Soft x-ray emission from laser-produced strontium ions

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Abstract

Soft x-ray spectra, in the range from 2 nm to 9 nm, were recorded from strontium plasmas formed by pulses from 20 ps, 170 ps and 5.5 ns Nd:YAG lasers operating at the fundamental wavelength of 1064 nm. Features due to 3d–4p and 3d–4f transitions were identified by comparison with spectra from adjacent ions and atomic structure calculations with both the Cowan code and the Flexible Atomic Code. As in the spectra of ions of other elements in the fifth row of the periodic table, resonant lines 3d−3d−4p1, 3d−3d−4f1 and satellite lines 3d−14s1−3d−24s14p1, 3d−14s1−3d−24s14p1 of \( \Delta n = 1 \) were observed over the 3.0–8.5 nm region, emitted by 10+ to 19+ ions. These \( \Delta n = 1 \) transitions provide a range of narrow band emission features which may match to specific multi layer combinations for reflective optics in the extreme ultraviolet region of the spectrum.

Keywords: laser produced plasma, strontium, soft x-ray spectroscopy, unresolved transition array, Cowan code, flexible atomic code

(Some figures may appear in colour only in the online journal)

1. Introduction

Spectra from laser produced plasmas generally display emission from a range of ion stages, particularly where they are integrated over both the lifetime and the spatial extent of the plasma. This range is determined, in the main, by the power density of the laser pulse at the target surface and the range of ionisation potentials of the target materials [1]. Emission spectra of laser produced plasmas, in addition to other sources, have been recorded for most elements in the periodic table, resonant transitions and 3d–4p satellite transitions in some strontium ions have already been reported collectively in a table of atomic data [6]. 6 lines from \( 3d^{10}4s^{1}−3d^{9}4s^{1}4p^{1} \) transitions in \( \text{Sr}^{9+} \) and 3 lines from the \( 3d^{10}−3d^{9}4p^{1} \) transitions in \( \text{Sr}^{10+} \) were reported in [12]. 20 lines from \( 3d^{9}−3d^{8}4p^{1} \) transitions in \( \text{Sr}^{11+} \) were reported by Ryabtsev and Reader [13], while Podobedova [14] reported 54 lines from the \( 3d^{2}−3d^{2}4p^{1} \) transition array in \( \text{Sr}^{12+} \) using experiments with a low inductance vacuum spark. In this work, a series of UTAs are reported between 2.5 and 9.0 nm. These are identified as the resonant lines 3d−14s1−3d−24s14p1, 3d−14s1−3d−24s14p1 and associated satellite lines 3d−14s1−3d−24s14p1, 3d−14s1−3d−24s14p1 in ions leading to discrete transition arrays being observed in the spectra [8]. This is in contrast to the overlapping frequently observed in \( \Delta n = 0 \) transitions, for example the single \( n = 4 \) to \( n = 4 \) unresolved transition array (UTA) [9] in spectra of laser produced plasmas formed on tin targets [10], which is generated by emission from a range of ion stages and has found application as a source for EUV lithography.

Strontium has been shown to be a candidate for high brightness x-ray laser applications. Soft x-ray amplification at 16.41 and 16.65 nm in a plasma generated by 0.53 micron laser irradiation in Ne-like Strontium ion have been reported [11]. 3d–4p resonant transitions and 3d4s–3d4s4p satellite transitions in some strontium ions have already been reported collectively in a table of atomic data [6]. 6 lines from \( 3d^{10}4s^{1}−3d^{9}4s^{1}4p^{1} \) transitions in \( \text{Sr}^{9+} \) and 3 lines from the \( 3d^{10}−3d^{9}4p^{1} \) transitions in \( \text{Sr}^{10+} \) were reported in [12]. 20 lines from \( 3d^{9}−3d^{8}4p^{1} \) transitions in \( \text{Sr}^{11+} \) were reported by Ryabtsev and Reader [13], while Podobedova [14] reported 54 lines from the \( 3d^{2}−3d^{2}4p^{1} \) transition array in \( \text{Sr}^{12+} \) using experiments with a low inductance vacuum spark. In this work, a series of UTAs are reported between 2.5 and 9.0 nm. These are identified as the resonant lines 3d−14s1−3d−24s14p1, 3d−14s1−3d−24s14p1 and associated satellite lines 3d−14s1−3d−24s14p1, 3d−14s1−3d−24s14p1 in ions
ranging from Sr$^{10+}$ to Sr$^{19+}$. A recent study on a laser plasma formed on a zirconium target [15] highlights the role played by a spectator 4s electron in broadening the transition arrays due to 3d–4p and 3d–4f which are found in the spectrum between 2 and 7 nm. The similarities in the XUV spectra of elements between $Z = 37$ (Rb) and $Z = 42$ (Mo) suggest that Sr would be a suitable candidate as a target material for radiation in the water window, between 2.3 and 4.3 nm [16].

