RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 38, NUMBER 12 DECEMBER 15, 1988

Rapid Communications

The Rapid Communications section is intended for the accelerated publication of important new results. Since manuscripts submitted to this section are given priority treatment both in the editorial office and in production, authors should explain in their submittal letter why the work justifies this special handling. A Rapid Communication should be no longer than 3½ printed pages and must be accompanied by an abstract. Page proofs are sent to authors, but, because of the accelerated schedule, publication is not delayed for receipt of corrections unless requested by the author or noted by the editor.

Fluorescence studies of multiphoton ionization of Sr: Production of excited ionic states

H. K. Haugen* and A. S. Othonos

Department of Physics, University of Toronto, Toronto, Ontario, Canada

(Received 24 August 1988)

Multiphoton ionization of strontium in the wavelength range of ~558–590 nm is studied via the fluorescence from excited $Sr^+(5p\,^2P_{3/2,1/2})$ ions. The fine-structure sublevels in the singly charged ion are fully resolved and are found to be nonstatistically populated. Four- and five- (or more) photon processes are observed to populate $Sr^+(5p\,^2P)$ in the intensity regime of $I \sim 1$–50 GW/cm^2, but no emission is detected from higher-lying states of Sr^+.

Multiple photon ionization (MPI) of strontium has been studied previously by several researchers.[1–10] Interest in alkaline-earth MPI processes was stimulated by early work of Aleksakhin and co-workers[1,2] which revealed that such species could be doubly ionized at surprisingly low-field strengths. Subsequently, Welge and co-workers[3–6] performed electron spectroscopy for strontium three- and four-photon ionization revealing a rich structure of autoionizing states above the first ionization limit. The energy analysis of ejected electrons revealed decay channels to $Sr^+(5s\,^2S)$ or $Sr^+(4d\,^2D)$ after absorption of three or four photons. In the latter case, one speaks of extra photon absorption in the continuum. It was further suggested that the $5p\,^2P$ state of the ion should play an important role in the double ionization of neutral strontium, but the electrons were not sufficiently energetic to be measured. Agostini and Petite[7–9] improved the electron-energy analysis in later experiments. By utilizing an extraction field, they obtained a low-energy electron peak which was tentatively identified as resulting from Sr^{**} autoionizing decay to $Sr^+(5p\,^2P)$. However, the fine-structure sublevel contributions, $J = \frac{1}{2}$ and $\frac{3}{2}$, could not be resolved in the measurements.

One of the outstanding questions in the multiphoton ionization of alkaline-earth atoms has been whether two-electron ionization is a direct or a two-step process. A simplified energy-level diagram of Sr is shown in Fig. 1, together with an indication of the two different pathways to the Sr^{2+} ionization limit. In the first (direct) case, the laser field pumps the atom above the first ionization limit and further through the spectrum of doubly excited states to the doubly ionized continuum. In the second (two-step) case, the atom is initially pumped above the first ionization limit, then decays to a specific state of the singly charged ion, which is subsequently ionized in the strong field. This paper presents preliminary results of fluorescence studies of multiphoton ionization of strontium in the laser wavelength range of $\lambda \sim 558$–590 nm and with laser focal intensities $\sim 1-50$ GW/cm^2. We monitor the fluorescence from $Sr^+(5p\,^2P_{3/2,1/2})$ ions which are formed via four- (or more) photon absorption by the neutral atom. The present approach demonstrates a new and complementary technique in MPI measurements with three major advantages: (1) It allows fine-structure sub-

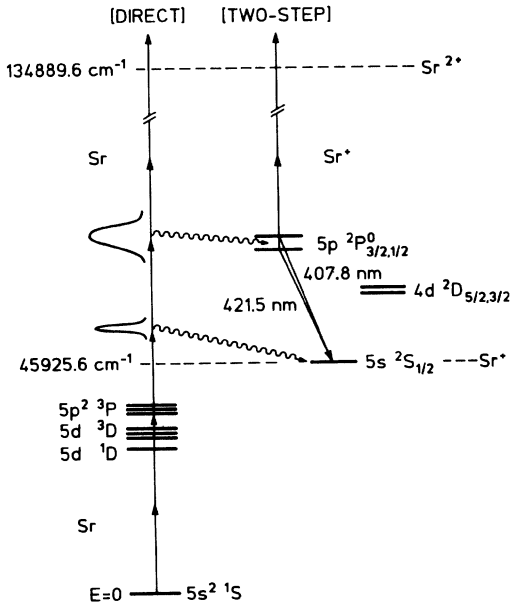

FIG. 1. Simplified energy-level diagram of Sr and Sr^+, illustrating the two possible mechanisms for double ionization of the neutral atom. In the present experiment, fluorescence from $Sr^+(5p\,^2P_{3/2,1/2})$ is monitored at 408 and 421 nm.

level resolution to be readily obtained in the final ionic states, (2) it facilitates the search for weak channels which can populate in general a multiplicity of ionic states, and (3) a wide variety of competing optical channels can be studied simultaneously.

