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LETTER TO THE EDITOR

Electron impact excitation out of the metastable levels of argon into the $3p^54p$ $J = 3$ level

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Abstract. We have measured the direct cross section for electron impact excitation out of the metastable $3p^54s[\frac{3}{2}]^o_2$ level ($1s_5$ in Paschen's notation) into the $3p^54p[\frac{5}{2}]_3$ level ($2p_9$) of argon from threshold to 800 eV. The direct cross section is 40×10^{-16} cm^2 at 10 eV.

Electron collisions with atomic argon play important roles in discharges, lighting, lasers and plasma processing. Due to their long lifetimes, extremely large cross sections, and large internal energy, metastable argon atoms are especially important in the understanding and modelling of argon plasmas (Bogaerts and Gijbels 1995). While electron excitation out of the ground state of argon has been extensively studied (Ballou *et al* 1973), only limited work has been performed on electron excitation out of the metastable levels of argon (excitation: Mityureva *et al* 1989, ionization: Dixon *et al* 1973). In our laboratory we have developed two apparatuses to study electron–metastable atom collisions using the optical method. This work on metastable Ar represents an extension of previous studies of metastable helium (Rall *et al* 1989, Lockwood *et al* 1992a, Lagus *et al* 1996, Piech *et al* 1996).

We have measured the direct cross section for electron-impact excitation out of the $3p^54s$ $J = 2$ metastable level ($1s_5$ in Paschen's notation) into the $3p^54p$ $J = 3$ level ($2p_9$). The argon atom in general does not conform to LS-coupling; the levels of the $3p^54p$ configuration are superpositions of different LS-terms of the same J. Within the $3p^54p$ configuration there is, however, only one $J = 3$ level, hence this level is accurately described by the LS-coupling level $^3D_3$. The two metastable levels of the $3p^54s$ configuration are also described by LS-coupling: the $1s_3$ is purely $^3P_0$ and the $1s_5$ is purely $^3P_2$.

Two separate apparatuses have been used in this work. A hollow-cathode discharge source was used to measure excitation functions from the threshold energy (~ 1.5 eV) up to the onset energy for excitation out of the ground state. For a detailed description of the apparatus the reader is referred to Lockwood *et al* (1992b); we only describe the general principles here. Metastable atoms are produced in an argon hollow-cathode discharge source and effuse out through a 1 mm diameter hole. Atoms exit the discharge source as a thermal beam ($v \sim 3 \times 10^4$ cm s^{-1}) and are crossed with a mono-energetic electron beam. We use a system of lenses, a narrow bandwidth interference filter (10 Å FWHM), and a photomultiplier tube (PMT) in photon counting mode to detect the light emitted by the argon atoms that were excited by electron collisions. This apparatus is limited to energies below

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the onset of ground-state excitation since the ground-state atom density is much higher than the metastable density ($\sim 10^6$:1). At energies above $\sim$ 13 eV the signal from ground-state excitation overwhelms any signal from metastable excitation. However, due to the relatively high target density of metastable atoms produced, $\sim 10^8$ cm^{-3}, we have obtained relative values of the cross sections for excitation into many levels of the $3p^5 4p$ configuration up to the onset energy for ground-state excitation.

A second, fast atomic-beam apparatus was used to obtain the absolute value of the direct cross section for the $1s_5 \rightarrow 2p_9$ excitation from just above the onset energy to 800 eV. A full description of the apparatus is found in Lagus *et al* (1996) and Boffard *et al* (1996), we only present the differences made for use with argon. A 2.1 keV Ar^+ beam is passed through a caesium vapour target yielding metastable atoms via the near-resonant charge exchange reaction,

$$Ar^+ + Cs \rightarrow Ar^*(1s_2, 1s_3, 1s_4, 1s_5) + Cs^+. \tag{1}$$

The $1s_5$ ($J = 2$) and $1s_3$ ($J = 0$) levels are metastable, whereas the $1s_2$ and $1s_4$ are $J = 1$ levels that are optically connected to the ground state. We assume the $3p^5 4s$ levels are populated according to their statistical weighting. Hence, after the $1s_2$ and $1s_4$ decay to the ground state, the ratio of ground state:$1s_5$:$1s_3$ atoms in the beam should be 6:5:1. Neynaber and Magnuson (1976) found that a 2 keV Ar^+ beam passing through a Rb vapour target (which is also near resonant) yields a 58% ground-state beam. This is consistent with a small amount of charge transfer into other manifolds (e.g. the $3p^5 4p$) which causes a slight deviation from the simple 6:5:1 ratio. Coggiola *et al* (1979) measured a 4.9:1 weighting for the two metastable levels of neon for 1.3 keV Ne^+ incident on Na. Hence, we assume our beam is 42% metastable atoms with a 5:1 weighting of $1s_5$ to $1s_3$.