2. Experimental apparatus

Three different Nd:yttrium-aluminium-garnet (Nd:YAG) lasers operating at 1064 nm were used to form the plasmas on solid strontium targets at normal incidence. These were a Continuum Surelite III 5.5 ns (FWHM), 605 mJ, root mean square (rms) deviation 0.48%, an EKSPLA SL312P 170 ps (FWHM), 350 mJ, rms deviation 20.2% and an EKSPLA PL2250 (20 ps (FWHM), 35 mJ, rms deviation 0.5%). The three lasers were focused using a 50 mm focal length aspheric lens, and the spot size diameters were 20–38 μm, 25–43 μm and 18–32 μm, respectively. Power densities on target were 9.7 × 10$^{12}$–3.6 × 10$^{13}$ (W cm$^{-2}$) for the 5.5 ns laser, 1.2 × 10$^{14}$–5.4 × 10$^{14}$ (W cm$^{-2}$) for the 170 ps laser and 2.2 × 10$^{14}$–7.4 × 10$^{14}$ (W cm$^{-2}$) for the 20 ps laser. The variability in these power densities was due mainly to the unevenness of the target surface and also the fluctuations in the laser energy. Spectra were recorded in single-shot (5.5 ns and 170 ps) or five-shot mode (20 ps), with integration times of 0.011 s for the 5.5 ns pulses and 5 s for the 170 and 20 ps lasers, where the camera was triggered manually. The spectra were corrected for background signal on the CCD and recorded under a vacuum better than 10$^{-4}$ mBar. A 0.25 m Flat-field grazing incidence spectrometer, equipped with a SHIMADZU 30-002 1200 grooves mm$^{-1}$ grating and an ANDOR DX436-BN CCD camera was set at an angle of 45 degrees to the incident angle of the laser. The resolving power of the system $R = \lambda/\Delta \lambda \sim 300$. The recorded spectra were time-integrated by the spectrometer.

Wavelength calibration was achieved using known emission lines from carbon, copper, aluminium, oxygen, silicon and nitrogen, based on reference targets of Cu, Al$_2$O$_3$ and reaction-bonded silicon nitride (RBSN) Si$_3$N$_4$. The typical spectral resolution was better than 0.01 nm. The residuals on the calibration fit were all below 0.095 nm.

3. Identification and analysis of spectra

Figure 1 shows observed time-integrated and normalised emission spectra in the 2.5–9.0 nm region from laser produced Sr plasmas by single shots of the 5.5 ns and 170 ps lasers, and five-shot accumulations of the 20 ps laser. The spectra are normalised to the maximum signal and shown with arbitrary units of intensity, derived from the summed vertical pixel counts at a given wavelength. The spectrometer response is not absolutely calibrated in this work. 1s–2p, 3p, 4p, 5p transitions in C$^{5+}$ and 1s$^2$–1s2p and 1s3p transitions in C$^{4+}$ appear due to contamination by oil in which the Sr targets were stored. The carbon lines in the 2.5–4.5 nm region are seen in second order between 5.5 and 8.5 nm. These carbon lines were used for wavelength calibration from identifications in the NIST Atomic Spectra Database. The dip visible in the 4.0–4.5 nm region is due to carbon absorption by a thin layer on the grating. The spectral emission remains above zero at all wavelengths in this region due to plasma continuum emission and the presence of multiple weak transitions in the spectrum.