Details of our experimental technique will be given in a separate publication.[11] A Nd:yttrium-aluminum-garnet laser pumps a pulsed dye laser. The ~10-ns duration dye laser pulses of ~3-cm^{-1} bandwidth are focused into a crossed heatpipe oven which is operated in a nonheatpipe regime. Here the strontium undergoes continuous evaporation and condensation with just a sufficient amount of He or Ar buffer gas to prevent coating of the windows. The fluorescence emission from the interaction region, and perpendicular to the direction of laser-light propagation, is detected in ~20-ns gate and then boxcar averaged. The wavelength selection is achieved using a monochromator or narrow-band interference filters. It should be pointed out that a vapor is used to provide sufficient target density to facilitate fluorescence measurements. In the early days of MPI measurements, several studies were undertaken in cells where the total ion or electron current was measured via collector plates. Later, low-density atomic beams were increasingly employed in measurements of electron energy and angular distributions, without interfering with collisional processes taking place simultaneously. In the present case, we revert to cell measurements to enhance the light-collection capabilities. However, as described in Ref. 11, a number of systematic checks have been performed in these ionic-fluorescence measurements. Collisional effects are shown to be negligible at moderate strontium densities in the range ~10^{-5}-10^{-1} Torr, while in a true heatpipe regime and pressures > 1 Torr, collisional excitation effects are observed in the vapor. Our typical strontium operating pressure is ~10^{-3} Torr. Collision effects are further shown to be negligible for all Ar-He buffer densities that are utilized.

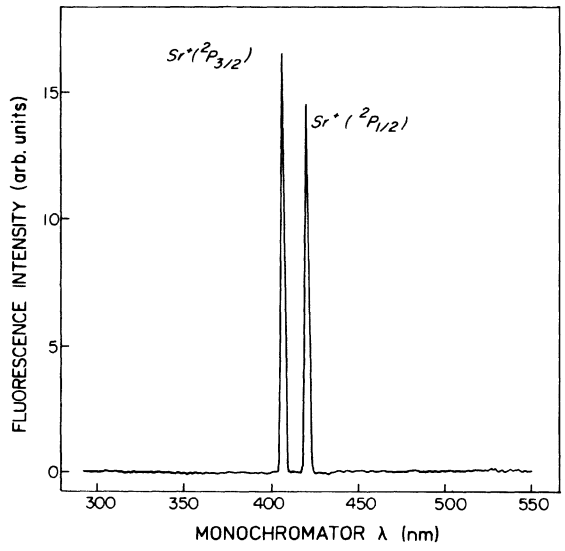

FIG. 2. Typical fluorescence spectrum for irradiation at $\lambda = 562.85$ nm and laser intensity $I \sim 50$ GW/cm^2.

Figure 2 shows a typical monochromator scan of the emission from the focal volume of the laser beam. The $Sr^+(5p\,^2P)$ fine-structure sublevels are readily resolved using either a monochromator or narrow-band interference filters. The spectrum is very clean in the region of 200–600 nm. The $J = \frac{1}{2}$ vs $J = \frac{3}{2}$ signals are typically nonstatistical. By isolating one of the fine-structure sublevels with the monochromator or appropriate narrow-band interference filter and scanning the dye laser wavelength, resonant and autoionizing structure is observed in the four-photon absorption. Spectra taken for $J = \frac{1}{2}$ and $\frac{3}{2}$ fluorescence from $Sr^+(5p\,^2P)$ with a laser intensity of $I \sim 5$ GW/cm^2 are shown in Fig. 3. The spectral features have been observed earlier by Welge and co-workers[3–6] and by Agostini and Petite,[7–9] but not with J sublevel resolution. Lambropoulos *et al.*,[12] by combining the totality of information from electron spectroscopy with theoretical calculations, have recently arrived at the following assignments. The resonances at 559.4 and 559.7 nm are, respectively, due to $4d(D_{3/2})4f[5/2]_3$ and $4d(D_{5/2})4f[5/2]_3$ states. The feature at 560.6 nm is associated with the $5p\,^2\,^3P_2$ two-photon resonance in the neutral atom. The peaks at 561.8 and 563 nm are, respectively, due to $5p\,5d(^3D_3)$ and $4d(D_{5/2})4f[7/2]_3$. Finally, the feature at 563.6 nm arises from the $5p\,5d(^3D_1)$ state.

