Spin-resolved negative ion resonances in zinc near 11 eV

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Abstract. Negative ion resonances overlapping the ionisation continuum were studied by spin-polarised electron impact on zinc atoms. The measured integrated Stokes parameters of the 636.2 nm and 468.1 nm photons from the respective 3\textit{d}\textsuperscript{10}4\textit{s}\textsuperscript{4}3\textit{d}\textsuperscript{1}D\textsubscript{2} and 3\textit{d}\textsuperscript{10}4\textit{s}\textsuperscript{5}3\textit{s}\textsuperscript{1}S\textsubscript{1} states show Fano-equivalent profiles reflecting the effects of spin-orbit interaction and electron exchange in the resonant states which have an open 3\textit{d} core shell.

1. Introduction

Zinc, unlike helium, has closed electron shells below the outer 4\textit{s}\textsuperscript{2} shell, which lead to changes of the state structure, and electron correlation effects are expected to play a major role in the scattering. The importance of these configuration correlations on the scattering is the main motivation for the present experiment.

Resonances and more generally interference effects are common processes occurring in almost every area of physics. Characteristic resonance profiles have been observed, for example, in cold-atom Efimov states [1] in nuclear physics or in the study of ferromagnetic resonances in thin films which have shown a clear Suhl threshold [2] for the spin-wave instability effects. In atomic and molecular physics the study of the negative ion resonances has been restricted mainly to rare-gas atoms and alkalis, as reviewed in [3]. It has been shown how the interference of the short-lived negative ion states with the excited or ionised atomic states can affect significantly the scattering cross-sections at the resonant energies. For example, the two negative ion resonances in helium near 58 eV causing classical Fano profiles [4] in the cross-sections have been studied extensively in transmission and energy-loss experiments [3] or in the studies of fluorescence of excited states [5, 6, 7].

The previous studies of resonances in zinc are restricted to the measurements of the optical excitation functions [8, 9] or the electron transmission spectroscopy [10, 11] which were mainly concerned with the classifications of the resonance states below the ionisation level with the 3\textit{d}\textsuperscript{10}4\textit{s} grandparent state configurations, i.e. the resonances with a closed inner 3\textit{d}-shell. However, in zinc the opening of the 3\textit{d}-shell can cause a significant core-valence correlation which in turn can affect the spin-dependent interactions [12]. This was also confirmed by observation of a breakdown in \textit{LS}-coupling for the 3\textit{d}\textsuperscript{2}4\textit{s}\textsuperscript{2}2\textit{D}_{3/2} state [13]. The fundamental aspects of spin-dependent electron scattering, developed by [14, 15, 16], describe a fine interplay of the
Figure 1. The apparatus showing source of polarised electrons, zinc oven and detection assembly.

electron exchange, i.e. relative orientation of the electron spins of incident and target electrons, and spin-orbit interaction, either within the target or the continuum electron. For example, in photoionisation the $\beta$ parameter, which depends on the electron couplings and correlations and on the phase differences between excitation amplitude, the departure from the value of 2 is a sign of spin-orbit interaction. Significant asymmetric Fano profiles for the linearly polarised photon impact on randomly oriented molecules have been demonstrated [17]. Moreover, it was shown how the terms for continuum, resonance and interference between them, which according to the Fano’s approach [18] can be used to describe a resonance effect on a cross-section, have a different angular asymmetry $\beta$ parameter.

The main aim of the present study is to investigate the spin-dependent effects for several negative-ion resonances overlapping the ionisation continuum in zinc near 11 eV. Integrated Stokes parameters were observed for the 636.2 nm and 468.1 nm decay radiation from the $4d\,^1D_2$ and $5s\,^3S_1$ states, respectively, after spin-polarised electron impact. The observed resonance profiles were fitted with the Fano-equivalent Shore parameters [19] and from the fits the resonance energies and widths were determined. The spin-dependent $P_2$ and $P_3$ Stokes parameters enabled analysis of the spin-dependent interactions in the negative ion states.

