Experimental observation of left–right asymmetry in outer s-shell photoionization

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Abstract. The double differential cross-sections of outer s-shells photoelectrons have been measured for noble gases He, Ne, Ar, Kr, Xe and the H$_2$ molecule by linearly polarized photons. A nonzero left–right asymmetry has been observed relative to the mirroring plane that is perpendicular to the polarization direction of the photon beam.

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1. Introduction

In the quantum mechanical picture the atomic ionization/excitation and rearrangement processes are invariant for the mirroring of the coordinate system. This means that the left and right side intensities in the angular distribution of the emitted particles are identical. A right-handed coordinate system can be defined for linearly polarized light (figure 1): the oscillation of the $\vec{E}(t)$ electric vector of the incoming photon determines the ($X$–$Z$) plane and the momentum vector $\vec{k}$ points to the direction of the $X$-axis, the polarization vector ($\vec{P}$) locates at $Z$-axis and $Y$-axis is perpendicular to this plane. The left- and right-hand sides are defined in the following way: the positive $Z$-axis is the right side (R) and the negative one is the left side (L) relative to the propagation direction of the incoming light (figure 1). The mirroring plane is perpendicular to the polarization direction of the photon beam (($X$–$Y$) plane in figure 1). The left–right asymmetry parameter can be determined in the following:

$$A_{LR} = \frac{\sigma_L - \sigma_R}{\sigma_L + \sigma_R},$$

where $\sigma_L$ and $\sigma_R$ are the left- and right-hand side cross-sections. According to the quantum mechanics the asymmetry parameter is zero for electromagnetic interaction due to the space inversion symmetry.

During recent years, we have investigated the angular distribution of photoelectrons excited by linearly polarized synchrotron radiation and determined the dipole and the non-dipole anisotropy parameters [1]–[3]. In re-evaluation of the data a left–right asymmetry has been observed in the double differential cross-sections of the photoelectrons. In order to verify this finding, the experimental set-up (including the beamline and the spectrometer system) has been tested carefully with the 2p and 3p photolines of Ar. A series of measurements were carried out to study the left–right asymmetry parameters for the H$_2$ molecule and for the following s-shells of noble gas atoms: He 1s, Ne 2s, Ar 3s, Kr 4s and Xe 5s. The measurements were repeated when the vacuum chamber and the spectrometer were rotated by 180$^\circ$ in order to check the systematic errors. The double differential cross-sections of the emitted photoelectrons were determined relative to the Auger-electron production cross-section of Ar.

2. Experiment

The present investigations were carried out at the beamline I411 of the third generation MAX-II storage ring at Max-laboratory, Lund, Sweden [4, 5]. The degree of the linear polarization of the photon beam was better than 98% [2, 5]. (We note this small difference (from 100%) can reduce the magnitude of the anisotropy parameters but the left–right symmetry behavior of the angular distribution of the photoelectrons remains unchanged.) The bandwidth of the photon beam was about 0.5 eV when an exit slit of 200 $\mu$m was used in the monochromator. The incident photon energy range was 220.4–255.5 eV in the measurements but the argon 2p photoelectrons were measured with photon energy of 461.2 eV and slit size of 60 $\mu$m. The high-resolution settings allowed us to fully separate the spin–orbit components of the Ar 2p photoelectron lines. The photon flux was measured by a photodiode. Its current was digitized and used as a clock signal for controlling the data collection.

The electrons emitted from the collision region were analyzed with the ESA–22 electron spectrometer. A detailed description of the analyzer and measuring geometry is presented in [6].

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In short, the spectrometer consists of a spherical and a cylindrical part where the spherical mirror transports the electrons from the scattering plane to the entrance of the cylindrical analyzer performing the energy analysis. A spherical deceleration lens was mounted around the scattering region to improve the energy resolution of the system. The emission angles of the electrons are conserved from the target to the detectors due to the applied radial electrostatic field. The analyzer and the interaction region are lined with three layers of µ-metal sheets to reduce the magnetic field of the Earth. The residual magnetic field in the scattering plane and in the analyzer was less than 5 mG. The photoelectrons were detected by channeltrons in the co-planar geometry, i.e. in the polarization plane at 20 polar angles \( \theta = 15^\circ – 345^\circ \) (except \(0^\circ, 90^\circ, 180^\circ\) and \(270^\circ\)) relative to the polarization vector (see figure 1). The acceptance angle of each channeltron was \( \Delta \theta = \pm 5^\circ \) and in the perpendicular direction \( \Delta \phi = \pm 1.7^\circ \) (see figure 1 and [6]). The energy and angular distributions of the Auger and photoelectrons were measured at the pass energy \( (E_{\text{pass}}) 40 \text{ eV} \) and the energy resolution of the analyzer was 90 meV full-width at half-maximum (FWHM).

