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Angle- and spin-resolved photoelectron spectroscopy in the region of the $6s6p^2$ autoionisation of Tl

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Abstract. An angle-, energy- and spin-resolved photoionisation experiment was performed in the region of the $6s6p^2$ autoionisation resonances of thallium. Using monochromatic circularly polarised synchrotron radiation the energy dependence of the spin-polarisation parameters A, ξ and α and the angular asymmetry parameter β of the differential cross section were determined. In the wavelength region investigated these dynamical parameters show a pronounced variation which agrees well with the results of the 'random phase approximation with exchange' calculation by Cherepkov. A detailed discussion of the resonance behaviour for the autoionising states is given in terms of dipole-matrix elements and phaseshift differences which are extracted from the experimental data.

1. Introduction

In the last decade studies aiming at the experimental determination of the energy dependence of the photoelectron spin-polarisation parameters A, ξ and α for free atoms have concentrated on closed-shell atoms such as noble gases (Ar, Kr, Xe) and metals (Ag, Cd, Hg, Yb) (for a review see Heinzmann 1986). Using these parameters and, in addition, the experimental results for the asymmetry parameter β of the differential cross section and the photoionisation cross section σ it was possible to characterise the photoionisation of these atoms in terms of 'experimental' dipole-matrix elements and phaseshift differences (Heinzmann 1980a, b, Schäfers *et al* 1982, Schönhense *et al* 1984, Heckenkamp *et al* 1986b). These evaluations revealed the importance of the spin-orbit interaction in the continuum states; furthermore, strong coupling between different continua has been established.

The results of *ab initio* theories (relativistic random phase approximation (RRPA) (Johnson *et al* 1978, 1979), random phase approximation with exchange (RPAE) (Amusia *et al* 1976) and R matrix (Scott *et al* 1980, 1982)) for the spin-polarisation parameters A, ξ and α of these closed-shell elements are, in general, in good agreement with the measured values.

The experimental data concerning photoelectron spin-polarisation for atomic systems with an open shell are considerably less complete. In earlier investigations on Cs(6s) (Heinzmann *et al* 1970a, b), Tl($6s^26p$) (Heinzmann *et al* 1975, 1976) and Pb($6s^26p^2$) (Heinzmann 1978) only the spin-polarisation parameter A was determined,

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since in these experiments the total photoelectron flux was analysed (the Fano effect) (Fano 1969a, b, Heinzmann *et al* 1970a).

In order to also obtain a complete characterisation of the photoelectron spin-polarisation vector for these systems, we have started to investigate open-shell atoms with one- and two-electron configurations. In this paper we present recent results for the spin-polarisation parameters A, ξ and α and the asymmetry parameter β for the differential cross section in the autoionisation region of Tl($6s6p^2$). In addition, we give a complete analysis of the photoionisation dynamics in terms of dipole-matrix elements and phaseshift differences.

For the theoretical calculation of the spin-polarisation parameters the above mentioned *ab initio* theories are not suitable, since in general these are restricted to closed-shell atoms. Up to now the only calculation available for an open-shell atom was performed by Cherepkov (1980) for Tl in the $6s6p^2$ autoionisation region using a modified version of the RPAE formalism (Cherepkov *et al* 1977). Therefore, a comparison of experimental data with these theoretical results constitutes a sensitive test for this modified theory.

2. Experiment

The measurements were performed with our angle- and spin-resolving apparatus for photoelectron spectroscopy at the electron-storage ring for synchrotron radiation in Berlin (BESSY). Details of the experimental set-up are described by Heckenkamp *et al* (1986a, b). Modifications to the experimental arrangement were made by adding a resistively heated atomic beam oven for the production of the metal vapour. Briefly, circularly polarised VUV synchrotron radiation, emitted off-plane, is dispersed by a 6.5 m normal incidence monochromator ($\Delta\lambda = 0.5$ nm) (Schäfers *et al* 1986) and crossed by an effusive beam of thallium atoms. The reaction plane is spanned by the momenta of photon and photoelectron. The photoelectrons emitted at the emission angle Θ are energy-analysed by a simulated hemispherical electron spectrometer (Heckenkamp *et al* 1986b) rotatable around the normal of the reaction plane. After two electrostatic deflections by 90 degrees the electrons are accelerated to 100 keV and scattered off the thin gold foil of a Mott detector for spin-polarisation analysis (Sherman function: -0.25 ± 0.01 (Kessler 1976)). Two transverse spin-polarisation components, $A(\Theta)$ (the component in the direction of the incident photon beam) and $P_\perp(\Theta)$ (the component perpendicular to the reaction plane), are determined simultaneously. The angular dependence is given by

