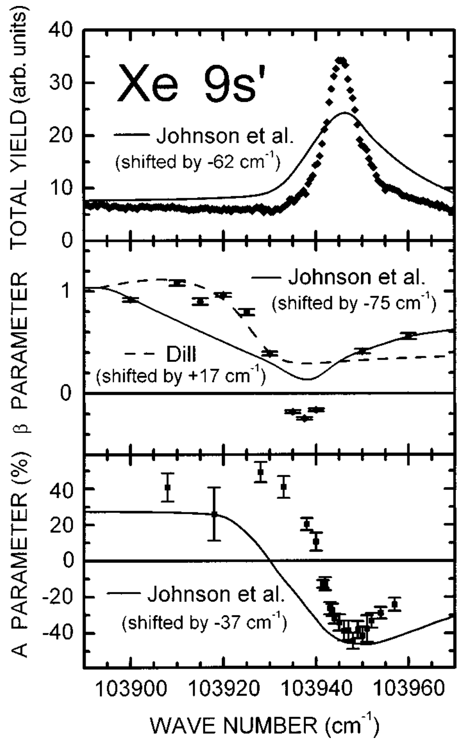

FIG. 2. Total yield, angular distribution parameter β, and integral spin polarization A of the photoelectrons at the Xe $9s'$ resonance. In addition to the experimental data theoretical curves from Dill [4] (dashed line) and Johnson *et al.* [3] (full line) are shown.

ments be performed with sufficiently small statistical errors. Each data point of the spin polarization A in Figs. 2 and 3 represents an average over 10 000–20 000 laser shots and was reproduced several times. Instrumental asymmetries have been eliminated by changing the helicity of the circular polarization between each measurement. The absolute values of the integral spin polarization are subject to an uncertainty of 10% due to limited determination of the effective Sherman-function (0.247 ± 0.021) [12] and the degree of circular polarization of the light ($97.7\pm2.2\%$).

In Fig. 2 the results for the Xe $9s'$ resonance are presented. The total yield was taken with a step width of $0.5\ \mathrm{cm}^{-1}$. The peak position of the resonance [full width at half-maximum (FWHM)$=7.6\ \mathrm{cm}^{-1}$] was determined to be $103\,945\pm2\ \mathrm{cm}^{-1}$. In the same plot the calculated cross section from Ref. [3] (shifted by $-62\ \mathrm{cm}^{-1}$) is displayed. Since the absolute wave-number position is different for different calculations, for the three theoretical curves from Johnson *et al.* in this figure (σ, β, and A) and for all other presented calculations different shifts are used to enable a comparison of the shape and width of the resonances with the experimental data.

The minimum of the integral spin polarization A was determined to be -0.444 ± 0.044 at $103948\ \mathrm{cm}^{-1}$. This is in perfect accordance with the calculations of Ref. [3] plotted in the same figure, if the theoretical curve is shifted by $-37\ \mathrm{cm}^{-1}$. However, there is a discrepancy between theory and experiment since the width of the observed resonance is considerably smaller than in theory, while the oscillator strengths are differing not so strongly. Therefore the experimental cross section in the resonance is five times higher than the background while the theoretical ratio is only about 3.5. This difference in the width leads also to essential differences between the shapes of the experimental and theoretical curves for the A and β parameters in the $9s'$ resonance [17]. This can be observed particularly on the low-energy side of the spin-polarization peak. Unfortunately the statistical error is large in this region due to very low VUV light intensity.

The results for the broad Xe $7d'$ resonance are shown in Fig. 3. The plotted experimental data of the total yield is scanned from Ref. [1], digitized and converted to a corrected wave-number scale. This procedure has been carried out also for all the theoretical curves in Figs. 2 and 3 to make a comparison possible. The experimental data for the integral spin polarization A obtained with the setup described above perfectly confirms the previous measurements by Heinzmann using synchotron radiation with a bandwidth of $\sim 53\ \mathrm{cm}^{-1}$ [1]. Theoretical calculations from Johnson *et al.* [3] are added to this figure, too. This time the shape of the theoretical curve (shifted by $-100\ \mathrm{cm}^{-1}$) fits very well the experimental spin-polarization data, but there is a difference of $\Delta A=0.1$ of the maximum values.

For measurements of the angular distribution parameter β in the case of a fixed electron detector it is necessary to rotate the polarization plane of the VUV radiation. For linearly polarized light β can unambiguously be derived from the ratio of intensities at 0° and 90° [18]. In order to exclude experimental asymmetries, β has also been determined by measuring a full angular distribution for selected energies and fitting to a second Legendre polynomial. The values obtained this way were in good agreement with those from the two-angle method. Each data point for β-parameter measurements in Figs. 2 and 3 represents 20 000 laser shots for each angle. The statistical error typically is less than 4%.

