J. Phys. B: At. Mol. Opt. Phys. 26 (1993) L445–L450. Printed in the UK

LETTER TO THE EDITOR

State-selective study of the direct double photoionization of the Ne valence shell

K-H Schartner†, G Mentzel†, B Magel†, B Möbus†, A Ehresmann‡, F Vollweiler‡ and H Schmoranzer‡

† I. Physikalisches Institut der Justus-Liebig-Universität, Heinrich-Buff-Ring 16, 6300 Giessen, Federal Republic of Germany

‡ Fachbereich Physik der Universität, Erwin-Schrödinger-Straße, 6750 Kaiserslautern, Federal Republic of Germany

Received 21 April 1993, in final form 28 May 1993

Abstract. Cross sections for double photoionization of the Ne L shell into the $2s2p^5\,{}^3P^o$ and ${}^1P^o$ and the $2s^02p^6\,{}^1S^e$ states were measured for energies from threshold up to 150 eV, using photon induced fluorescence spectroscopy. Both $2s2p^5$ channels were observed with comparable magnitude in contradiction to a propensity rule based on the Wannier–Peterkop–Rau theory. A comparison of the summed ${}^3P^o$ and ${}^1P^o$ cross sections with MBPT calculations results in a deviation of 50%.

Direct double photoionization (DPI) as the simultaneous ejection of two electrons results from interelectron interactions. While in earlier work predominantly electron emission from different shells was studied which was modelled by core relaxation, recent efforts concentrated on the simultaneous emission from the same shell, especially from the valence shell of the rare gases (Huetz *et al* 1991, Schmidt 1992 and references therein). From these experiments and the corresponding calculations, a deeper understanding of the electron correlation is expected (Amusia 1981). This holds especially for experiments in the threshold range of the direct DPI (Kossmann *et al* 1988), for experiments which study the angular correlations of the emitted electrons (Mazeau *et al* 1991) or which are able to discriminate between different states of a double ionized atom (Krässig and Schmidt 1992 and references therein). A prominent example of the latter case is the ground state configuration of Ar^{2+} or Xe^{2+} which splits into the ${}^3P^e$, ${}^1D^e$ and ${}^1S^e$ states. First investigations of the relative population of these states were carried out by Lablanquie *et al* (1987), followed by experiments by Price and Eland (1989). The interest in the population of different levels of one configuration resulted from the Wannier–Peterkop–Rau theory (Wannier 1953, Peterkop 1971, Rau 1971) which seemed to favour the ${}^3P^e$ state by kinematically preferred partial waves of the two outgoing electrons. The application of this theory in the form of classical arguments combined with symmetry aspects of quantum mechanical wavefunctions was already criticised in the discussion of the experimental results by Price and Eland (1989, 1990). Meanwhile it has been realized that for the discussion of a state selectivity the Wannier–Peterkop–Rau theory should be regarded as a weak propensity rule (Huetz *et al* 1991). These recent interpretations are also of interest for our experiments on the DPI of Ne into the $2s2p^5\,{}^3P^o$ and ${}^1P^o$ states and moreover also on the DPI of two 2s electrons into the $2s^0\,2p^6\,{}^1S^e$ state. On the basis of the mentioned symmetry aspect of the wavefunction of the escaping two electron, the ${}^1P^o$ state should be populated stronger than the ${}^3P^o$ state.

