Dynamically Induced Spin Polarization of Resonant Auger Electrons

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(Received 22 October 1998)

Resonant Auger electrons from the decay of the atomic Xe $4d_{5/2}^{-1}6p(J = 1)$ state excited with linearly polarized light are shown to have a strong spin polarization. The core hole states are not oriented in this case. We found polarization values ranging from -0.46 to 0.57. For the decay to the $5p^4(^1D_2)6p(^2P_{1/2})$ state the ratio of the radiationless decay matrix elements and their relative phase, including its sign, are derived. [S0031-9007(99)09447-8]

PACS numbers: 32.80.Dz, 32.80.Hd, 32.90.+a

The dynamics of photo- and Auger electron emission is completely determined by the transition matrix elements involved in the particular process, including their relative phases. Whereas the photo- and Auger emission intensities are basically determined by the absolute size of the matrix elements, either dipole or Coulomb, the emission anisotropies and polarization properties are governed by the phase shift between different matrix elements. Therefore knowledge of the phase shift makes it possible to estimate the size of the polarization effects and emission anisotropies which may be observed in an experiment. This is one of the reasons why complete photoionization experiments [1] have attracted rapidly increasing interest during the last couple of years. As a result it is now possible to approximately predict the photoionization properties of most atoms by either theoretically calculated or semiempirically determined photoionization parameters within a relatively simple 4-parameter model [2].

This favorable situation is in marked contrast to the situation in Auger spectroscopy. Although Auger spectroscopy is a well established field of intense research activity in gas phase, surface and solid state physics, it is nearly solely concerned with the Auger intensities. A few studies have yielded phase-shift-dependent information such as angular distribution parameters, i.e., [3–5], or spin polarization parameters [6,7], but, as these experiments probed polarization terms depending on the cosine of the relative phases, they can be compared to angular distribution measurements in photoionization and, hence, their sensitivity to the phase shifts is not very high.

In this Letter we report on the observation of spin polarization from the radiationless decay of Xe atoms after $4d_{5/2} \rightarrow 6p$ $(J = 1)$ excitation. For several lines a strong spin polarization is observed. In contrast to earlier experiments with circularly polarized light [6,7], we excite inner-shell electrons using linear polarization. This case, a so-called dynamical spin polarization of Auger electrons, was theoretically discussed nearly twenty years ago [8]. The effect should have a pronounced dependence on the Auger phase shifts. In an early experimental attempt to verify this prediction, a very weak polarization effect in Kr Auger decays after impact of (unpolarized) 1.5 keV electrons was reported [9]. The dynamical spin polarization of Auger electrons after photoionization of free Ba atoms has been measured to be zero within experimental uncertainty [10]. Thereafter it was generally believed that impact of particles or photons possessing no helicity is not effective in transferring a spin polarization to secondary processes. Our measurements differ in two points from these earlier experiments: (i) Instead of probing inner-shell ionized states we probed the decay of an inner-shell excited neutral state, the so-called resonant Auger decay. In excited neutral states the alignment, or electric quadrupole moment, which is the origin of dynamical spin polarization, may greatly exceed the values found for a photoion. (ii) Angular momentum and parity selection rules may restrict the number of outgoing partial waves in a way that is favorable for the creation of dynamic spin polarization.

We used a combination of a time-of-flight electron drift tube and a retarding spherical field Mott polarimeter [7] to measure the energy and spin-polarization of electrons emitted by Xe after excitation by linearly polarized synchrotron radiation. The TGM5 beam line of the BESSY I storage ring (Berlin, Germany) running in single-bunch mode ($\Delta t = 208$ ns) served as a pulsed light source. An effusive gas beam crossed the synchrotron light to produce photoelectrons. Adjustment of the Xe target density resulted in a background pressure of 10^{-4} mbar in the vacuum chamber. The electrons were detected in the plane perpendicular to the direction of incident light $\mathbf{k}_\gamma$; the angle ϕ from horizontal to the detector within that plane was 135°. The z axis of our coordinate system aligns with the electron propagation direction $\mathbf{k}_e$; the x axis is perpendicular to the reaction plane spanned by $\mathbf{k}_e$ and the electric field vector. Two micro-channel-plate (MCP) counters are set up to detect the spin polarization component P_x antiparallel to $\mathbf{k}_\gamma$. A negative voltage of approximately -30 V was applied to the drift tube to increase the energy resolution. Two spectra of the $4d_{5/2}^{-1}6p$ resonant Auger lines with a total of 175 000 events were accumulated simultaneously for about 4200 s.