Next to the spin polarization data the results for the β parameter of the Xe $9s'$ photoelectrons are presented in Fig. 2. The minimum ($\beta=-0.245$) is located at $103\,938\pm2\ \mathrm{cm}^{-1}$. In addition, two theoretical curves from Dill [4] and Johnson *et al.* [3] are displayed, shifted by $+17$ and

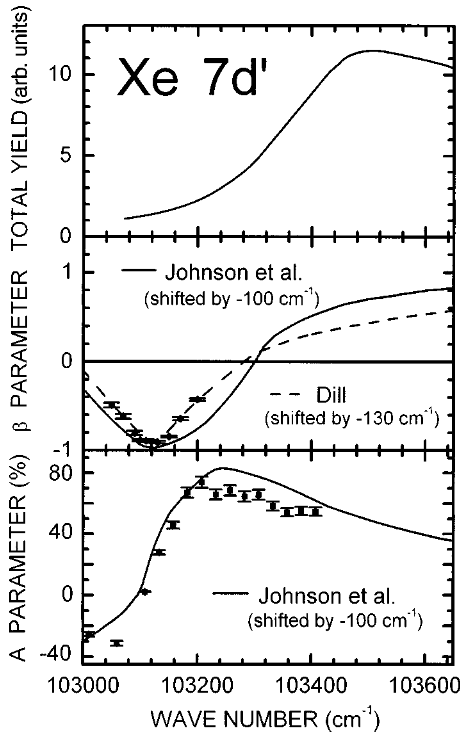

FIG. 3. Total yield, angular distribution parameter β, and integral spin polarization A of the photoelectrons at the Xe $7d'$ resonance. In addition to the experimental data theoretical curves from Dill [4] (dashed line) and Johnson *et al.* [3] (full line) are shown. The experimental data for the total yield is scanned from Ref. [1], digitized and converted to a wave-number scale.

$-75\ \mathrm{cm}^{-1}$, respectively. For the low-energy side of the peak the calculation of Dill is in good agreement with the experiment while the curve from Johnson *et al.* is not in accordance with the measured values. At the high-energy side of the peak the situation changes completely. Here a good agreement between theory and experiment is given only for the curve of Johnson *et al.* But both theories do not reach the minimum value of β. For the angular distribution of s resonances a discrepancy remains between theoretical calculations and experimental data.

It is interesting to make the same comparison for the broader Xe $7d'$ resonance (Fig. 3): Here the minimum value was determined to be $\beta=-0.91$ at $103\,130\ \mathrm{cm}^{-1}$. The experimental data is in excellent agreement with the calculation from Dill (shifted by $-130\ \mathrm{cm}^{-1}$). There is an agreement with the theory of Johnson *et al.*, too, but the width of this curve (shifted by the same value as for the spin polarization) is too large.

IV. CONCLUSION

In this report we present measurements of the angular distribution parameter and integral spin polarization of Xe $9s'$ and $7d'$ photoelectrons. Due to a VUV bandwidth considerably smaller than the natural linewidth it was possible to obtain exact experimental data for the resonance position, magnitude, and shape. Though the theory describes well the general characteristics, like the total photoionization cross section above thresholds, as well as the broad d-autoionization resonances, there is still a lack of agreement to be observed for the narrow s resonances. A closer consideration of both the d and s series will lead to a more refined theoretical description of the autoionization process of closed shell atoms.

ACKNOWLEDGMENTS

We thank Professor N. A. Cherepkov for encouraging this work and for reading the manuscript. The support of the Deutsche Forschungsgemeinschaft (SFB 216) is gratefully acknowledged.

[1] U. Heinzmann, J. Phys. B **13**, 4353 (1980).
[2] M. Y. Amusia and N. A. Cherepkov, Case Stud. At. Phys. **5**, 47 (1975).
[3] W. R. Johnson, K. T. Cheng, K.-N. Huang, and M. Le Dourneuf, Phys. Rev. A **22**, 989 (1980).
[4] D. Dill, J. A. R. Samson, and J. L. Gardner, Phys. Rev. Lett. **31**, 1327 (1973).
[5] D. M. P. Holland *et al.*, Nucl. Instrum. Methods Phys. Res. **195**, 331 (1982).
[6] U. Heinzmann and N. A. Cherepkov, in *Spin Polarization in Photoionization* (Plenum, New York, 1996), Chap. 15.
[7] N. A. Cherepkov, *Polarisation of Electrons Ejected by Circularly Polarized Light from Rare Gas Atoms* (Inst. Phys., Beograd, Yugoslavia, 1973), pp. 583–4.
[8] M. Drescher, R. Irrgang, A. Elizarov, N. Böwering, and U. Heinzmann, Phys. Rev. Lett. **75**, 2936 (1995).
[9] M. Drescher, A. Brockhinke, N. Böwering, U. Heinzmann, and H. Lefebvre-Brion, J. Chem. Phys. **99**, 2300 (1993).
[10] R. Irrgang, N. Böwering, M. Drescher, M. Spieweck, and U. Heinzmann, J. Chem. Phys. **104**, 8966 (1996).
[11] R. Irrgang, M. Drescher, F. Gierschner, M. Spieweck, and U. Heinzmann, Meas. Sci. Technol. **9**, 422 (1998).
[12] T. J. Gay and F. B. Dunning, Rev. Sci. Instrum. **61**, 1635 (1992).
[13] G. D. Fletcher, T. J. Gay, and M. S. Lubell, Phys. Rev. A **34**, 911 (1986).
[14] G. J. Bjorklund, IEEE J. Quantum Electron. **11**, 287 (1975).
[15] C. R. Vidal, *Four Wave Frequency Mixing*, Topics in Appl. Phys.–Tunable Lasers (Springer-Verlag, Heidelberg, 1986).
[16] A. Lago, G. Hilber, R. Hilbig, and R. Wallenstein, Laser Optoelektron. **4**, 367 (1985).
[17] N. A. Cherepkov (private communication).
[18] J. A. R. Samson and J. L. Gardner, J. Opt. Soc. Am. **62**, 856 (1972).