Laser Produced Plasma Light Sources for Short Wavelength Applications

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Outline

• Laser Produced Plasma Properties

• Line Sources.

• $\Delta n = 0$ UTA

• $\Delta n = 1$ UTA

• Conclusions
Laser produced plasmas (LPPs)

Expansion velocity $\approx 10^6 - 10^7$ cms$^{-1}$

Average charge $\approx 0.67 (ZT_e)^{1/3}$

Temperature depends on laser power density ($\Phi$),
$T_e$(eV) $\alpha (\lambda^2\Phi)^{3/5}$ ...CR model

Critical density (absorption front)

High density plasma

For ns duration plasmas
Collisional Radiative (CR) Equilibrium assumed

Critical electron density, $n_{ec} = 10^{19} - 10^{21}$ cm$^{-3}$ depends on laser wavelength
($n_{ec} \sim [10^{21}/\lambda(\mu m)^2]$ cm$^{-3}$)
Effect of Increasing Power Density $\Phi$

Ion populations and average ionization of a Gd plasma as a function of $T_e$ computed with the Collisional Radiative (CR) model.

Increasing charge state requires higher power density and tighter focusing that causes increased kinetic losses due to higher temperature.

High power density usually means tighter focus, increased kinetic loss due to lateral expansion.

Competition between kinetic and radiative losses.

Harilal et al. used a CO$_2$ pulse with $\tau = 25 - 55$ns. Typically FWHM = 30 ns, $\Phi = 6 \times 10^9$ W cm$^{-2}$.
Typical LPP EUV Spectrum

Spectrum consists of:

- lines (bound-bound transitions), because of high density, lines from high n states are usually not seen.

**Strongest lines from closed shell or single electron outside closed shell species.** Also in LPPs spectra often dominated by these species (especially at short pulse lengths)

- recombination radiation (bound-free transitions): \( I \propto n_e^2 \langle z \rangle^4 \) where \( \langle z \rangle \) is the average ionic charge

- bremsstrahlung (free-free): \( I \propto n_e^2 \)

In some cases lines cluster together to form a UTA (unresolved transition array)

**Strongest lines always involve resonance transitions to the ground configuration**
Most important isoelectronic sequences for line emission

Because of increase in ionization potential required to remove an electron from a closed shell, get large populations of closed shell and single electron outside closed shell species. Spectra are relatively simple, so lines are strong. Must allow for level rearrangement with ionization.
Subshell ordering with increasing ionization

Ground configurations can change along isoelectronic sequences, levels reorder by principal quantum number.

• In Ca I ground configuration is \((\text{Ar})\ 4s^2\), this changes to \((\text{Ar})\ 3d^2\).
• Hyperalkali ions: PmI: \(4d^{10} 5s^2 6p^6 4f^5\) changes to \(4d^{10} 4f^{14} 5s\) at the 15\(^{\text{th}}\) ion stage along the sequence \((\text{Curtis and Ellis PRL 45, 2099 1989})\).
• Xe sequence: transitions based on \(5p^6\) vanish at Pr VI, ground configuration changes to \(\{5p4f\}^6\).

**Reason:**

The effective radial potential is of the form:

\[-Z'e^2/4\pi\varepsilon_0 r + l(l+1)h^2/8\pi^2 r^2\]

in neutrals and low ion stages the centrifugal term dominates and causes irregularities in filling of subshells. With increasing ionisation the Coulomb term becomes dominant..............H-like structure
Early studies @13.5 nm, line emission CE

Laser wavelength dependence of extreme ultraviolet light and particle emissions from laser-produced lithium plasmas

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**Line Source:**
Li: 1s-2p transition at 13.5 nm

Conversion efficiency (CE)
~ 2.3% @ λ = 532 nm,
~ 1.8% @ λ = 1064 nm

Up to 4% using forced recombination predicted.

Spot size ~400μm, τ = 10 ns
CE dropped at small spot sizes due to lateral expansion

More ablation at λ = 532 nm,
Opacity reduced output at 355 nm
1s-2p lines in He- and H-like oxygen

Spectrum of Sm: the strongest lines are actually due to 1s-2p resonance transitions in H- and He-like oxygen. The Sm lines are from Ni- and Cu-like Sm ions.