2. Experimental Technique

The apparatus and the experimental geometry are shown in Figure 1. The detailed description was given previously [20]. In summary, transversely polarised electrons with momentum along $z$ axis and spin along $y$ axis impact zinc atoms produced by a resistively heated oven. Photons emitted after excitation of zinc atoms are detected along the $y$ axis and their polarisation is analysed using a combination of liquid crystal variable retarder and a linear polariser. A particular wavelength is selected by an interference filter. From the observed intensities the integrated Stokes parameters [21] are determined as $P_1 = [I(0^\circ) - I(90^\circ)]/[I(0^\circ) + I(90^\circ)]$, $P_2 = [I(45^\circ) - I(135^\circ)]/[I(45^\circ) + I(135^\circ)]$ and $P_3 = [I(\sigma^+) - I(\sigma^-)]/[I(\sigma^+) + I(\sigma^-)]$. Here $I(\beta)$ is the intensity of photons with a polarisation vector at an angle $\beta$ with respect to the $z$ axis, and $I(\sigma^+)$ and $I(\sigma^-)$ are, respectively, the intensities of photons with positive and negative helicity.
Scattered electrons are not observed. For $P_1$ measurements the incident electron resolution was 180 meV and 200 meV for the $^1D_2$ and $^3S_1$ transitions respectively. $P_2$ and $P_3$ were measured with 250 meV incident electron resolution and 66% polarisation.

While unpolarised electron impact can result in non-zero values of $P_1$, $P_2$ and $P_3$ are spin-dependent parameters. For incident transversely polarised electrons non-zero values of $P_2$ can be caused only by a significant spin-orbit interaction. For a light atom like zinc ($Z=30$) the spin-orbit interaction between the continuum electron and the target is negligible and the non-zero values of $P_2$ will be mainly the result of the spin-orbit interaction within the atom. Furthermore, non-zero values of $P_2$ can be caused if a well LS-coupled excited state is populated by a non LS-coupled intermediate states, for example via cascades or via negative ion resonances. For the latter, if the lifetime of the resonance state is comparable or longer than the fine structure relaxation time the spin of the temporary captured incident electron may be rotated by the magnetic field and hence $P_2$ may be non-zero. The non-zero values of $P_3$ can be caused by two processes, the spin-orbit interaction discussed above and by electron exchange which causes angular momentum to be transferred parallel to the incident spin vector.

Hence the measurements of the integrated Stokes parameters $P_2$ and $P_3$ allow disentanglement of the spin-dependent interactions from the usually much stronger Coulomb interactions and often from each other, not only in the excited states but also in the resonance states [22].

3. Results and Discussion

The measured Stokes parameters and the Fano-equivalent fits to the polarisations are shown in Figure 2 for the $^1D_2$ and $^3S_1$ transitions. The justification of the fitting of Fano-equivalent profiles directly to the polarisations is given in [5]. For the $^1D_2$ transition, $P_1$ and $P_3$ clearly show two resonances superimposed on a polarisation background while $P_2$ only shows the lower energy resonance. For the $^3S_1$ transition $P_1$ shows two structures on a polarisation background while in $P_3$ there are possibly two unresolved resonances with the same sign. The lower energy resonance possibly appears as a tail on the larger higher energy structure. The resonance contributions to $P_2$ for this transition do not indicate clearly presence of resolved structures as in other Stokes parameters. However, the scatter of the points may indicate the possibility of resonant structures whose observation requires a better statistical accuracy. Interestingly, although the energy resolution of the incident electrons did not allow the resonances to be resolved in the photon excitation functions for $P_2$ and $P_3$ (not shown here) the polarisation measurements clearly show two structures.

The first part of the discussion of resonances is in terms of likely configurations and coupling schemes. The average energies and widths extracted from the fits of the Fano profiles to the polarisations are $10.98 \pm 0.02$ eV and $11.33 \pm 0.02$ eV respectively for the $^1D_2$ transition and $10.92 \pm 0.02$ eV and $11.40 \pm 0.03$ eV respectively for the $^3S_1$ transition. Assuming that the additional electron is bound in the field of a known excited state with a negative electron affinity of up to approximately 0.5 eV, the resonance energies can be compared with the calculated centre-of-gravity energies [23, 3] of states from the nearby configurations [24]. The most probable resulting configuration of the parent state is then $3d^34s^24p$. For the resonances observed in the $^1D_2$ channel it is also possible to propose $J = 5/2$ and $J = 3/2$ configurations of the $3d^34s^2$ ion core for the lower and higher energy resonances respectively. The spin-orbit splitting of 0.337 eV for this ion core [24] is in agreement with the measured energy difference of $0.36 \pm 0.02$ eV.