$$A(\Theta) = \gamma \frac{A - \alpha P_2 \cos\Theta}{1 - \frac{1}{2}\beta P_2 \cos\Theta} \qquad (1a)$$

$$P_\perp(\Theta) = \frac{2\xi \cos\Theta \sin\Theta}{1 - \frac{1}{2}\beta P_2 \cos\Theta} \qquad (1b)$$

where $\gamma \pm 1$ is the helicity of light; A, ξ and α are photoelectron spin-polarisation parameters; β is the asymmetry parameter of the differential cross section and $P_2 \cos\Theta$ is the second Legendre polynomial.

By measuring $A(\Theta)$ and $P_\perp(\Theta)$ at the 'magic angle', $\Theta_m = 54°44'$ (where $P_2 \cos\Theta_m = 0$), one obtains directly the spin-polarisation parameters A and ξ, respectively. The spin-polarisation parameter α and the asymmetry parameter β of the differential cross section are determined from the angular dependence of $A(\Theta)$ (see equation (1*a*)) by a least-squares fit, as described elsewhere (Heckenkamp *et al* 1986b).

3. Experimental results and discussion

Our investigation of Tl($6s^2 6p$) was focused on the wavelength region between 167.0 and 129.0 nm, where the photoionisation is strongly influenced by the excitation of a 6s electron to the $6s6p^2$ configuration. Since the fine-structure splitting of the ground state of thallium is $\Delta E = 0.9$ eV, the population of the Tl$(6s^2 6p)^2P_{3/2}$ level can be neglected at our evaporation temperature of 1000 K. The photoionisation of Tl is then described by the following reaction scheme:

ground state

$\mathrm{Tl}(6s^2 6p)^2P_{1/2} + h\nu(\sigma^+, \sigma^-)$

$\longrightarrow$ autoionisation resonances: $\mathrm{Tl}(6s6p^2)^2S_{1/2}, {}^2P_{1/2,3/2}, {}^2D_{3/2}$ (2*a*)

$\downarrow$ $\Delta J = 0$

$\longrightarrow$ continuum states: $[\mathrm{Tl}^+(6s^2)^1S_0 + e^-(\varepsilon s, \varepsilon d)]_{J=1/2,3/2}$. (2*b*)

We have used the *LS*-coupling scheme in order to compare our results with the RPAE calculation of Cherepkov (1980, 1988). The ionic core of thallium has filled shells, $L = S = J = 0$; thus the total angular momentum J of the final state, ion plus electron (see equation (2*b*)), is equal to the angular momentum j of the outgoing photoelectrons. Therefore, in the region investigated the photoionisation of $\mathrm{Tl}(6s^2 6p)^2P_{1/2}$ can be described by the energy dependence of two real dipole-matrix elements, $D_{1/2}$ ($\varepsilon s_{1/2}$ outgoing partial wave) and $D_{3/2}$ ($\varepsilon d_{3/2}$ outgoing partial wave), and the corresponding phaseshift difference $\Delta = \delta_{1/2} - \delta_{3/2}$. The autoionisation ($\Delta J = 0$) of a $6s6p^2$ level with angular momentum $J = \frac{1}{2}$ or $J = \frac{3}{2}$ takes place only into one of the two orthogonal continua $\varepsilon s_{1/2}$ or $\varepsilon d_{3/2}$, enhancing the $D_{1/2}$ or $D_{3/2}$ dipole transition amplitude, respectively. Assuming pure *LS* coupling with the selection rules for autoionisation (Cowan 1981):

$$\Delta S = \Delta L = \Delta J = 0, \tag{3}$$

only the ^2S and ^2D states can autoionise; however, taking into account the spin-orbit interaction, the ^2P states can autoionise, as well (Karamatskos *et al* 1984).