The unique capability of the \( E S A–22 \) electron spectrometer is to measure the total angular range of emitted electrons simultaneously. Therefore, the confidence in the pattern of the angular distribution is very high. Any instabilities in the photon flux or in the density of the target atoms produce the same effect at all angles. This is also valid for the current of the photodiode and the high voltage power supplies of the spectrometer and detectors. Changes in the experimental conditions modify simultaneously the line shapes or the intensities in different angular channels but the relative values remain unaffected.

The relative efficiencies of the detectors were determined by measuring the Ar \( L_2 – M_{3,3} M_{2,3} \) (3p)\(^{-2} \) \(^3\)\( P_{0,1,2} \) diagram Auger transitions at the average kinetic energy of 207.0 eV [6]. The angular distribution of these Auger electrons is isotropic since there is no alignment due to the \( J = 1/2 \) angular momentum of the initial state. If there is any left–right asymmetry, it changes only the magnitude of the measured asymmetry parameters but it cannot modify e.g. its dependence on the atomic number. The \(^3\)\( P_{0,1,2} \) Auger lines were measured at 400 eV photon energy, except for photoelectrons ejected from Ar 2p-shells where \( h\nu = 461.2 \text{ eV} \) was used to minimize the energy differences between the photoelectron and the Auger electron lines.
During the evaluation of the experimental data, the angular distribution of the photoelectrons was determined by normalizing the intensities of the photoelectron lines to the Auger lines in all angular channels separately (after linear background substraction). The angle dependent relative double differential cross-sections were summed up separately for the left and right half-spectrometers (see figure 1 and [6]) for determination of the $\sigma_L$ and $\sigma_R$ experimental cross-sections. Finally, the asymmetry parameter was calculated by using equation 1 and its error was derived from statistical uncertainty, background subtraction, normalization and reproduction.

2.1. Test of the experimental system

In order to reduce the possible effect of retardation, all photoelectrons ejected from the outer s-shells were collected at around the kinetic energy of the Ar $^3P_{0,1,2}$ Auger-electrons (207.0 eV). The deceleration ratios ($E_{\text{kin}}/E_{\text{pass}}$) were varied between 4.83–5.31 during the measurements at 40 eV pass energy ($E_{\text{pass}}$) of the spectrometer. These relatively small variations in the ratios did not affect the measured angular distributions [2]. The collection times of the photoelectrons were some ten seconds and the energy sweeps were repeated 5–50 times depending on the magnitude of the photoionization cross-sections. This procedure also increased the reliability of the measured data. The data were collected as follows: first an Auger-spectrum was recorded for normalization of the photoelectron data, then a photoelectron spectrum was collected and finally an Auger-spectrum was measured again. This scheme was repeated several times.

In order to exclude any possible systematic instrumental effects (mechanical differences of the two parts of the analyzer, effect of the residual magnetic field, etc) the spectrometer and vacuum chamber were rotated by 180°. This means that the left side of the analyzer became the right side and the forward detectors turned into the backward ones and vice versa.

A further test was carried out by measuring the Ar 2p photoelectron line and the Ar LMM Auger peak in the same spectrum. The photon energy of 461.2 eV was used to bring the Auger and photoelectron lines close to each other. Figure 2(a) shows the comparison between the left (circle and black shaded peak area) and right (triangle and gray shaded peak area) side spectra normalized to the corresponding isotropic Auger line and figure 2(b) presents the difference spectrum ($I^L_j - I^R_j$) where $I^L_j$ and $I^R_j$ are the relative intensities in the left and right spectra for the $j$th electron energy. The left–right asymmetry is clearly seen. The values of the asymmetry parameters were found to be $A^{1/2}_{LR} = 0.015(10)$ and $A^{3/2}_{LR} = 0.0094(90)$ for the fine structure components of Ar 2p photoelectrons (the numbers in the brackets show the errors).