A study of the behavior of the resonances is performed

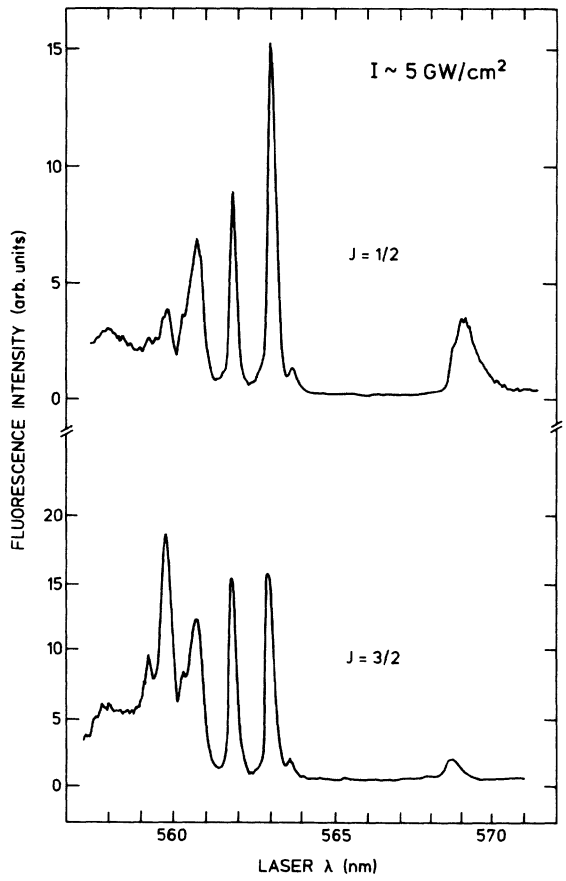

FIG. 3. Fluorescence spectra vs laser wavelength for each J sublevel of $Sr^+(5p\,^2P)$ at an intensity $I \sim 5$ GW/cm^2. The vertical scale ($J = \frac{1}{2}$ vs $J = \frac{3}{2}$) is internally normalized.

over the intensity range $I \sim 1$–$50\,\mathrm{GW/cm^2}$. At the highest field intensities, the structure is broadened considerably. The relative contribution of the various peaks to the fine-structure $J = \frac{1}{2}$ and $J = \frac{3}{2}$ sublevels is also found to depend sensitively on laser intensity. For $I \sim 1.5\ \mathrm{GW/cm^2}$, the peaks are sharp, and broad autoionization-state contributions are virtually absent. Other emissions from the medium are not observed for strontium densities less than a few Torr. The only exception arises for exact tuning to the $5s^2\,{}^1S \rightarrow 5p^2\,{}^3P_{2,0}$ two-photon resonances, where fluorescence emission is observed in the neutral-atom triplet spectrum. Further details concerning the $J = \frac{1}{2}, \frac{3}{2}$ populations versus laser intensity as well as relative ionic versus neutral fluorescence contributions are given in the upcoming publication.

Agostini and Petite,[7–9] by utilizing an extraction field, obtained a peak representing the convolution of population in $J = \frac{1}{2}$ and $J = \frac{3}{2}$ of $5p\,{}^2P$. A scan of the electron yield versus wavelength suggested discontinuities at the expected four-photon thresholds for the $J = \frac{1}{2}$ and $\frac{3}{2}$ sublevels at 567.8 and 574.4 nm, respectively. In the present fluorescence measurements, and at laser intensities $I \sim 50\ \mathrm{GW/cm^2}$, a sharp decrease in the signal is observed at the four-photon thresholds. We observe the effect *individually* for the two sublevels, not for the sum of populations, as in Ref. 7. Small but finite fluorescence signals at 408 and 421 nm are also detected beyond the four-photon thresholds. These signals are linear in Sr density and exhibit a much greater sensitivity to laser intensity. We associate these spectra for $\lambda \sim 580$–590 nm with five-photon absorption leading to $Sr^+(5p\,{}^2P)$. At the highest field strengths achieved, the five-photon signal at 580.4 nm is still ~ 30 times weaker than the four-photon signal at 562.85 nm.