Figure 1(*a*–*e*) shows the photoionisation cross section σ and the spectral dependence of the photoelectron spin-polarisation parameters A, ξ, α and of the asymmetry parameter β of the differential cross section for $\mathrm{Tl}^+(6s^2)^1S_0$ in the region from 167–129 nm. The full circles are the data of this work ($\Delta\lambda = 0.5$ nm); the results of a previous (angle integrated) measurement for the spin-polarisation parameter A (Heinzmann *et al* 1975, $\Delta\lambda = 3.1$ nm) are indicated by the open circles. The full curves in figure 1(*b*–*e*) for A, ξ, α and β represent the RPAE calculation of Cherepkov (1980) (convoluted to the radiation bandwidth of our experiment ($\Delta\lambda = 0.5$ nm)). The photoionisation cross section σ was determined from the measured photoelectron intensity, recorded at the magic angle (Θ_m) and normalised to the photon flux (grating efficiency, Schäfers *et al* 1986) and to the electron storage ring current. The absolute scaling was performed by the comparison of our relative data to the absolute cross section measurement of Krylov *et al* (1979) in the broad $^2D_{3/2}$ resonance near 160 nm, where the influence of the instrumental resolution is negligible.

In the wavelength region investigated the photionisation process is dominated by three autoionisation resonances (see figure 1(a)). In the range $\lambda = 165$–155 nm lies the broad $Tl(6s6p^2)^2D_{3/2}$ resonance, and at $\lambda = 149$ nm the much narrower $Tl(6s6p^2)^2S_{1/2}$ resonance occurs. The classification of these two resonances with respect to the J values was established by Heinzmann *et al* (1975) using spin-polarisation data. As was first shown by these authors, the measured values of the spin-polarisation parameter A enables the determination of the J value of an autoionising state. This can be seen from the expression of the spin-polarisation parameter A in terms of dipole-matrix elements given in table 1 equation (T3) (Huang 1980, Cherepkov 1983). Assuming that $D_{1/2} \gg D_{3/2} (D_{3/2} \gg D_{1/2})$, the spin-polarisation parameter A approaches a limiting value of $+1.0\,(-0.5)$; thus the sign of the spin-polarisation parameter A reflects the enhanced dipole-transition amplitude and the J value of the resonance can be obtained.

Table 1. Dynamical photoionisation parameters, i.e. cross section σ, asymmetry parameter β for the differential cross section, and spin-polarisation parameters A, ξ and α, as functions of the dipole-matrix elements $D_{1/2}$, $D_{3/2}$ and the phaseshift difference $\delta_{1/2}$-$\delta_{3/2}$ (Huang 1980, Cherepkov 1983) for $Tl(6s^26p)^2P_{1/2}$. α', a_0 and ω are the fine-structure constant, the Bohr radius and the photon energy in atomic units, respectively.

$$\sigma = \sigma_{s_{1/2}} + \sigma_{d_{3/2}} = 4\pi^2 \alpha' a_0 \omega (D_{1/2}^2 + D_{3/2}^2) \qquad \text{(T1)}$$

$$\beta = \frac{D_{3/2}^2 + 2\sqrt{2} D_{3/2} D_{1/2} \cos(\delta_{1/2} - \delta_{3/2})}{D_{1/2}^2 + D_{3/2}^2} \qquad \text{(T2)}$$

$$A = \frac{D_{1/2}^2 - 0.5 D_{3/2}^2}{D_{1/2}^2 + D_{3/2}^2} \qquad \text{(T3)}$$

$$\xi = \frac{3\sqrt{2} D_{1/2} D_{3/2} \sin(\delta_{1/2} - \delta_{3/2})}{4(D_{1/2}^2 + D_{3/2}^2)} \qquad \text{(T4)}$$

$$\alpha = \frac{-D_{3/2}^2 + \sqrt{2} D_{1/2} D_{3/2} \cos(\delta_{1/2} - \delta_{3/2})}{D_{1/2}^2 + D_{3/2}^2} \qquad \text{(T5)}$$

The results for the spin-polarisation parameter A (figure 1(b)) show negative values in the $^2D_{3/2}$ resonance ($\rightarrow J = \frac{3}{2}$), followed by a sharp change to large positive values in the $^2S_{1/2}$ resonance ($\rightarrow J = \frac{1}{2}$) while in the resonance at $\lambda = 130$ nm the values are again negative ($\rightarrow J = \frac{3}{2}$). The classification for the resonance at $\lambda = 130$ nm which has been the subject of discussion in the past ($^2P_{1/2}$ or $^2P_{3/2}$, see Connerade *et al* (1981) and references therein) can therefore be established as $^2P_{3/2}$ in accordance with the Hartree–Fock calculation of Connerade *et al* (1981). The comparison of this measurement for the parameter A with the experimental data of Heinzmann *et al* (1975) ($\Delta\lambda = 3.1$ nm) shows good agreement in the range of the broad $^2D_{3/2}$ resonance. Due to the different radiation bandwidth used, the two sets of experimental data differ in the $^2S_{1/2}$ resonance. Nevertheless, details such as the strong asymmetry of this resonance (not seen in the photoionisation cross section σ, see figure 1(a)) are revealed by the