It should be noted that the present data indicates that the two resonances observed in two different transitions are not the same. The resonant energies appear to be different and the energy differences between the lower and higher resonances are different for the two transitions. This is confirmed by the spin-dependent analysis given below. Previous calculations [24] show a large number of states with the $3d^34s^24p$ configuration and, additionally, our high energy resolution (120 meV) scans of electron and photon excitation functions of $4p \ ^3P$ states [25]
Figure 2. The integrated Stokes parameters $P_1$, $P_2$ and $P_3$ as a function of incident electron energy. (a), (b), (c) - $^1D_2$ transition, (d), (e), (f) - $^3S_1$ transition. The full line is a fit to the Fano profile, the dotted line shows the fit to the polarisation background. All error-bars are one standard deviation.

clearly indicate at least three resonances with an asymmetric tail on the high energy side of the highest energy resonance possibly indicating further unresolved structures.

However, the real advantage of the measurement of the integrated Stokes parameters using incident polarised electrons lies in the possibility of the direct observation of the spin-dependent interactions via $P_2$ and $P_3$ in the excited and also in the resonance states. For example only the lower resonance observed in $^1D_2$ transition has non-zero values of $P_2$ indicating spin-orbit interaction in this resonance state. In physical terms this means that the electron charge cloud of the resonant state is rotated in the $xz$ scattering plane. Immediately after a collision the spin is oriented along the $y$ direction but over the fine structure relaxation time of the state the final charge cloud tilts in the $xz$ plane under the influence of the atomic magnetic field after which
it emits a photon. Due to interference between the resonant and direct excitations the charge cloud of the direct excitation is also rotated and this effect can be observed in different channels. The present measurements of $P_2$ indicate that only one of the observed resonance states is not well $LS$-coupled. Furthermore, the $P_3$ parameter for the $1D_2$ transition shows two resonances with opposite signs. For the lower energy resonance the negative sign is indicative that the spin-polarisation vector $\vec{P}$ of the incident electron and $P_3$ have different signs which, for the $1D_2$ state, can be caused only by the spin-orbit interaction. This is consistent with the observation of non-zero $P_2$. On the other hand, the higher energy resonance is dominated by electron exchange, demonstrated by positive sign in $P_3$ and vanishing $P_2$. For the $3S_1$ transition $P_3$ is positive which, similarly for the $1D_2$ state, indicates the same orientation of the spin vector of the incident electron and $P_3$. Furthermore, $P_3$ shows one wide structure with a peak at the same energy as the higher energy resonance in $P_1$. The asymmetric shape of the structure with a wing on the lower energy side indicates unresolved structure with the lower energy resonance having weaker electron exchange than the higher energy resonance.

The origin of the spin-orbit interaction in the lower resonance state observed in the $1D_2$ channel comes most likely from the $3d^94s^24p^2$ configuration with an open $3d^9$ shell. The spin-orbit interaction in zinc was previously linked to states with open $3d^9$ shell [13]. Furthermore, the structure calculations of the states with $3d^94s^24p^2$ configuration show strong configuration mixing [24] in some of the states which is caused by the spin-orbit interaction.

4. Conclusion

In conclusion, Fano profiles caused by resonances were observed in the polarisation of decay photons in zinc near 11 eV. The spin-polarised incident electrons enabled exploration of the weak spin-dependent interactions in those negative ion complexes for the first time. The measured integrated Stokes parameters for the $1D_2$ and $3S_1$ states identified spin-orbit interaction only in one of the resonance states while electron exchange was dominating the others. Furthermore, the Stokes parameters offered a significant advantage over excitation functions which did not resolve the resonances due to insufficient resolution of the incident electron beam. The present experiment has identified $3d^94s^24p^2$ as the most probable configuration of the resonances and that different resonances with similar energies were observed in the two exit channels.

While the present experiment answers questions about possible configuration, coupling scheme, the effects of electron exchange and spin-orbit interaction and angular momentum transfer mechanisms, several questions remain open. The obvious ones concern, for example, unresolved and unobserved resonances, different exit channels, the exact configurations of the resonances, the effects of the cascades on the resonance polarisations and most importantly the origin of the spin-dependent interaction in the resonance states. Future work will include observation of these resonances in different exit channels as well as in angular differential modes.

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