0953-4075/93/150445+06$07.50 © 1993 IOP Publishing Ltd

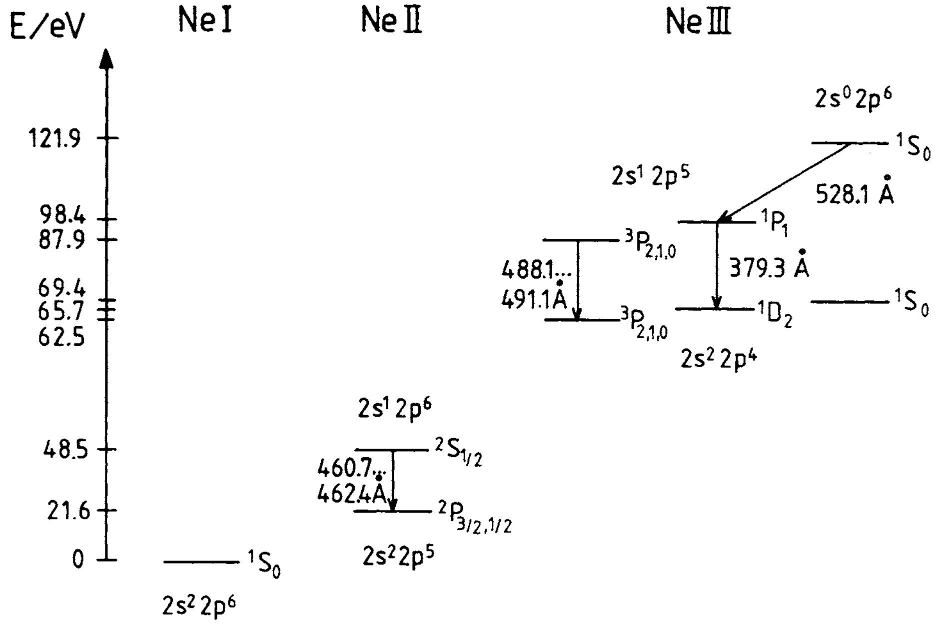


Figure 1. Term diagram of Ne, Ne^{+} and Ne^{2+} containing the relevant energy levels and transitions.

In the present experiment the DPI was followed for the first time from threshold on up to 150 eV exciting photon energy. The photon induced fluorescence spectroscopy was applied (Schartner *et al* 1990), and absolute total cross sections were measured. First results for the respective states of the $3s3p^5$ configuration of Ar^{2+} obtained with this technique were already published (Magel *et al* 1990, Schartner 1990) showing no preferential population of the $^1P^o$ state near threshold. This is in contrast with the result of a recent study using threshold photoelectron coincidence spectroscopy which is in agreement with the prediction of the propensity rule (Hall *et al* 1992a, b). For the Ne^{2+} $2s2p^5\,^1P^o$ and $^3P^o$ states these authors found relative signals of comparable magnitude at threshold, again opposite to the Ar case.

The threshold energies of the three considered states amount to 87.9 eV, 98.4 eV and 121.9 eV for the $^3P^o$, $^1P^o$ and $^1S^e$ state, respectively. They are shown in figure 1 which also contains the transitions from these states observed in fluorescence. Their wavelengths are 49.0 nm, 37.9 nm and 52.8 nm. Additionally, figure 1 shows the $2s2p^6\,^2S_{1/2}$–$2s^22p^5\,^2P_{1/2,3/2}$ doublet at 46.1/46.2 nm. The $^3P^o$–$^3P^e$ multiplet was not resolved in the present experiment.

The ionizing photons were provided by the synchrotron radiation facility BESSY I at Berlin. An undulator followed by a toroidal grating monochromator was used. The experimental set-up consists in its main components of a differentially pumped target cell connected to a specially constructed entrance slit unit of a 1 m normal incidence 225 McPherson monochromator. The exit slit unit was replaced by a position sensitive channel plate detector. Its two-dimensional read-out ability is based on a wedge and strip anode. The ionizing photons were detected by the secondary electron emission of an Al anode. The target pressure amounted to 10 μbar. The photon beam could be replaced by an electron beam, used for determination of the relative detection sensitivity of the monochromator–detector combination.

A set of spectra is displayed in figure 2, registered for discrete increasing photon

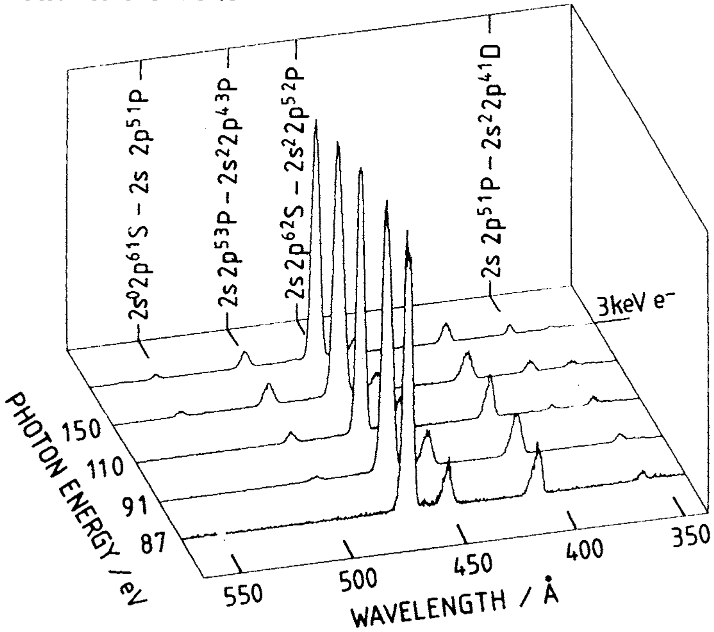


Figure 2. Fluorescence spectra for selected energies of the ionizing photons, scaled to equal height of the 46 nm line. 3 keV electron impact excitation spectrum shown for comparison.

energies. The single spectra are scaled to equal height of the strongest line at 46.1/46.2 nm resulting from the single 2s-electron photoionization. A spectrum for 3 keV electron impact was added for comparison. The spectra are not corrected for the detection sensitivity. Below the thresholds for DPI into the considered channels, satellites appear additionally to the 2s main line. With increasing photon energy the thresholds for DPI into the here relevant states at 87.9 eV, 98.4 eV and 121.9 eV are passed sequentially with more lines appearing accordingly.