As an additional check the asymmetry parameters for the Ar 2p line were also measured by two Scienta analyzers (SES-100 and SES-200) at the beamline I411 at several angles. The angular distribution of the photoelectrons was measured simultaneously by both analyzers (one of them was on the left side and the other one on the right side) and the analyzers were rotated in the dipole plane ((Z-Y)-plane on figure 1) keeping the same angles. The measured asymmetry parameter of $A^{Sc}_{LR} = 0.010(3)$ is in good agreement with the value obtained with ESA–22 spectrometer.

3. Results and discussion

After the above mentioned experimental tests of the whole equipment the asymmetry parameters were measured for the photoionization of H$_2$ molecule and of the noble gases He 1s, Ne 2s, Ar 3s, Kr 4s and Xe 5s during two different beamtimes. In the re-measurements the energy
Figure 2. Comparison between the left (circle and black shaded peak area) and right (triangle and gray shaded peak area) side $2p_{1/2}$ and $2p_{3/2}$ photoelectron spectra of Ar after the normalization for the corresponding area of the $^3P_{0,1,2}$ Auger line (a) and the differences between the left and right side spectra (b).

of the photoelectrons was exactly the same as the energy of the Auger-electrons and the exit slit of the monochromator was 100 µm. The effect of the different slit size on the left–right asymmetry was tested with 3p photoelectrons of Ar. $A_{LR}$ was found to remain constant within 2.5% when the width of the exit slit was changed in the range of 50–200 µm. The values of the measured asymmetry parameters are displayed in figure 3 as a function of atomic mass. The results of the two independent measurements are plotted in the figure (the first one with circles and the second one with squares). The triangles represent the data measured after 180° rotation of the whole equipment. (The solid line is drawn only to guide the eye.) The agreement among the three datasets is very good. The figure clearly shows that the asymmetry parameters differ from zero and increase with decreasing nuclear mass.

As it was mentioned the nonzero value of the asymmetry parameter indicates the breakdown of space inversion symmetry in photoionization. The parity non-conservation (PNC) has been observed in a few cases in atomic physics (see the overview articles [7]–[9] and references therein). The parity violation was measured in the resonant photoexcitation processes of the valence electrons by linearly and circularly polarized laser light. Only a few heavy atoms were examined from the point of view of PNC, namely Cs [10, 11], Tl [12, 13], Pb [14, 15] and Bi [16]. The findings were explained in the frame of the standard model (SM) as the result of the electroweak interaction between the nucleons and atomic electrons mediated by the exchange of the $Z_0$ boson.
The experimental and theoretical s-shell left–right asymmetry parameters as a function of atomic mass for $\text{H}_2$ molecule and for the noble gases from He to Xe. The circles show the first measurement, the squares are the reproduction of data a half-year later and the triangles represent the asymmetry parameters after rotating the whole system by 180°. (The solid line is drawn to guide the eye.) The dashed line and right-hand scale denote the theoretical estimation of the left–right asymmetry parameters.

As far as we know, no experimental investigations and theoretical predictions have been published for the left–right asymmetry in photoionization. Accordingly, we made a simple estimation for the parity non-conserving transition amplitudes in the frame of the SM. We used similar description as Bouchiat and Bouchiat \[17, 18\] for the calculation of the asymmetry parameters for photoionization. A first-order nonrelativistic independent particle model was applied for the theoretical estimation of the left–right asymmetry parameters for photoionization. The initial and final states of atomic electrons were represented by hydrogen-like wavefunctions in both the parity violating ($A_{PV}$) and the parity conserving transition amplitudes ($A_{EM}$). The parity non-conserving interaction potential was reduced only for the spin independent part (see equation (10) in \[7\]) and dipole approximation was applied for the photoionization \[19\]. In this description, the left- and right-hand side cross-sections are $\sigma_{L,R} = |A_{EM} \pm A_{PV}|^2$ and the asymmetry parameter is $A_{LR} = 2Re(A_{PV}/A_{EM})$, when the second-order terms are neglected. The estimation of the left–right asymmetry parameters is plotted on figure 3 as a dashed line (with right side scale). It can be seen that the order of magnitude and the tendency of the calculated data are in contradiction with the measured one. The disagreement indicates that the observed nonzero asymmetry cannot originate from the weak interaction among the atomic electrons and nucleons. This is true especially for the $\text{H}_2$ molecule where the theory predicts a very small value for the asymmetry parameter and the experiment shows a maximum.