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In Fig. 1 the sum of the spectra obtained in both MCP counters is shown. The resulting lines can be grouped according to the coupling of the pertaining $5p^4$ valence double hole (^1S,^1D, or ^3P), with an additional splitting caused by the outer $6p$ electron. Most recently, angle-independent [11] and angle-resolved [3–5] experiments used an excitation by a bandpass smaller than the natural linewidth to disentangle this complex line structure. Several attempts to calculate the intensity distribution and angular anisotropy parameter for this spectrum have also been undertaken (see [4,11,12], and references therein). However, even in the most recent studies, significant discrepancies between calculated and measured values exist.

To extract spin polarization data from our time-of-flight electron spectra the time axis was converted to kinetic energy using line positions from Aksela *et al.* [11]. From the spectra of both counters the spin polarization P was determined for intervals 10 meV wide by $P = (\gamma R - 1)[(\gamma R + 1)S_{\mathrm{eff}}]^{-1}$. Here R is the intensity ratio I_1/I_2 of the counters, $S_{\mathrm{eff}} = -0.20(3)$ is the polarization sensitivity (Sherman function) of the Mott polarimeter [13], and γ is a correction factor for the instrumental asymmetry. In contrast to spin polarization experiments with circularly polarized light or magnetized samples, the instrumental asymmetry in our experiment cannot be eliminated from the data by combining measurements with reversed experimental conditions. We, therefore, recorded the He $1s$ photoelectron line, which cannot be polarized, before the actual Xe spectrum under identical conditions and determined γ as $\gamma = I_2^{\mathrm{He}}/I_1^{\mathrm{He}}$. By this procedure we found one group of Xe lines with a polarization significantly different from zero. The spin separated intensities $I_{+(-)} := [1 + (-)P](I_1 + I_2)/2$ and the spin polarization values P of this group are displayed in the top and bottom panels of Fig. 2. Error bars in this figure include only the statistical error. Errors in determination of γ will result mainly in a shift of the zero line of the polarization axis, while the error in S_{eff} affects all polarization values as a multiplication with a common factor.

The degree of light polarization at a photon energy of 93.8 eV was measured by a polarimetric setup using a transmission multilayer. We found nearly complete linear polarization within the orbit plane. The Stokes parameters were $p_1 = 0.97(2)$, $p_2 = -0.06(1)$, and circular polarization of 0.01. The same parameters were applied to our photon energy of 65.11 eV. This assumes that the polarization state of the undulator light does not change significantly with photon energy when the undulator gap is moved accordingly, which has been our experience in a number of photoelectron angular distribution studies.

For ease of comparison between our results to the data from other experiments and theoretical calculations, besides the interval-oriented analysis, a line-oriented analysis of our data is benefical. Unfortunately, the resolution obtained at the TGM5 beam line is not sufficient to resolve the close lying final states of the $5p^46p$ configuration. Therefore least squares fits using a "simulated annealing" algorithm were performed for both spectra. We used the line positions from Aksela *et al.* [11] for the major components and fixed them relative to each other. The intensities and widths of the lines were allowed to vary, as the resolution of the spectrum expressed as a function of E_{kin} is not

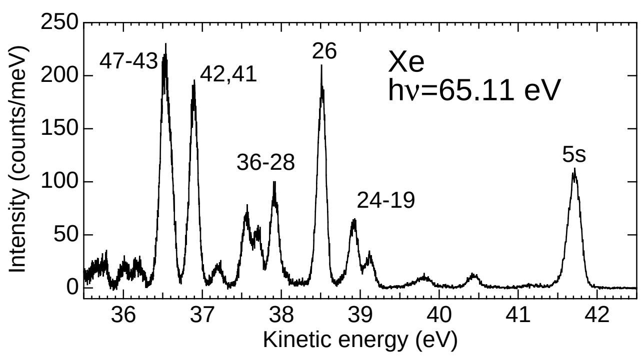

FIG. 1. A part of the Xe $4d_{5/2}^{-1}6p(J = 1) \rightarrow 5p^46p$ decay spectrum. The line numbers adhere to [11].