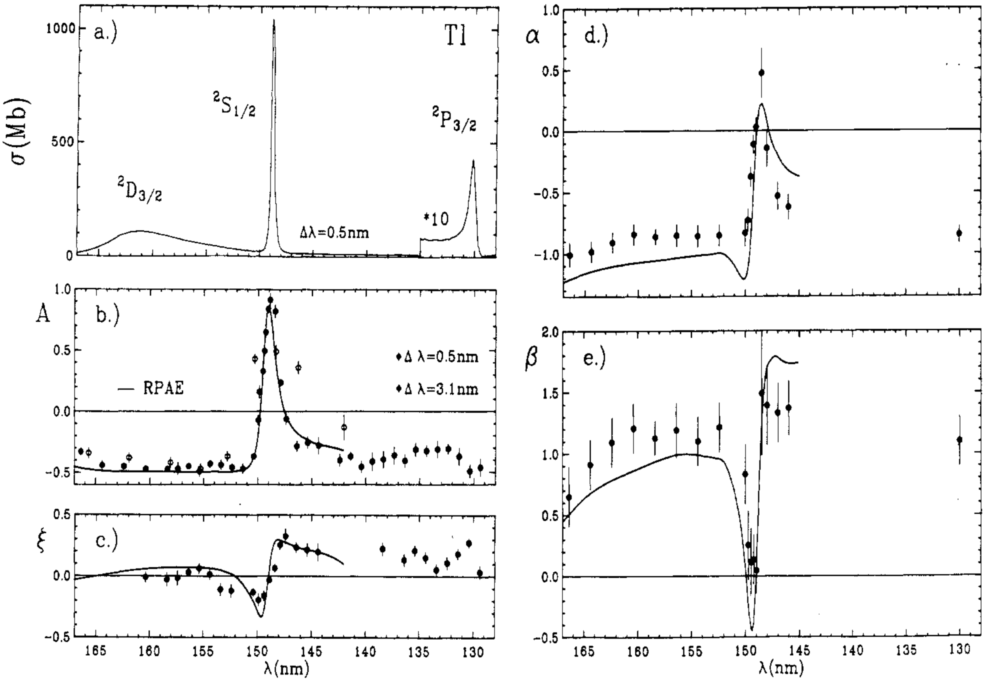

Figure 1. Measured photoionisation cross section σ of $Tl(6s^26p)^2P_{1/2}$ in the autoionisation region of the $6s6p^2$ configuration together with the experimental results (full circles) of the spin-polarisation parameters A, ξ and α and the asymmetry parameter β of the differential cross section. Open circles (for A): Heinzmann *et al* (1975), full curve in A, ξ, α and β: RPAE calculation of Cherepkov (1980, 1988), convoluted to the radiation bandwidth of the experiment ($\Delta\lambda = 0.5$ nm).

measurement at $\Delta\lambda = 3.1$ nm (see also Heinzmann and Kessler 1978, and Cherepkov 1983). The RPAE results for the parameter A in the range of the $^2D_{3/2}$ and $^2S_{1/2}$ resonance agree very well with the experimental values; in particular, the calculation shows the same asymmetry and resonance width for the $^2S_{1/2}$ state. Since the RPAE calculation (Cherepkov 1980) is performed in the framework of pure LS coupling, the autoionisation of the $^2P_{3/2}$ resonance is not considered (see equation (3)). A sensitive test of the RPAE calculation is possible by the ξ parameter. From equation (T4) of table 1, it follows that this parameter is very susceptible to phaseshift variations and sign changes of the dipole-matrix elements. Usually these quantities are strongly affected by an autoionisation resonance as shown by experimental and theoretical studies (Schäfers *et al* 1982, Schönhense *et al* 1984, Heckenkamp *et al* 1986, Fano 1961). On the left wing of the $^2S_{1/2}$ resonance the measured values of the ξ parameter in figure 1(c) are negative ($\xi \simeq -0.2$), while at the resonance position a rapid change to positive values ($\xi \simeq +0.35$) occurs. For the $^2P_{3/2}$ resonance a reverse trend occurs. For the broad $^2D_{3/2}$ resonance we observe only a weak variation of the ξ parameter, the measured ξ values are small and at $\lambda \simeq 158$ nm a change of sign is indicated. The RPAE results ($^2S_{1/2}$ and $^2D_{3/2}$ resonance) for the ξ parameter agree very well with the experimental data as seen best for the $^2S_{1/2}$ resonance, while in the range of the $^2D_{3/2}$ resonance the values are slightly different, in particular, the change of sign takes place at $\lambda = 165$ nm.

