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Energy dependence of the electron spin polarisation parameters for Hg 5d photoionisation with circularly polarised light

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Abstract. Circularly polarised VUV radiation from the storage ring BESSY was used to measure all three spin polarisation parameters of photoelectrons from the Hg 5d shell in the photon energy range from threshold to approximately 35 eV. The experimental results are compared with theoretical calculations in the relativistic and non-relativistic random-phase-approximation scheme (RRPA and RPAE respectively) and Dirac-Slater (DS) calculations. A discussion of the influence of relativistic and potential barrier effects on the experimental data is given.

1. Introduction

In the last decade the experimental determination of the spin polarisation of photoelectrons has become an important probe of the electronic structure of matter.

This paper presents new experimental results for the photoelectron spin polarisation for the outermost d subshell of Hg. All three spin polarisation components were measured in the energy region above the $^2D_{3/2}$ threshold (16.7 eV) up to approximately 35 eV photon energy. The measurements were made at the storage ring BESSY using the circularly polarised synchrotron radiation of the 6.5 m normal incidence monochromator (Schäfers *et al* 1986).

Though intensively studied experimentally and theoretically the low- energy photoionisation dynamics of mercury is far from being understood completely. For the Hg 5d subshell up to now only data of the spin polarisation of the total electron flux A, where the fine structure was not resolved, and of the polarisation component $P_\perp$, perpendicular to the reaction plane described by the parameter ξ, existed (Schönhense *et al* 1982). Those measurements in combination with data for the total cross section measured by Shannon and Codling (1978) and for the differential cross section described by the parameter β (Schönhense 1981) were used to extract 'experimental matrix elements in the LS-coupling scheme (Schönhense and Heinzmann 1984). This analysis revealed strong interchannel interaction between the $5d \rightarrow \varepsilon p$ and $5d \rightarrow \varepsilon f$ channels in the threshold region, which was adequately described only by the RPAE model (including intertransition correlations), while the single-electron (Hartree–Fock and Dirac–Slater) calculations failed to reproduce the experimental data.

Recent extended measurements of the β parameter (McQuaide *et al* 1987) resulted in systematic deviations from former measurements (Schönhense 1981) and from theoretical results. Although it neglects correlation effects, the Dirac–Fock calculation (Tambe *et al* 1981) gives a better agreement with experiment than the eight-channel RRPA calculation of Johnson *et al* (1982). In the threshold region, however, deviations from both theories exist.

In particular, the threshold region is sensitive to the coupling schemes and theoretical approaches used. As pointed out by Keller and Combet-Farnoux (1982, 1985), the potential barrier effect in the εf channel (which is the dominant one) prevails over the spin–orbit effect in the near-threshold region. On the other hand, the influence of the spin–orbit interaction is stronger when it is superimposed on a small cross section. Thus the relativistic effects should be more pronounced in the εp continua (spin–orbit interaction between $\varepsilon p_{1/2}$ and $\varepsilon p_{3/2}$) than in the εf channels ($\varepsilon f_{5/2}$ and $\varepsilon f_{7/2}$). Therefore the process starting from the $j=\frac{3}{2}$ d level should be more sensitive to relativistic effects (since it can couple to the two p continuum channels), while the transition from the $j=\frac{5}{2}$ d level should be dominated by the potential barrier effect.

The new data of the photoelectron's spin-polarisation were taken in order to address these open questions. The data are compared with relativistic and non-relativistic random-phase approximation (RRPA and RPAE respectively) calculations and with Dirac–Slater (DS) results.

2. Experimental

The experiment was carried out at the storage ring BESSY using circularly polarised light delivered by the 6.5 m normal incidence monochromator (Schäfers *et al* 1986). The monochromatic light has a circular polarisation of 92% and a bandwidth of approximately 0.3 nm. Details of the rotatable electron spectrometer system and the Mott detector for spin polarisation analysis are given elsewhere (Heckenkamp *et al* 1986a, b).

The spin polarisation components are described by the energy-dependent parameters A, α and ξ, which are related to the Cartesian components of the spin polarisation vector $A(\theta)$ (component parallel to the photon momentum) and $P_\perp(\theta)$, (component perpendicular to the reaction plane) according to the equations:

$$P_\perp(\theta)=\frac{2\xi\sin(\theta)\cos(\theta)}{1-\frac{1}{2}\beta P_2(\cos(\theta))} \tag{1}$$

$$A(\theta)=\frac{A-\alpha P_2(\cos(\theta))}{1-\frac{1}{2}\beta P_2(\cos(\theta))}. \tag{2}$$

(The reaction plane is spanned by the momenta of the photon and photoelectron, which include the reaction angle θ.)

The denominator represents the differential cross section which is described by the β parameter. Equation (1) is valid for unpolarised or circularly polarised light, equation (2) is valid for completely circularly polarised light only, while $A(\theta)=0$ for linearly polarised or unpolarised light.

For the case of elliptically polarised light the relations are more complex (Heckenkamp *et al* 1986b).