Cross sections were derived by normalization onto the single 2s-electron photoionization cross section from Wuilleumier and Krause (1979). The relative detection sensitivity which was needed in this procedure was obtained from the 3 keV electron impact spectrum in connection with published cross sections for electron impact induced line radiation. For the $2s2p^5\,{}^3P^o$ state and the single 2s-electron ionization, the corresponding cross sections are known with a very good uncertainty of 7% (Jans 1992). For the ${}^1P^o$ state and the double 2s-electron ionization the mean values of the cross sections determined by Flaig *et al* (1983) and by Eckhardt and Schartner (1983) were used. These cross sections are quoted to be uncertain within 25%. Consequently, uncertainties of 20% and 50% result for the presented ${}^3P^o$, ${}^1P^o$ and ${}^1S^e$ photoionization cross sections, respectively.

The measured cross sections for state selective direct DPI are shown as a function of the photon energy in figures 3(*a*) and 3(*b*). The different symbols mark independent experimental runs documenting a good reproducibility. The bandwidth of the ionizing photons was 0.5 eV. The larger scattering of the ${}^1P^o$ data is due to low count rates caused by the small detection sensitivity of the normal incidence spectrometer at 37.9 nm. The signals were obtained by subtracting the background from the total counts within the ${}^1P^o$ line. Due to the low statistics also negative signals resulted especially below threshold. As mentioned above, the cross sections were derived by normalization onto the signal for the 2s-electron ionization and the respective cross section. Its value for each investigated photon energy was taken from a mean curve through the data points summarized by Wuilleumier and Krause

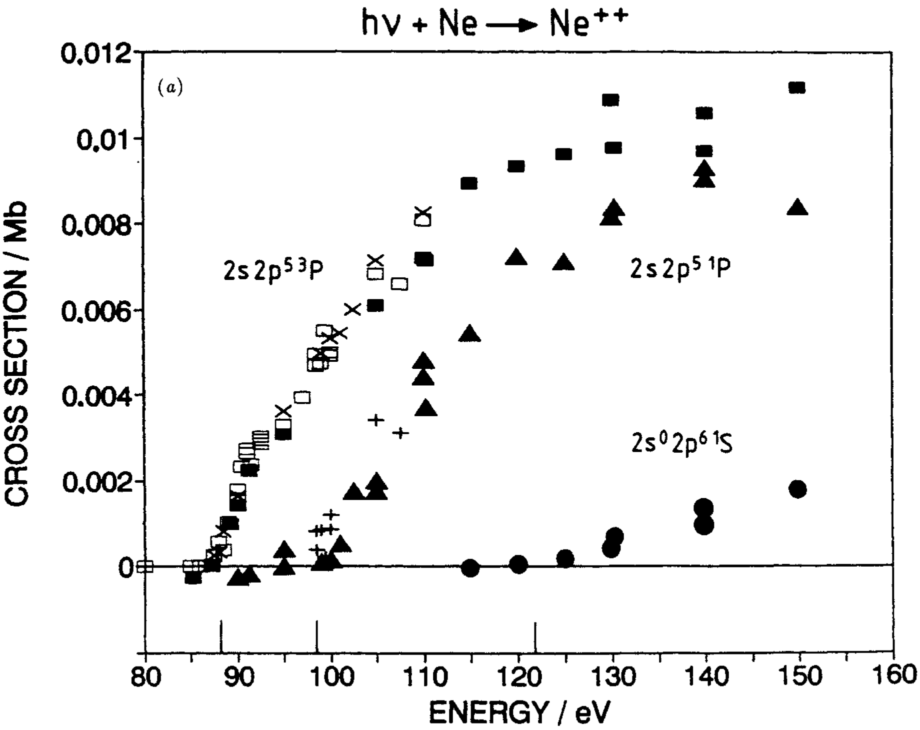
(1979). A possible alignment causing an anisotropy of the fluorescence radiation was not considered in the data evaluation. Its influence is—at least for the higher energies—assumed to be smaller than the error bars. This assumption is supported by an earlier observation of a statistical population of the J states of the Ar^{2+} $3s3p^5\,{}^3P^o$ state at 100 eV photon energy (Magel *et al* 1990). The cited experiment was carried out under the same conditions as the present experiment. Nevertheless, the population of the Ar^{2+} ${}^3P^o_{0,1,2}$ fine structure states close to threshold will be studied in the future.