As with the hollow cathode source, the resulting beam is crossed by a mono-energetic electron beam, and the resulting fluorescence is collected by a system of lenses, a narrowband interference filter and PMT. The neutral atoms are detected using a secondary-electron detector that is calibrated with a pyroelectric thermal detector. The metastable target density achieved is $\sim 10^5$ cm^{-3}, so signal rates are significantly lower than with the hollow cathode source. We obtain absolute results by measuring both the metastable flux and the profiles of the metastable beam, electron beam and optical viewing region. The optical detection efficiency is removed by ratioing the metastable signal to the signal from a separate experiment on ground-state argon, and then using the known ground-state cross section (Ballou *et al* 1973). Details of this procedure are given by Lagus *et al* (1996).

For both experiments, there is a mixture of $1s_3(^3P_0)$ and $1s_5(^3P_2)$ in the target beam, and thus the excitation functions may have contributions from both levels. As with the fast beam, we assume that the $1s_5$:$1s_3$ ratio for the hollow-cathode source is 5:1. In this letter we report our results for excitation into the $2p_9$ ($J = 3$) level obtained by measuring the intensity of the $2p_9 \rightarrow 1s_5$ emission at 8115 Å. The $2p_9 \rightarrow 1s_5$ is the only optically allowed emission from the $2p_9$ level, thus the $2p_9 \rightarrow 1s_5$ emission cross section is equal to the apparent cross section for the $2p_9$ level. Since excitation out of the $1s_5$ level ($J = 2$) into the $2p_9$ level corresponds to a $\Delta J = +1$ dipole allowed excitation, we expect the cross section to be large. The cross section for excitation out of the $1s_3$ level into the $2p_9$ level ($\Delta J = +3$) should be much smaller, because the direct coupling potential between the $1s_3$ and $2p_9$ levels due to the colliding electron can be shown to be zero by extending the arguments of Sharpton *et al* (1970) and Ballou *et al* (1973), e.g. equation (7) of Sharpton *et al* (1970) and equation (5) of Ballou *et al* (1973) .

In general, the observed fluorescence arises from four sources, direct excitation from the three initial levels ($1s_5$, $1s_3$ and ground), and cascades from higher levels. Combining the lower number density of $1s_3$ atoms, along with the argument in the preceding paragraph on the relative size of the $1s_3$ cross section, we conclude that the $1s_3$ contribution to the observed signal is negligible. The hollow-cathode experiment is used only below 13 eV so that the ground-state excitation does not occur. The target in the fast-beam experiment has slightly more ground-state atoms than metastable atoms, but since the cross section for excitation out of the metastable levels is two to three orders of magnitude larger than the cross section for excitation out of the ground level we can neglect the ground-state contribution to the total signal. Thus for both experiments the observed $2p_9$ signal is attributed almost exclusively to excitation out of the $1s_5$ metastable level.

In order to measure the cascade contribution to the $2p_9$ cross section, one would have to measure cross sections for excitation into manifolds higher than the $3p^54p$. This is difficult since many of the transitions out of these higher-lying levels lie in the infrared. While we have not *directly* measured the cascade contribution to the signal, we experimentally show that cascades contribute less than 20% to the apparent cross section. We expect the cascade contribution to be small since the argon $3p^54s \rightarrow 3p^54p$ excitation is similar to the $1s2s \rightarrow 1s2p(2^3S \rightarrow 2^3P)$ excitation in helium. For the $He(2^3S \rightarrow 2^3P)$ excitation, we observed a peak cross section that was 15 times as large as the next largest peak cross section out of the 2^3S level (Piech *et al* 1996). We expect a similar trend to hold for $Ar(1s_5 \rightarrow 2p_x)$ excitation, i.e. that the cross sections for excitation out of the $1s_5$ into the levels of the 2p manifold should be much larger than the cross sections for excitation out of the $1s_5$ into the levels of any other manifold. In the $He(2^3S \rightarrow 2^3P)$ excitation, cascade only contributed $\sim$ 10% to the apparent cross section.