Two components of the spin polarisation vector can be measured simultaneously; the third spin parameter is obtained by a measurement of the angular dependence of $A(\theta)$.

3. Results

The reaction considered is the removal of a d electron from the filled outermost d shell leaving the residual ion according to the spin-orbit interaction in two possible final states. The reaction is described by the following equation:

$$\mathrm{Hg}(5\mathrm{d}^{10}6\mathrm{s}^2)(^1\mathrm{S}_0) + h\nu$$

$$\rightarrow \mathrm{Hg}^+(5\mathrm{d}^9 6\mathrm{s}^2)(^2\mathrm{D}_{5/2}) + \mathrm{e}^-(\varepsilon \mathrm{f}_{7/2}, \varepsilon \mathrm{f}_{5/2}, \varepsilon \mathrm{p}_{3/2}) \qquad (3)$$

or

$$\rightarrow \mathrm{Hg}^+(5\mathrm{d}^9 6\mathrm{s}^2)(^2\mathrm{D}_{3/2}) + \mathrm{e}^-(\varepsilon \mathrm{f}_{5/2}, \varepsilon \mathrm{p}_{3/2}, \varepsilon \mathrm{p}_{1/2}).$$

A photoelectron spectrum obtained at 25 eV photon energy is shown in figure 1. It demonstrates the fine-structure splitting of 1.86 eV between the d thresholds and, with a lower intensity, shows the electrons from the outermost s subshell with a kinetic energy which is 4.4 eV higher. This spectrum was recorded with a monitor channeltron behind the spectrometer using circularly polarised light. Note that the count rates in the Mott detector are three orders of magnitude smaller, so that the accumulation time for only one spin polarisation value is in the order of some hours for a reasonable statistic (statistical error less than 5%) to be achieved.

To determine the spin parameters A and α the spin polarisation component $A(\theta)$ is measured at different emission angles θ (equation (2)). Figure 2 shows the result of such a measurement of $A(\theta)$ for a wavelength of 50 nm and for the $^2\mathrm{D}_{3/2}$ final ionic state (open symbols with error bars).

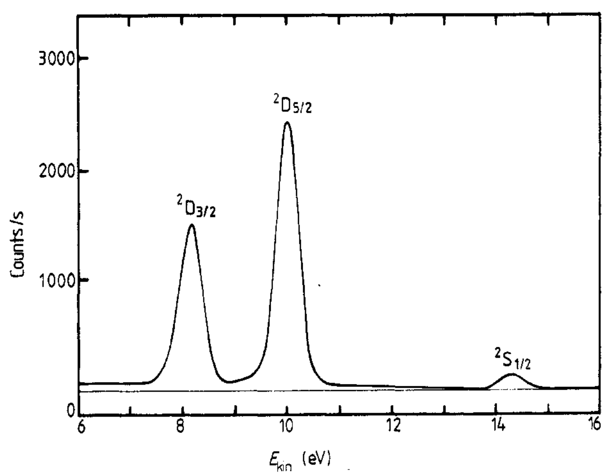

Figure 1. Photoelectron spectrum of Hg ($5\mathrm{d}^{10}6\mathrm{s}^2$) recorded with a monitor channeltron behind the spectrometer using circularly polarised light of 50 nm wavelength. The final ionic states are indicated.

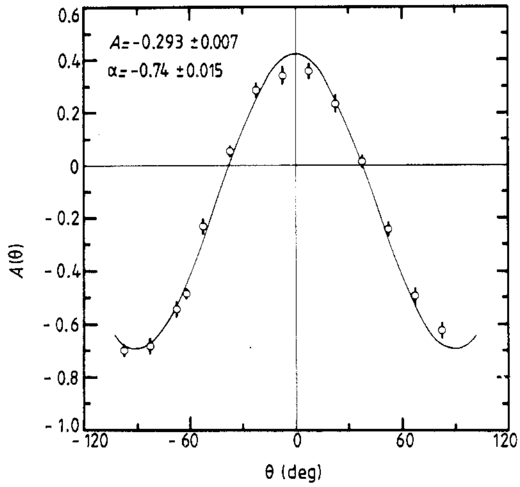

Figure 2. Angular dependence of the spin polarisation component $A(\theta)$ at 50 nm for the Hg^+ $^2D_{3/2}$ final ionic state. A least-squares fit yields the parameters A and α as indicated. The β parameter has also been determined from the least-squares fit (-0.3 ± 0.3).

The fit procedure, however, is not very sensitive to variations of the β parameter and therefore, though in principle possible, not very well suited to obtain accurate β values. It is, however, within the error bars consistent with the literature data for the β parameter ($\beta=-0.05\pm0.08$ (McQuaide *et al* 1987))

The full curve in figure 2 is the result of a least-squares fit of $A(\theta)$ to the experimental values according to equation (2) yielding the spin parameters $A=-0.293\pm0.007$ and $\alpha=-0.74\pm0.015$. Note that

$$A(\theta_{\rm m})=\frac{\int {\rm d}\Omega\ I(\theta)A(\theta)}{\int {\rm d}\Omega\ I(\theta)}=A \tag{4}$$

that is, the polarisation of the total electron flux A can be obtained by a measurement of $A(\theta_{\rm m})$ ($\theta_{\rm m}$ = magic angle 54° 44′), since $P_2(\theta_{\rm m})=0$). The parameter ξ is evaluated by a measurement of $P_\perp(\theta_{\rm m})$ $(=2\ \xi \sin(\theta_{\rm m})=0.943\ \xi)$.