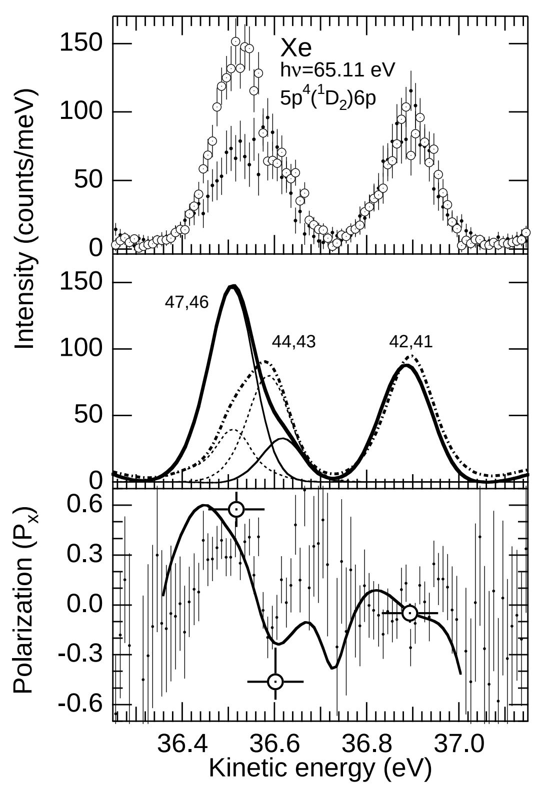

FIG. 2. Experimental results for the decay of Xe $4d_{5/2}^{-1}6p(J = 1)$ to the multiplet of $5p^4(^1D_2)6p$ states. Top panel: spin separated intensities I_+, I_- derived from 10 meV wide intervals of the measured spectra (open symbols: I_+); middle panel: I_+, I_- from our least squares analysis (solid lines: I_+, bold solid line: sum of the two underlying components for I_+, dotted and dot-dashed lines: I_-); bottom panel: spin polarization data derived analogous to the top panel (dots) and spin polarization results of the least squares analysis (solid line and open symbols).

constant. As a result, model independent values of the spin polarization for each pair of adjacent lines could be obtained (Table I). The row "Area determination" refers to the scatter of least squares fits using different model assumptions for peak and background shapes. A consistency check to the previously published relative intensities [11] showed that our fits had a tendency towards underestimating the area of the $^2D_{3/2,5/2}$ lines. Therefore, we allowed for an asymmetrical error bar in this case. Results of the fits are displayed in the middle panel of Fig. 2 in the form of spin separated intensities of the fit curves, and in the bottom panel of Fig. 2 as open symbols. It can be seen that the result of the interval-oriented analysis is corroborated by the line-oriented analysis. Obviously, the peak at 36.55 eV, unresolved in the intensity spectrum, consists of two components with opposite spin polarization of large absolute value (compare the bold solid and the dash-dotted line in the middle panel of Fig. 2). The horizontal error bar in Fig. 2 indicates the total apparatus broadening of ≈122 meV.

A parametrization of the spin polarization vector in the electron system has been published by Huang [14]. For the component P_x measured in our arrangement we obtain

$$P_x = \frac{-2\xi_2 p_1}{\sqrt{2} + \alpha_2(3p_2 - 1)/2}. \qquad (1)$$

Here ξ_2 and α_2 are the "intrinsic" parameters consisting of coupling coefficients and the decay matrix elements [15]. For excitation of closed shell atoms the value of the alignment is an exact number and has been inserted. From Eq. (1) it follows that the spin polarization parameter ξ_2 cannot be extracted without knowledge of the angular distribution parameter α_2. Therefore, to yield ξ_2 for the lines listed in Table I data for α_2 were taken from Aksela *et al.* [4].

Substituting everything into Eq. (1) gave the results displayed in Table II. The dominating contribution in the experimental error for ξ_2 is from the spin polarization measurement. Calculated values for the spin polarization parameters are given wherever available. No clear picture emerges from a comparison with these theories.

However, for at least one of the lines a more sophisticated comparison with the calculations is possible. The component showing the strongest polarization effect is assigned as a $(J_f = 1/2)$ state [11]. This entails that the resonant Auger decay can take place via only two different partial waves, $s_{1/2}$ and $d_{3/2}$. The intrinsic parameters α_2 and ξ_2 can, therefore, be expressed by the ratio of decay matrix elements $r := M_{1/2}/M_{3/2}$ and the difference of the total scattering phases $\Delta := \sigma_{3/2} - \sigma_{1/2}$. M_j is the real part of the Coulomb matrix element. Using expressions of Kabachnik and Sazhina [18] we get

$$\alpha_2 = (-2^{-1/2} + 2r\cos\Delta)/(1 + r^2) \qquad (2)$$

and

$$\xi_2 = -3r\sin\Delta/(2 + 2r^2). \qquad (3)$$

Inspection of Eq. (3) explains why a large polarization is to be expected in this case: As the phase difference Δ includes the difference in the Coulomb phase between s and d waves, it may deviate strongly from zero. Therefore the factor of $\sin\Delta$ in the expression will have an appreciable value as well.

These relations are displayed in the two dimensional plot shown in Fig. 3. Here, a possible influence of the weak shoulder line 47, pertaining to a satellite state of opposite parity to the $^2P_{1/2}$ line 46, has been neglected. A determination of r and Δ is possible by finding the intersection of the contours defined by Eqs. (2) and (3). By visual inspection we arrive at values of $r = -0.8 \pm 0.2$ and $\Delta = -0.85 \pm 0.1$. The only theoretical investigation from which we may extract these values yields $r = -1.24$ and $\Delta = -1.46$ [16]. While the disagreement for the phase difference in this case may result from the use of the spectator model in the determination of matrix elements, the too small value for r in our interpretation indicates once again the difficulties in describing the ionized $5p^4 6p$ states correctly.