Figure 2 shows published transitions from the National Institute of Standards and Technology (NIST) atomic spectra database for comparison with an experimental spectrum in the 6.0–9.0 nm region from a 170 ps laser produced plasma. 3d–4p resonant transitions in Sr$^{10+}$ to Sr$^{12+}$ were observed in low-inductance vacuum spark experiments [12–14] and the 5p–3d resonant transition in Sr$^{20+}$ was reported from a calculation [17].
by the 170 ps laser, combined with 2.5 G to (3d^9-3d^{10}4p^1) (red), 3d^{10}4s^2-3d^{10}4s^24p^1 (blue), 3d^{10}4s^23d^{10}4p^1 (green) and 3d^{10}4s^23d^{10}4f^1 (light blue) in the 2.5–4.5 nm region.

We undertook calculations using the Flexible Atomic Code (FAC) [18] and the Cowan atomic structure code [19] to identify the main features in the spectra by comparison with weighted transition probabilities, (gA values). For the Cowan code calculations the Slater Condon Integrals were left unscaled. This scaling, when applied to the direct F^2(3d, 4p) and exchange G^1(3d, 4p) integrals, which largely determine the array width, was previously found to give optimum agreement for 3d–4p transitions in the Co-like isoelectronic sequence [2, 20, 21]. Here, g = 2J + 1, and A is the Einstein A coefficient. Figures 3 and 4 show comparison of the measured emission spectra in the 2.5–4.5 nm and 4.0–9.0 nm region, both from plasmas formed by the 170 ps laser, combined with gA values of Δn = 1 transitions of the 3d^{10}–3d^{10}4p^1 (red), 3d^{10}4s^2–3d^{10}4s^24p^1 (blue), 3d^{10}4s^23d^{10}4p^1 (green) and 3d^{10}4s^23d^{10}4f^1 (light blue) by FAC for each ion stage. The gA values were convolved with a Gaussian (σ = 0.003 nm), binned in order to match the pixel spacing of the CCD camera mounted on the spectrometer, and finally normalised to best reflect experimental spectra.

Table 1 lists the peak wavelengths of the observed emission features due to Δn = 1 transitions in Sr^{10+} to Sr^{19+}. The mean wavelengths and widths of the transition arrays, calculated using the procedure of Bauche et al [9] are also provided, based on gA values calculated using the Cowan code and FAC code. The Slater-Condon F^2, G^1 and R^6 parameters were unscaled in both the Cowan code and FAC calculations. Peaks a to d (figure 3) are a combination of 3d^{10}–3d^{10}4f^1 resonant transitions in Sr^{16+} to Sr^{19+} and 3d^{10}4s^2–3d^{10}4s^24f^1 satellite transitions in Sr^{15+} to Sr^{18+}. Peaks p to t (figure 3) are a combination of 3d^{10}–3d^{10}4p^1 resonant transitions in Sr^{16+} to Sr^{19+} and 3d^{10}4s^2–3d^{10}4s^24p^1 satellite transitions in Sr^{16+} to Sr^{19+}. Peaks e to o (figures 3 and 4) are a blend of resonant and satellite transitions in a range of ions.

Transition arrays due to each ion stage are clearly identifiable. In the case of peaks a to d in figure 3 and table 1, progressing from short to long wavelength, 3d–4f transitions from ions 19+ through 16+ dominate the spectrum. Peaks e to g are combinations of 3d–4f and 3d–4p transitions. By comparison with calculations it is clear that satellite transitions broaden the peaks from a to f. At wavelengths longer than 4 nm the resonant transitions and their associated satellite transitions are separated from each other as peaks g and h, respectively. In figure 3, again the resonant transitions and their associated satellite transitions form separate peaks. Here peaks g, i, j, l, n, o, p, r and t are created by resonant transitions, while peaks h, k, m, q and s are created by satellite transitions, some of which overlap each other. Calculated gA value of satellite transitions were smaller than those for resonant transitions in our calculations. The peaks in the measured spectra were mainly formed by resonant transitions, which were stronger than those due to satellite transitions. However, the intensities of peaks g and h are irregular, the strength of h is stronger than g. There is a possibility that peak g may be affected by carbon absorption due to contamination of the grating, thus the measured intensity was reduced. The effects of satellite transitions on the spectrum are thus clearly visible.