In order to show experimentally that the cascade contribution to the measured cross section is small, we have performed a type of time-resolved electron excitation experiment using the fast-beam apparatus. The velocity of the 2.1 keV Ar^* atoms is approximately 0.01 cm ns^{-1}. In one atomic lifetime ($\sim$ 30 ns for the $2p_9$) the excited atoms in the beam move approximately 0.3 cm, a size comparable to our viewing region. The lifetimes of the nine levels in the $3p^55s$ and $3p^53d$ configurations that can decay into the $2p_9$ are 45 ns or longer. Compared to the $2p_9$ atoms created by direct excitation, those $2p_9$ atoms formed via cascade must undergo two decays to be detected (e.g. $2s_5 \rightarrow 2p_9$ and then $2p_9 \rightarrow 1s_5$), and thus stand a greater chance of exiting the viewing region. Hence, the cascade contributions to the cross sections measured in this fast-beam experiment are significantly smaller than they would be in a thermal-beam or static-gas target experiment. If we translate the electron gun (mounted on a translation stage) out of the viewing region towards the ion source, the longer-lived cascading levels will contribute more to the total signal. In figure 1 we present our data for signal versus electron-gun position (relative to the centre of the viewing region) along with the calculated response assuming no cascades and 20% cascades. The data demonstrate that cascades make up less than 20% of the apparent cross section as measured by the fast-beam experiment. For comparison we also show the curve for a static target.

The observed $2p_9$ excitation function is shown in figure 2. A corresponding table of values is located in table 1. The cross sections are taken to be the direct cross sections. The data below 12 eV are primarily from the hollow-cathode source, since it has significantly higher signal/noise in this regime. The higher-energy data are from the fast-beam source exclusively. Absolute calibration is performed using the fast-beam source, and the two experiments are normalized at 10 eV. We note that the shapes of the excitations from both experiments agree well in their region of overlap, and therefore conclude that the cascade contribution to the cross section as measured by the hollow-cathode experiment is also small (both experiments also agreed extremely well in the shapes of the excitation

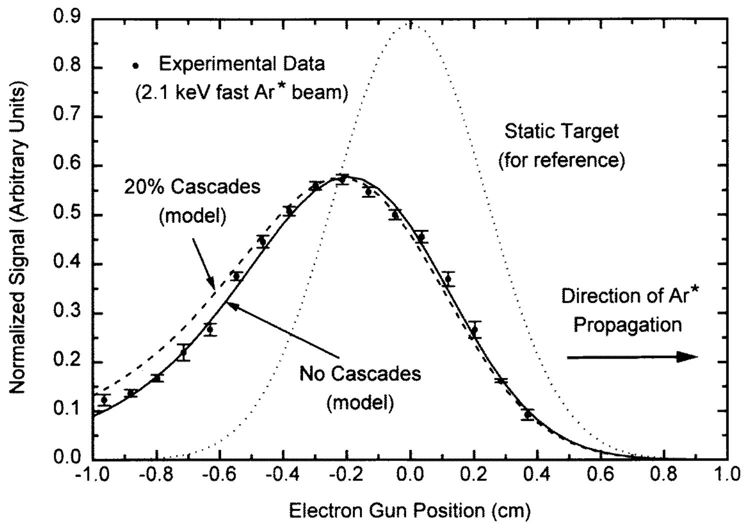

Figure 1. Cascade analysis of $1s_5 \rightarrow 2p_9$ excitation at 50 eV. Experimental data are obtained by translating the electron gun along the Ar* beam axis (relative to the fixed centre of the viewing region) and recording the photon counts. The model calculations (no cascades and 20% cascades) take into account the known electron-beam profile and the known optical profile of the apparatus. As a worst-case scenario, we have used the $2s_5$ level, which has both the shortest lifetime ($\sim$ 45 ns) and largest branching ratio of the levels that can decay into the $2p_9$ level. Calculations for a static target ($\cdots\cdots$) are shown to illustrate the effects of a fast target beam.