There may be another explanation of our results in that the photon package is extremely short. It is an established fact for synchrotron radiation that the length of the photon packet is long in time. Many oscillations of the electric vector occur during the photoabsorption of an atom and the intensity of the photoelectrons emitted to the opposite direction should be identical due to the parity conservation of electromagnetic interaction. The present experimental data are contradiction with this principle. However, a ‘virtual’ breakdown of space inversion
symmetry was observed in multi-photon ionization by an ultra short laser pulse. Paulus and co-workers [20] measured the photoelectrons perpendicular for the momentum vector of the photon in two opposite directions with 6 femtoseconds long laser light from Kr atoms (wavelength \( \lambda = 780 \text{ nm} \) or \( h\nu = 1.59 \text{ eV} \)). A few percent left–right asymmetry (anti-correlation) was observed relative to the photon propagation direction. This finding was explained as a phase effect between the carrier-envelop and the few cycle laser light. The time average value of the oscillating electric vector can differ from zero in the left or right direction depending on the phase difference (see the review paper by Milosević et al [21] for details). The order of magnitude of our asymmetry parameter is approximately same as was observed in Paulus et al’s [20] experiment, so we can extrapolate from their laser pulse length for the time structure of the synchrotron radiation in our measurement. If the present experimental observations originate from the shortness of the photon packet, we get approximately 42 atoseconds duration time for the pulse at 230 eV photon energy. This time is shorter by a factor of \( 10^{-6} \)–\( 10^{-7} \) than expected from traditional synchrotron radiation. We note that the time-dependent description of photoionization by an ultra short (few femtoseconds) single photon produces the same shape for the angular distribution of photoelectrons as a long one, and the asymmetry parameter is zero in this model (see [22, 23]). As far as we know no experimental data and theoretical calculation have been published for the atomic number dependence of the left–right asymmetry in the case of high-energy photoionization processes excited by few tens of attoseconds pulses.

4. Conclusion

In conclusion, a left–right asymmetry has been observed in photoionization created by linearly polarized synchrotron radiation. The experimental left–right asymmetry parameters were determined for photoelectrons ejected from the \( \text{H}_2 \) molecule and from the outer s-shells of noble gas atoms He, Ne, Ar, Kr and Xe. The kinetic energy of the measured photoelectrons was chosen to be close to the energy of the \( (2p)^{-1}\,2^1 P_{1/2} - (3p)^{-2}\,2^3 P_{0,1,2} \) Auger transition of Ar, and all spectra were normalized to this isotropic Auger electron line. The measurements were repeated when the equipment was rotated by 180°. The re-measured data agreed well with the previous ones. Nonzero asymmetry parameters were also obtained for the fine structure components of Ar 2p photoelectrons. The values obtained with two Scienta and \( E\,S\,A-22 \) spectrometers were in good agreement with each other. The observation strongly indicates that the left–right asymmetry is a result of a real physical process and cannot be interpreted as an instrumental effect.

Experimental asymmetry parameters do not increase with increasing nuclear mass. The tendency and the order of magnitude of the measured data do not agree with the predictions for the parity violation process in photoexcitation/photoionization estimated by the SM (see figure 3 and [7]–[9]). The present experimental asymmetry parameters can be explained with the few cycles photon beam but that is in contradiction to the knowledge of the time structure of the synchrotron radiation. We note that normalization with respect to the Auger line can shift the magnitude of the measured asymmetry parameters but it cannot change their dependence on the atomic mass.

The present results cannot be explained with existing models and experiments for breakdown of atomic space inversion symmetry. Further experimental work is necessary to investigate the behavior of the left–right asymmetry in more detail. There is also a need for a theoretical model which could describe the observed behavior.
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