4. Discussion

Figure 5 shows the positions of calculated gA values of Δn = 1 transitions of the 3d^{10}–3d^{10}4p^1 (red), 3d^{10}4s^2–3d^{10}4s^24p^1 (blue), 3d^{10}4s^2–3d^{10}4s^24p^1 (green) and 3d^{10}4s^2–3d^{10}4s^24f^1 (light blue) in the spectra of Strontium ions calculated using the Cowan code. The gA values were convolved with a Gaussian, binned and normalised as in figures 2 and 3. The resonant transitions, 3d^{10}–3d^{10}4p^1 emit in the 2.9–5.7 nm region, and the 3d^{10}–3d^{10}4p^1 transitions appear between 3.6 and 8.5 nm from Sr^{10+} to Sr^{19+}. Satellite transitions of 3d^{10}4s^2–3d^{10}4s^24f^1 and 3d^{10}4s^2–3d^{10}4s^24p^1 are
Table 1. Identifications of observed emission peak wavelengths. \( \Delta n = 1 \) transitions for Strontium ions calculated by Cowan code and FAC. Mean wavelength and width of a transition array calculated with \([9]\). Note: no width is given for single-line transitions.

| Peak No. | Measured Wavelength (nm) | Cowan Wavelength (nm) | FAC Wavelength (nm) |
|----------|--------------------------|-----------------------|----------------------|
|          | UTA Peak Spectrum Transition | UTA Peak Width | UTA Peak Width |
| a        | 2.97 | Sr XX \( 3d^2-3d^4f^1 \) | 2.96 | 2.97 |
| b        | 3.12 | Sr XIX \( 3d^4s^2-3d^4s^4f^1 \) | 3.00 | 0.010 | 3.01 | 0.010 |
| c        | 3.28 | Sr XVIII \( 3d^4s^2-3d^4s^4f^1 \) | 3.11 | 0.027 | 3.12 | 0.027 |
| d        | 3.50 | Sr XVII \( 3d^4s^2-3d^4s^4f^1 \) | 3.16 | 0.028 | 3.17 | 0.028 |
| e        | 3.70 | Sr XX \( 3d^4s^2-3d^4s^4f^1 \) | 3.34 | 0.040 | 3.35 | 0.039 |
| f        | 3.97 | Sr XIX \( 3d^4s^2-3d^4s^4f^1 \) | 3.71 | 0.055 | 3.72 | 0.056 |
| g        | 4.27 | Sr XVIII \( 3d^4s^2-3d^4s^4f^1 \) | 4.07 | 0.066 | 4.08 | 0.063 |
| h        | 4.38 | Sr XVII \( 3d^4s^2-3d^4s^4f^1 \) | 4.07 | 0.066 | 4.08 | 0.063 |
| i        | 4.56 | Sr XVII \( 3d^4s^2-3d^4s^4f^1 \) | 5.02 | 0.098 | 5.03 | 0.099 |
| j        | 4.63 | Sr XIII \( 3d^4s^2-3d^4s^4f^1 \) | 5.09 | 0.063 | 5.12 | 0.067 |
| k        | 4.77 | Sr XIII \( 3d^4s^2-3d^4s^4f^1 \) | 5.19 | 0.140 | 5.21 | 0.134 |
| l        | 5.06 | Sr XI \( 3d^4s^2-3d^4s^4f^1 \) | 5.27 | 0.073 | 5.29 | 0.067 |
| m        | 5.23 | Sr XIII \( 3d^4s^2-3d^4s^4f^1 \) | 5.49 | 0.115 | 5.50 | 0.115 |
| n        | 5.47 | Sr XI \( 3d^4s^2-3d^4s^4f^1 \) | 5.62 | 0.165 | 5.66 | 0.157 |
| o        | 5.61 | Sr XIV \( 3d^4s^2-3d^4s^4f^1 \) | 5.69 | 0.165 | 5.71 | 0.157 |
| p        | 6.00 | Sr XIV \( 3d^4s^2-3d^4s^4f^1 \) | 6.03 | 0.130 | 6.05 | 0.130 |
| q        | 6.19 | Sr XIV \( 3d^4s^2-3d^4s^4f^1 \) | 6.27 | 0.190 | 6.30 | 0.180 |
| r        | 6.67 | Sr XIV \( 3d^4s^2-3d^4s^4f^1 \) | 6.68 | 0.139 | 6.71 | 0.137 |
| s        | 6.96 | Sr XIV \( 3d^4s^2-3d^4s^4f^1 \) | 6.96 | 0.214 | 7.00 | 0.204 |
| t        | 7.47 | Sr XIV \( 3d^4s^2-3d^4s^4f^1 \) | 7.46 | 0.129 | 7.50 | 0.121 |
| u        | 7.79 | Sr XIV \( 3d^4s^2-3d^4s^4f^1 \) | 7.79 | 0.232 | 7.85 | 0.222 |
| v        | 8.41 | Sr XIV \( 3d^4s^2-3d^4s^4f^1 \) | 8.41 | 0.232 | 8.48 | 0.222 |