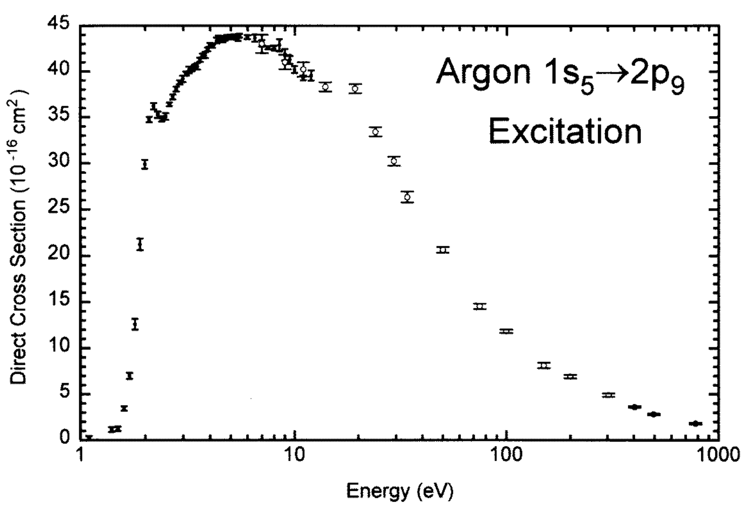

Figure 2. Direct cross section for electron excitation out of the $1s_5$ level into the $2p_9$ level. Results from the hollow cathode experiment (●) are normalized to the results of the charge exchange experiment (○). The error bars represent statistical error only and do not include the uncertainty of the absolute calibration.

functions observed for metastable He, see Piech *et al* 1996). The excitation function overall is relatively broad, which is expected for an excitation into an optically allowed level. The small peak at $\sim$ 2 eV may be due to resonances. No attempt has been made to correct the data for the effects of polarization of the emitted light. We have not measured the

Table 1. $1s_5 \rightarrow 2p_9$ direct excitation cross section in units of 10^{-16} cm^2.

E (eV)	Q^{dir}	E (eV)	Q^{dir}
2	30[a]	30	30
3	39	50	21
4	43	100	12
5	44	200	6.9
6	44	300	5.0
10	40	400	3.6
20	37	500	2.8

[a] All values have a ±35% uncertainty from the absolute calibration.

polarization fraction $(I_{\parallel} - I_{\perp})/(I_{\parallel} + I_{\perp})$, but even if it were as large as ±0.3 this would only change our cross section results by 10%.

Mityureva *et al* (1989) found a peak cross section for excitation into the $2p_9$ level of $(180 \pm 50) \times 10^{-16}$ cm^2, a value four times larger than ours. They also find that the cross section decreases by 25% from the peak value at 4 eV to their highest energy at 12 eV; we observe a much more modest 8% decrease. Similar discrepancies in magnitude and shape exist between our measurements and theirs on metastable helium (Lagus *et al* 1996).

We are unaware of any theoretical calculations for electron excitation out of the metastable levels of argon into the $2p_9$ level. Hyman (1978) reported average values of the Born cross sections for excitation out of the $3p^5 4s$ manifold into the $3p^5 4p$ manifold, which do not directly correspond to our measurements. We have used the Born–Bethe approximation along with the tabulated transition probabilities of Wiese *et al* (1989) to obtain a first-order estimate of the theoretical cross section as shown in figure 3. At high energies our data lie slightly above the theoretical curve, although the difference is within the uncertainties of our experiment and the transition probability used in the Born–Bethe approximation.

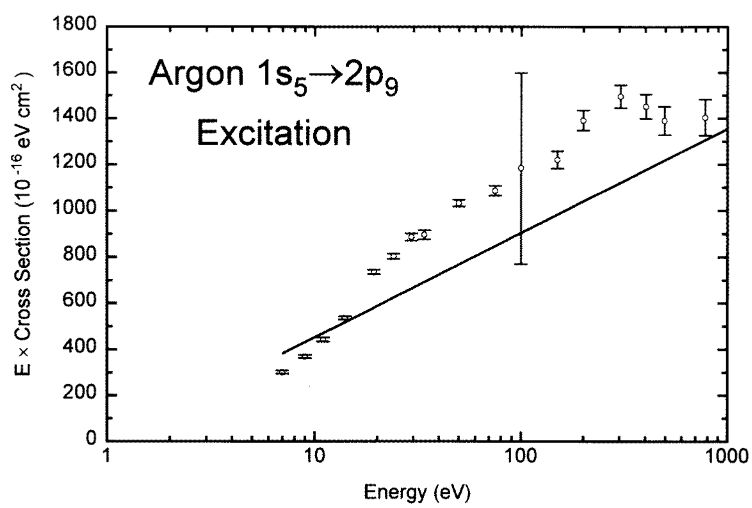

Figure 3. Bethe plot of experimental results (○) and a calculation using the Born–Bethe approximation (——). The error bars shown represent statistical uncertainty only, except for the point at 100 eV which includes the absolute uncertainty of ±35%.

We expect to extend our measurements to the other levels in the $3p^5 4p$ manifold. Analysis of the results for these other levels is complicated by the possibility of significant contributions from both of the initial metastable levels. Measurements of the cross sections into manifolds above the $3p^5 4p$ are particularly important, since one can then directly determine the cascade into the $3p^5 4p$ manifold. More sophisticated theoretical calculations of the cross sections are also desirable.

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