First of all it is evident that there is no suppression of the ${}^3P^o$ channel with respect to the ${}^1P^o$ channel. This result is in agreement with the relative threshold signals measured by photoelectron coincidence spectroscopy (Hall *et al* 1992a). It is in contradiction to the propensity rules for direct DPI (Huetz *et al* 1991). Within this context a further result from Hall *et al* (1992a, b) is of interest: at threshold also the ${}^1D^e$ state of Ne^{2+} $2s^22p^4$ is more strongly populated than the ${}^3P^e$ state, this again is in contrast to Ar^{2+}. Thus, the two rare gases behave quite differently. As a possible explanation indirect ionization processes via resonance-like excited satellites and thier autoionization into the ${}^3P^e$ state have been discussed by Hall *et al* (1992a, b) who showed that they will probably be of no influence on the population of the Ne^{2+} $2s^22p^4$ states.

The cross sections in figure 3 increase practically linearly from threshold on and seem to reach a maximum around 140 eV for the ${}^3P^o$ and ${}^1P^o$ states. The cross section maximum for the double 2s-electron ionization is expected around 160 eV because of the larger threshold energy. It is also evident that at the present status of the experiment no evaluation of a Wannier exponent of the energy dependence is possible. Nevertheless the plot with enlarged energy scale in figure 3(*b*) shows a change of the photon energy dependence for the ${}^3P^o$ state near 92 eV and thus below the ${}^1P^o$ threshold which is so far not understood. No resonances in the energy dependence of the ${}^3P^o$ channel were observed unambiguously in the present experiment. The cross section for DPI of both 2s electrons is by one order of magnitude smaller than the sum of the ${}^3P^o$ and ${}^1P^o$ cross sections. A factor of $\frac{1}{12}$ would follow from the possibilities to select the two s electrons with respect to selecting one s electron and one p electron, neglecting the differences in binding energy and wavefunction.

Only the MBPT calculations from Carter and Kelly (1977) are available for a numerical comparison. This comparison is possible for the sum of the ${}^1P^o$ and ${}^3P^o$ channels at 130.6 eV, the only tabulated energy in our range. The theoretical value is 0.033 Mb while the experiment yields 0.019±0.006 Mb, the deviation being outside the experimental uncertainty. For 278 eV a ratio of two for DPI into the ${}^3P^o$ and ${}^1P^o$ channels of the $2s2p^5$ configuration can be extracted from Carter and Kelly (1977) which we could not test so far, because an excitation energy of 278 eV was not available but also because of the large uncertainty of the relative detection efficiency at 37.9 nm.

Summarizing, cross sections for direct double photoionization of Ne into the $2s2p^5\,{}^3P^o$ and ${}^1P^o$ states and the $2s^02p^6\,{}^1S^e$ state were measured for photon energies from threshold to 150 eV for the first time. The motivation of the experiment was a study of the properties of electron correlations being responsible for the direct DPI, especially of a correct consideration of the intershell correlations since s and p electrons are participating. No preferential population of the ${}^1P^o$ component was observed as expected from propensity rules based on the Wannier–Peterkop–Rau theory of direct DPI. This result is in agreement with the behaviour of the direct DPI of Ne observed by threshold photoelectron coincidence spectroscopy. The only existing prediction of the cross section from MBPT overestimates the sum of ${}^3P^o$ and ${}^1P^o$ cross sections by 50%. With respect to the complexity of the state selective DPI this result is regarded as satisfactory. On the other hand the present data should