Observed in the 3.0–5.3 nm and 3.7–7.9 nm regions from Sr\(^{10+}\) to Sr\(^{18+}\) respectively. The satellite transitions from \( 3d^4f \) and \( 3d^4p \) appear on the shorter wavelength side of their associated resonant transitions. The widths of the transition arrays depend on the number of lines in each array. As can be seen in table 1 and figure 5, the transitions are simply line spectra at 10+ and 19+ due to the closed shell structure, whereas around 14+ and 15+, the open shell structure gives rise to a wide spectral shape. The satellite transitions, from states with one electron in the 4s orbital are observed in ions as high as 18+. Figure 6 shows a comparison of the experimental spectrum recorded from a laser plasma formed with the 170 ps pulse with synthetic spectra derived from the \( gA \) values obtained from the Cowan code and FAC calculations. Both the value of the calculations as an aid to interpreting the experimental spectra, and the similarity between the output from the two codes, are evident in figure 6. Figure 7 shows the scaled mean positions and widths as a function of the degree of ionisation of the \( 3d^4f \) and \( 4f \) resonant transitions and the \( 3d^4s^2-3d^4s^4p \) and \( 3d^4s^2-3d^4s^4f \) satellite transitions, from \( gA \) values calculated using the Cowan code and the FAC code. The mean energy and widths of the transition arrays, calculated using the procedure of Bauche et al., are scaled by \( \zeta \), where \( \zeta = Z - (N - 1) \) \([22]\). Here, \( \zeta \) is the residual charge, \( Z \) is the number of electrons in the neutral ion and \( N \) is the number of electrons in the ionised ion. For \( 3d^4f \) and \( 4f \) transitions, the satellite transitions lie at higher energies than the resonant transitions, the \( \zeta \) being smaller than \( Z \).
transitions for a given ion. The $3d$–$4p$ transitions tend to higher energies with increasing ionisation, while the $3d$–$4f$ transitions are almost constant in energy.

5. Conclusion

Soft x-ray emission spectra from Nd:YAG laser produced strontium plasmas in the wavelength range 2.0–9.0 nm were observed and features identified with the aid of Cowan and FAC code calculations and comparison with similar spectra from neighbouring elements. The spectra are dominated by the $\Delta n = 1$ resonant lines $3d$–$4p$–$4f$ and satellite lines $3d4s$–$4s4p$–$4s4f$, emitted by 10+ to 19+ ions. Above 3.5 nm the

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**Figure 5.** The positions of $gA$ values of $\Delta n = 1$ transitions of the $3d^{n-1}3d^{n-1}4p^1$ (red), $3d^{n-1}4s^1$–$3d^{n-2}4s^14p^1$ (blue), $3d^{n-1}3d^{n-1}4f^1$ (green) and $3d^{n-1}4s^1$–$3d^{n-2}4s^14f^1$ (light blue) in the spectra of Strontium ions calculated using the Cowan code. The $gA$ values are normalised and binned to match the pixel interval of the CCD camera on the spectrometer.

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**Figure 6.** Comparison between the experimental spectrum from a plasma formed with a 170 ps laser pulse (top) and spectra simulated from the Cowan code (middle) and FAC (bottom). The $gA$ values were normalised and binned to match the pixel interval of the CCD camera on the spectrometer. They were not weighted to account for ion populations.

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**Figure 7.** The degree of ionisation with scaled mean positions and widths of $3d^{n-1}3d^{n-1}4p^1$ (red), $3d^{n-1}4s^1$–$3d^{n-2}4s^14p^1$ (blue), $3d^{n-1}3d^{n-1}4f^1$ (green) and $3d^{n-1}4s^1$–$3d^{n-2}4s^14f^1$ (light blue) transitions be calculated by Cowan code and FAC.
spectrum is increasingly influenced by satellite transitions. The strong emission in the water window region of the spectrum makes strontium plasmas interesting candidates as light sources for work with multilayer optics in this region.

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