TABLE I. Results for the spin-polarization component P_x and contributions to the experimental error. The first four lines of the table follow Aksela *et al.* [11]. Assignments for the lines 41–46 refer to a $5p^4(^1D_2)6p$ configuration; relative intensities refer to the $5s$ photoelectron line as 100. L and S of the assigned final state are only approximate. For the sum 43–47 we get $P_x = 0.20(8)$.

Line designation	41, 42	43, 44	46, 47
Assignment	$^2P_{3/2}, ^2F_{7/2}$	$^2D_{3/2,5/2}$	$^2P_{1/2}$, sat.
Kinetic energy (eV)	36.90, 36.85	36.62, 36.59	36.52, 36.47
Rel. intensity	139, 25	70, 77	102, 6.7
P_x	−0.049	−0.462	0.574
Statistical	0.031	0.043	0.032
Area determination	0.05	−0.1, +0.2	0.10
S_{eff}	0.006	0.062	0.077
Apparatus asymmetry	0.05	0.05	0.05
Total error	0.08	−0.14, +0.22	0.14

TABLE II. Experimental and theoretical values for the intrinsic spin polarization parameter ξ_2 and experimental values for α_2 [3,4]. Values are averaged over two or four lines (rightmost column) as indicated. Contributions from the photoionization satellite line 47 are neglected in all theoretical values.

No.	41, 42 $^2P_{3/2}, ^2F_{7/2}$	43, 44 $^2D_{3/2,5/2}$	46, 47 $^2P_{1/2}$, sat.	43–47
ξ_2 (Expt.)	0.04(6)	$0.27(^{0.08}_{-0.13})$	−0.60(15)	−0.16(6)
ξ_2 [12]	0.09[a]			−0.23
ξ_2 [16]	0.13	0.22	−0.73	−0.29
ξ_2 [17]		0.02	−0.61	−0.19
α_2	−0.19(3)	0.49(8)	−1.03(6)	−0.16(7)

[a] Additionally includes the (weak) line 39.

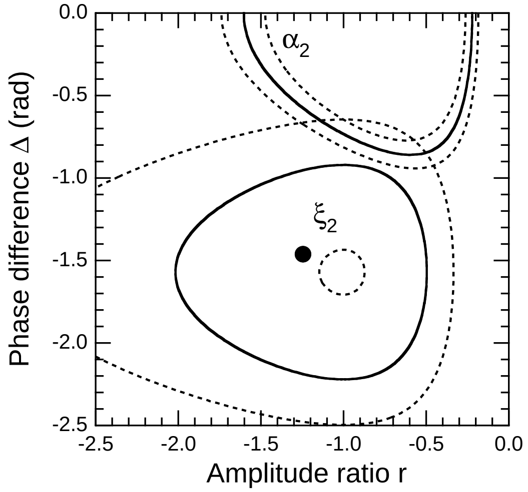

FIG. 3. Contour diagram of intrinsic Auger decay parameters α_2 and ξ_2 in dependence of the partial amplitude ratio r and relative phase Δ [Eqs. (2) and (3)]. Solid contours pertain to the measured values of $\alpha_2 = -1.10(6)$ [4] and $\xi_2 = -0.60(15)$, broken contours to 1 standard deviation. $\bullet$: Theoretical value for (r, Δ) from [16].

The particular resonant Auger decay discussed above bears an analogy to photoionization from a closed p shell. In this case the outgoing photoelectron is also described by s and d partial waves. Large spin polarizations have been found, for example, for photoionization of the Xe $5p$ shell [19].

In conclusion we have shown for the first time that a strong spin polarization of Auger electrons in radiationless decays can be observed even after photoexcitation by linearly polarized light. Combining the results of our spin polarization measurements and data from angular distribution measurements available in the literature, we determined the phase difference of the outgoing partial waves, including its absolute sign. This is an important first step towards a complete experiment for Auger decays. Since the main prerequisite for the dynamical spin polarization observed in this Letter is the large quadrupole moment of the intermediate state, this effect may also be observable in molecules and adsorbates, where the resonant Auger decay is a well known process.

While this manuscript was in preparation, we learned of a theoretical work showing that large ξ_2 values, as reported here, can be explained by a large phase shift of the $\varepsilon s_{1/2}$ wave [20]. Our work has been funded in part by the Deutsche Forschungsgemeinschaft. A critical reading of the manuscript by E. Rennie is acknowledged.

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