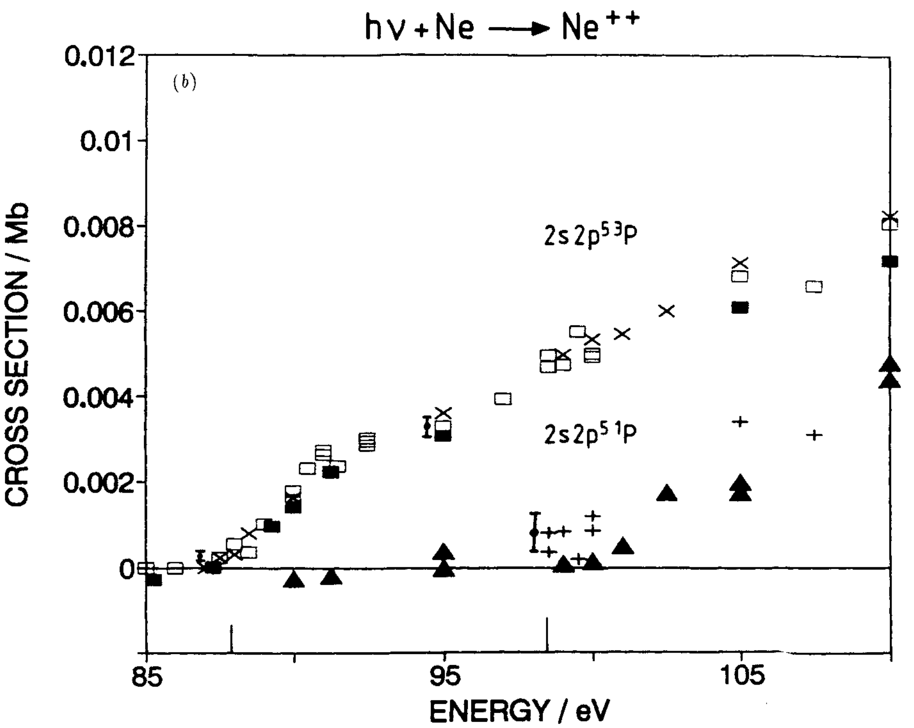


Figure 3. (*a*) Cross sections for DPI of Ne into the $2s2p^5\,{}^3P^o$ and ${}^1P^o$ and the $2s^02p^6\,{}^1S^e$ states as a function of the impinging photon energy with different symbols for independent experimental runs. Ionization thresholds are marked. (*b*) As (*a*), energy scale expanded, ɸ: statistical uncertainty at selected energies.

stimulate further detailed theoretical investigations of the simultaneous ionization of s and p electrons of the rare gas valence shell.

The technical support by the staff of BESSY is gratefully acknowledged. The work has been funded by the Bundesminister für Forschung und Technologie under contracts Nos 05 452 AXI and 05 447 AXI.

References

Amusia M Ya 1981 *Comment. At. Mol. Phys.* **10** 155–70
Carter S L and Kelly H P 1977 *Phys. Rev.* A **16** 1525–34
Eckhardt H and Schartner K H 1983 *Z. Phys.* A **312** 321–8
Flaig H J, Schartner K H and Kaase H 1983 *Nucl. Instrum. Methods* **208** 405–7
Hall R I, Dawber G, McConkey A G, MacDonald M A and King G C 1992a *Z. Phys.* D **23** 377–81
Hall R I, McConkey A G, Ellis K, Dawber G, MacDonald M A and King G C 1992b *J. Phys. B: At. Mol. Opt. Phys.* **25** 799–810
Huetz A, Salles P, Waymel D and Mazeau J 1991 *J. Phys. B: At. Mol. Opt. Phys.* **24** 1917–33
Jans W 1992 *Thesis* Technische Universität Berlin
Kossmann H, Schmidt V and Andersen T 1988 *Phys. Rev. Lett.* **60** 1266–9
Krässig B and Schmidt V 1992 *J. Phys. B: At. Mol. Opt. Phys.* **25** L327–33
Lablanquie P, Eland J H D, Nenner I, Morin P, Delwiche J and Hubin-Franskin M J 1987 *Phys. Rev. Lett.* **58** 992–5
Magel B, Möbus B, Schartner K H, Schmoranzer H, Wildberger M and Ehresmann A 1990 *BESSY Jahresbericht* 104–5
Mazeau J, Selles P, Waymel D and Huetz A 1991 *Phys. Rev. Lett.* **67** 820–3
Peterkop R 1971 *J. Phys. B: At. Mol. Phys.* **4** 513–21
Price S D and Eland J H D 1989 *J. Phys. B: At. Mol. Opt. Phys.* **22** L153–8
—— 1990 *J. Electron Spectrosc. Relat. Phenom.* **52** 649–60
Rau A R 1971 *Phys. Rev.* A **4** 207–20
Schartner K H 1990 *26th Int. Conf. on the Physics of Electronic and Atomic Collisions (New York, 1989, AIP Conf. Proc. 205)* ed A Dalgarno *et al* (New York: AIP) pp 215–23
Schartner K H, Lenz P, Möbus B, Magel B, Schmoranzer H and Wildberger M 1990 *Phys. Scr.* **41** 853–6
Schmidt V 1992 *Rep. Prog. Phys.* **55** 1483–659
Wannier G H 1953 *Phys. Rev.* **90** 817–25
Wuilleumier F and Krause M O 1979 *J. Electron Spectrosc. Relat. Phenom.* **15** 15–20