The fitted values of the angular dependence $A(\Theta)$ for the α and β parameters (see figure 1(*d*, *e*) are of the order of -1 and $+1$, respectively, in the wavelength region of $\lambda = 166$–152 nm. These data reflect the dominance of the $D_{3/2}$ amplitude as seen by equations (T2) and (T5) of table 1. The variation of these parameters in the $^2S_{1/2}$ resonance is caused by the enhancement of the $D_{1/2}$ amplitude and by the phaseshift variation (see below). The RPAE results for the α and the β parameters show the same dependence, with values slightly smaller than the experimental data.

In the $^2S_{1/2}$ resonance at 148.9 nm the spin-polarisation parameter A was measured to be 0.92 ± 0.05 and the parameters α, ξ and β were found to be zero within the experimental error limits. This means that here a particular limiting case of complete photoelectron spin-polarisation is approached where the length of the polarisation vector is close to unity at all emission angles (see equation (1*a*) and Böwering *et al* 1990). The limiting case is not fulfilled exactly since there is also a small but non-vanishing $D_{3/2}$ amplitude at this photon energy (see also below).

From the data for the dynamical parameters σ, A, ξ, α and β the three unknown quantities $D_{1/2}$, $D_{3/2}$ and $\delta_{1/2} - \delta_{3/2}$ were determined using their analytical dependence as given in table 1. Since for the photoionisation of $Tl(6s^2 6p)^2P_{1/2}$ three suitably measured parameters are already sufficient to determine the matrix elements and the phaseshift difference, the analysis served to check the consistency of the experimental data. The results of our evaluation of the dipole-matrix elements and phaseshift difference are shown in figure 2 as function of the wavelength (full circles) together with the data of the RPAE calculation (full curve) (Cherepkov 1988). We have plotted the quantum-defect differences $\mu_{1/2} - \mu_{3/2}$ as a measure for the phaseshift difference. The relation of the quantum defect μ_i to the phaseshift δ_i is given by the equation

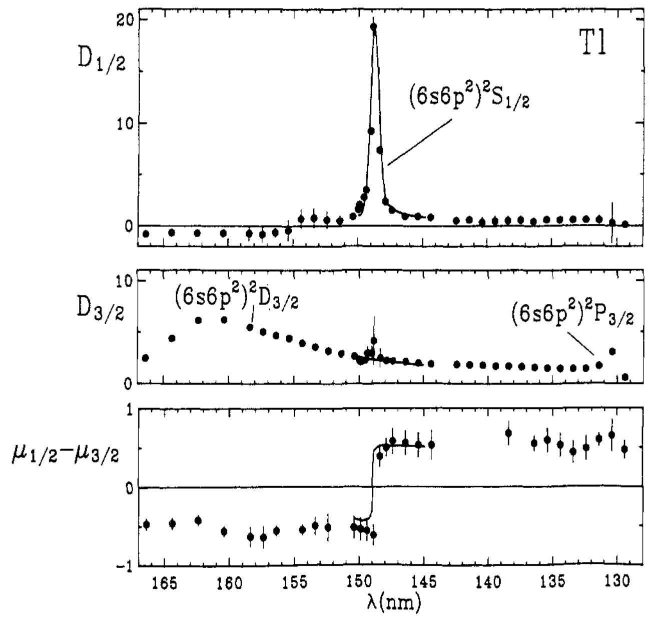

Figure 2. Dipole matrix elements $D_{1/2}$ and $D_{3/2}$ and quantum-defect difference $\mu_{1/2} - \mu_{3/2}$ for photoionisation of $Tl(6s^2 6p)^2P_{1/2}$ as function of the wavelength in the $6s6p^2$-autoionisation region. The full curve represents the RPAE calculation of Cherepkov (1980, 1988), convoluted to the radiation bandwidth used in the experiment ($\Delta\lambda = 0.5$ nm).