By measurement of such an angular distribution (figure 2) at different photon energies and by evaluation of the parameters A and α for each energy one obtains the energy dependence of A and α.

Figure 3 shows all experimental spin resolved photoelectron spectroscopy data available for Hg 5d. The results are shown for both final ionic states (open symbols: $j=\frac{3}{2}$, full symbols: $j=\frac{5}{2}$); the vertical broken lines represent the D thresholds.

The figure shows the results for the spin parameters α, A and ξ. The circles represent the new set of measurements; the earlier measurements of ξ obtained at resonance lines are indicated by triangles.

The full curves represent the RRPA calculation of Johnson *et al* (1982) (experimental thresholds are used; correlations with the 6s and within the 5d shell are included); the broken curve reproduces the RPAE calculation of Ivanov *et al* (1979) and Cherepkov (1981). The chain curve is a one-channel Dirac-Slater calculation (DS) of Keller and Combet Farnoux (1985). In the non-relativistic approximation, neglecting the spin-orbit interaction in the continuum states, the ratio of the spin parameters is equal to the statistical ratio ($A_{3/2}/A_{5/2}=-1.5$, the same for α and ξ). In accordance with this the results for the two fine-structure components differ in sign.

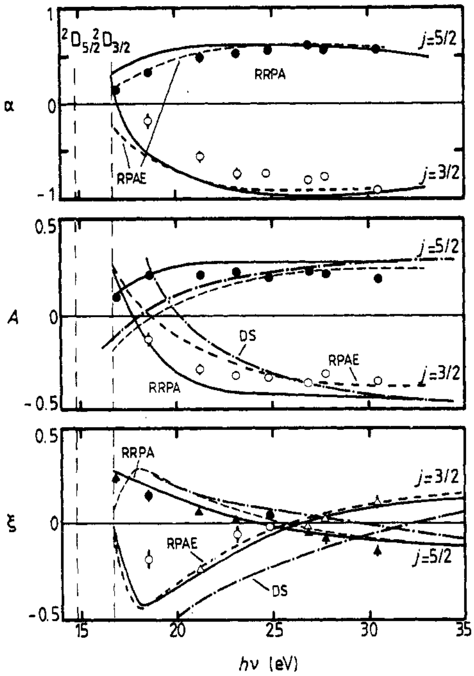

Figure 3. Photoioionisation of Hg 5d. Experimental results for the spin parameters α, A and ξ for the final ionic state $^2D_{5/2}$ (full symbols) and $^2D_{3/2}$ (open symbols). Triangles (for ξ), Schönhense *et al* (1982); full curves, RRPA calculation of Johnson *et al* (1982); broken curves, RPAE calculation of Ivanov *et al* (1979); chain curve, DS calculation of Keller and Combet-Farnoux (1985). The vertical broken lines indicate the ionisation thresholds.

The general energetic trend of the parameters can be explained by the following.

All three parameters are complicated functions of the three transition matrix elements and the relative phases of the continuum wavefunctions. (The quantitative evaluation of the 'experimental' dipole matrix elements and phaseshift differences is in progress and will be published elsewhere.) The parameter ξ is determined mainly by phaseshift differences between p and f waves, which due to the Coulomb part vary rapidly at threshold and change sign at 25 eV.

The A parameter, on the other hand, is determined by the phaseshifts between partial waves with the same l (spin-orbit interaction) which are known to be small and energetically nearly constant (Keller and Combet-Farnoux 1985). Therefore A is influenced mainly by the ratio of the matrix elements, which is maximum in the shape resonance at approximately 20 eV above threshold (Shannon and Codling 1978). This resonance, therefore, also causes the broad maximum of A starting approximately 10 eV above threshold.

All theoretical results reproduce the general energy dependence quite well, but in the threshold region systematic discrepancies remain. The overall agreement of the experimental data is best with the RRPA results, although considerable differences remain. The agreement with the DS results could be improved by introducing experimental thresholds.

In general the agreement for the $j=\frac{5}{2}$ level seems to be slightly better than with the $j=\frac{3}{2}$ level. This seems to indicate indeed that the potential barrier effect which is dominant in the $j=\frac{5}{2}$ level (Keller and Combet-Farnoux 1985) is treated more adequately, while the spin-orbit effects between the outgoing p waves in the $j=\frac{3}{2}$ level are underestimated.

Concluding, in the present investigation the three components of the spin polarisation vector were measured for photoelectrons emitted from the Hg 5d subshell in the energy region from threshold to approximately 35 eV photon energy. In combination with the cross section data and the asymmetry parameter β a parameter set is now available which in a quantum mechanical sense is a complete characterisation of the photoionisation process.

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