One of the important questions to be elucitated by the present work is whether the double-ionization process is a direct or a two-step mechanism. At present, the question can be answered only qualitatively. The possible emission from higher-lying states than $5p\,{}^2P$ is a qualitative monitor of population resulting from decay of autoionization states well above the first ionization limit. We search for such emissions by increasing the Sr density and checking that any observed fluorescence emission scales linearly with the strontium density. In particular, a careful search is made for emissions from the $6s\,{}^2P$-$5p\,{}^2P^0$ ($\lambda = 416.3$, 430.7 nm) and $5d\,{}^2D$-$5p\,{}^2P^0$ ($\lambda = 338.2$, 346.5 nm) from the next two higher-lying levels. No signals are observed at these wavelengths within our detection sensitivity. The sensitivity is defined via an internal normalization to be greater than $1{:}10^3$ with respect to the ionic resonance fluorescence at 408 and 421 nm.

Relating the fluorescence observations to limits on populations of various upper states involves a number of considerations. First, the individual oscillator strengths must be taken into account. Second, the ionization of singly charged ions needs to be considered, particularly for higher-lying states of Sr^+. Finally, a variety of autoionization states, and field-induced effects on the states, must ultimately be identified and characterized to fully interpret the results. More data will also be helpful. So far we have explored the interactions through a rather limited "window" defined by relatively strongly allowed dipole transitions in the wavelength range of 200–600 nm. Further work could extend the approach to incorporate laser-induced fluorescence measurements, thereby extracting relative populations of the, e.g., low-lying $Sr^+(^2S)$ and $Sr^+(^2D)$ states. Also, only the incoherent fluorescence is presently collected from the interaction region at 90° to the beam direction. Measurements in the ultraviolet could also serve to probe the fate of the autoionization states and explore coherent emissions in the forward direction due to harmonic generation.

The qualitative conclusions of the present work support the findings by electron-spectroscopy techniques. Prior to undertaking second-generation experiments, we are extending the empirical work to other species such as Ba, where a wealth of spectroscopic studies has been performed to date. Further exploration of the limitations of the "all optical" approach will also be necessary. The possible influence of radiation trapping, polarization of emission, and the potential detrimental role of fine-structure mixing at intermediate densities ($\sim 10^{-1}$ Torr) will need to be studied in detail. However, the preliminary results are very encouraging. They demonstrate a density regime where fluorescence measurements can be performed without significant collisional effects in the medium. The approach, even in its more simple form, allows the exploration of weak population channels and the resolution of fine-structure sublevel populations in the final ionic states. A better understanding of the behavior of pseudo-two-electron systems in strong laser fields will ultimately require the combined insight gained through many complementary experiments.

---

*Present address: Institute of Physics, University of Aarhus, DK-8000 Aarhus C, Denmark.

[1] I. S. Aleksakhin, I. P. Zapesochnyi, and V. V. Suran, Pis'ma Zh. Eksp. Teor. Fiz. **26**, 14 (1977) [JETP Lett. **26**, 11 (1977)].

[2] I. S. Aleksakhim, N. B. Delone, I. P. Zapesochnyi, and V. V. Suran, Zh. Eksp. Teor. Fiz. **76**, 887 (1979) [Sov. Phys. JETP **49**, 447 (1979)].

[3] S. L. Chin, D. Feldmann, J. Krautwald, and K. H. Welge, J. Phys. B **14**, 2353 (1981).

[4] D. Feldmann, H.J. Krautwald, and K. H. Welge, J. Phys. B **15**, L529 (1982).

[5] D. Feldmann and K. H. Welge, J. Phys. B **15**, 1651 (1982).

[6] D. Feldmann, H. J. Krautwald, S. L. Chin, A. von Hellfeld, and K. H. Welge, J. Phys. B **15**, 1663 (1982).

[7] P. Agostini and G. Petite, Phys. Rev. A **32**, 3800 (1985).

[8] P. Agostini and G. Petite, J. Phys. B **18**, L281 (1985).

[9] G. Petite and P. Agostini, J. Phys. (Paris) **47**, 795 (1986).

[10] I. I. Bondar, M. I. Dudich, and V. V. Suran, Zh. Eksp. Teor. Fiz. **90**, 1952 (1986) [Sov. Phys. JETP **63**, 1142 (1985)].

[11] H. K. Haugen and A. S. Othonos (unpublished).

[12] P. Lambropoulos, X. Tang, P. Agostini, G. Petite, and A. L'Huillier, Phys. Rev. A **38**, 6165 (1988).