(Lee 1974):

$$\delta_i = \sigma_l + \pi\mu_i - \pi l/2. \tag{4}$$

σ_l is the Coulomb phaseshift for an outgoing partial wave with angular momentum l in a pure Coulomb field. Deviations of the Coulomb field are incorporated by the additional phaseshift $\pi\mu_i$; the term $\pi l/2$ satisfies the sign convention of the matrix elements.

From the wavelength dependence of the matrix elements $D_{1/2}$ and $D_{3/2}$ (upper and middle parts of figure 2) the resonance behaviour of the autoionising states is clearly demonstrated. The values for the $D_{1/2}$ amplitude are strongly enhanced in the region of the $^2S_{1/2}$ resonance, whereas they remain unaffected in the region of the $^2D_{3/2}$ and $^2P_{3/2}$ resonance. A sign change for the $D_{1/2}$ amplitude occurs at about 155 nm as also indicated by the measured ξ parameter. The comparison with the available RPAE results (Cherepkov 1988), convoluted with our radiation bandwidth of $\Delta\lambda = 0.5$ nm shows excellent agreement.

The data for the $D_{3/2}$ matrix elements reflect the dominance of the broad $^2D_{3/2}$ autoionisation resonance which affects the whole energy range investigated. It is clearly seen that for the $\varepsilon d_{3/2}$ continuum we have the case of an overlapping resonance structure (Mies 1968), since the $^2P_{3/2}$ resonance is situated at the right wing of the broad $^2D_{3/2}$ resonance. Therefore, a reasonable parametrisation of the cross section σ in terms of Fano profiles (Fano 1961) is only possible with the knowledge of the partial continuua $\varepsilon s_{1/2}$ and $\varepsilon d_{3/2}$. Furthermore, one has to use the extension of the work of Fano (1961) given by Mies (1968). This is in contrast to the evaluation of Krylov *et al* (1979). As was the case for $D_{1/2}$ dipole-matrix elements, the available RPAE data (Cherepkov 1988) for $D_{3/2}$ are in very good agreement with the experimental values. It is worth noting, that the agreement between theory and experiment within the given error bars is excellent even for $D_{3/2}$ at the $^2S_{1/2}$ resonance energy (148.9 nm), where the strong $D_{1/2}$ contribution influences the length of the error bars for $D_{3/2}$.

The data evaluated for the quantum defect difference $\mu_{1/2} - \mu_{3/2}$ (lower part of figure 2) show a change from negative to positive values in the region of the $^2S_{1/2}$ resonance. The difference $\Delta(\mu_{1/2} - \mu_{3/2})$ of the values to the long and short wavelength side of the $^2S_{1/2}$ resonance is of the order of 1 demonstrating a phaseshift variation by π (see equation (4)) in accordance with Fano's theory for the interaction of an isolated state with a single continuum channel (Fano 1961). A similar behaviour is given by the RPAE results of Cherepkov (1988).

From the absolute value of the experimental quantum defect differences $|\mu_{1/2} - \mu_{3/2}|$ which is nearly constant ($\simeq 0.5$) in the energy range investigated, it is obvious that the phaseshift variation is caused only by the energy dependence of the Coulomb phase. Moreover, the 'constant' quantum defect difference confirms the basic idea of quantum defect theory (QDT) (Seaton 1966a, b), which predicts a slowly varying quantum defect μ_i with respect to the photon energy.

4. Conclusions

In the present investigation a complete characterisation is given for the photoionisation of the open-shell atom $Tl(6s^26p)$ in the autoionisation region of the $6s6p^2$ configuration. Dipole-matrix elements and quantum defect differences were determined from the experimental data of the photoelectron spin-polarisation parameters A, ξ, α, the angular asymmetry parameter β and the photoionisation cross section σ. In the case of the $^2S_{1/2}$ resonance the experimental results show good agreement with Fano's theory for an isolated state interacting with one continuum. The experimental data for the $\varepsilon d_{3/2}$ continuum demonstrates clearly the overlapping resonance structure for the $^2D_{3/2}$ and $^2P_{3/2}$ resonance. The comparison of the experimental results with the data of the extended RPAE theory for open-shell atoms shows that the shape and energy dependence of the resonances are in general well reproduced, despite some minor discrepancies. A possible reason for these discrepancies can be attributed to the fact that the RPAE calculation is performed in pure *LS* coupling, neglecting mixing amplitudes of the autoionisation resonances caused by the spin-orbit interaction (Karamatskos *et al* 1984).

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