Lines in lower ion stages from lower Z targets more intense.
Two types of UTA in XUV spectra

$\Delta n = 0$

Increasing power density ↓

$4p^64d^N - 4p^64d^{N-1}4f + 4p^54d^{N+1}$ in Sn @13.5 nm

$\Delta n > 0$

$\Delta n = 0$ transitions overlap in adjacent ion stages.

$\Delta n > 0$ transitions do not overlap in adjacent ion stages and move to shorter wavelengths with increasing ionization.
Δn = 0 UTA: important isoelectronic sequences

3p - 3d

4d - 4f

4p - 4d

Periodic Table of Elements
$3p-3d \ \Delta n = 0 \ \text{UTA}$

$3p^n-3p^{n-1}3d$ UTA, important in 2$^{\text{nd}}$ transition row, Broad ($\sim 20$ eV), overlaid with a few strong lines.

Relatively weak. Spin orbit split, effectively two arrays in SXR.
4p-4d $\Delta n = 0$ UTA

$4p^n - 4p^{n-1}4d$ UTA, important in 3rd transition row and lanthanides and beyond,

Spin orbit split, overall, relatively weak and high energy array lies on the short wavelength side of the 4d-4f UTA
Evolution with $Z$ of $\Delta n=0$, $n=4 - n=4$ UTAs

- $13.5$ nm Litho: $T_e \sim 40$ eV
- $6.7$ nm Litho: $T_e \sim 100$ eV
- WW Imaging: $T_e > 500$ eV

Plasma electron temperature required increases with $Z$.
Spectral narrowing due to Configuration Interaction

For $4p^64d^{N+1} - 4p^64d^N4f + 4p^54d^{N+2}$
configuration interaction (CI) causes
- Spectral narrowing.
- Strong peaking of oscillator strength.

(Mandelbaum et al. PRA 35, 5051 (1986),

Spectral narrowing greatly enhances UTA intensity

$5d$-$5f$ UTA weaker as CI effects are less important.
CI regroups component lines

CI groups $4d^N \rightarrow 4p^5 4d^{N+1}$ transitions on high energy side. $4d^N \rightarrow 4p^6 4d^N 4f$ transitions lie at lower energy side of the overall array which is shifted to higher energy. Kilbane and O’Sullivan PRA 82, 062504 (2010)
4p-4d and 4d-4f arrays separate as $Z$ increases

Note that 4d-4f moves to longer wavelength with increasing ionization

$4p^64d^{N+1} - 4p^64d^N4f$

$4p^54d^{N+2} - 4p^64d^{N+1}$

$\delta E = N \frac{(2l + 1)(2l' + 1)}{4l + 1} \left( \sum_{l \neq 0} f_k F_k(l, l') \right) + \sum_k g_k G_k(l, l')$
Observed $\Delta n=0$ transitions in Pt

Comparisons of measured and calculated emission spectra of Pt. ($\Delta n=0$, $4p^64d^m\rightarrow 4p^54d^{m+1}+4p^64d^{m-1}4f$) (Pt XXXIV-Pt XLII). The theoretical spectra are calculated gA-value distributions.

The strongest lines are observed from $4d^N \rightarrow 4p^64d^N4f$ transitions. Lines from $4p^64d^m\rightarrow 4p^54d^{m+1}$ do not contribute strongly.
Detailed analysis of $4p^64d^n \rightarrow 4d^{n-1}4f$ UTA

Initial analysis for Sn by Churilov and Ryabtsev *Phys. Scr.* 73 614-619, (2006) UTA mainly due to $\text{Sn}^{10+}-\text{Sn}^{13+}$: $4p^64d^n \rightarrow 4d^{n-1}4f + 4p^54d^{n+1}$

Subsequently Toretti et al *PRA* 95 042303 (2017) found that the earlier analysis is in need of revision. Major problem due to spectral complexity.

Levels of $4p^64d^N$ & $(4p^54d^{N+1}+4d^{N-1}4f + 4d^{N-1}5p)$ calculated with Cowan suite of codes. **Overlap in adjacent ion stages makes analysis almost impossible.**
Need to isolate the contribution from each ion stage

Use ECR or EBIT Source

ECR Source Spectra
At low gas pressure in target chamber, Sn ions from ECR capture one electron.

\[ \text{Sn}^{q+} + X \rightarrow \text{Sn}^{(q-1)+*} + X^+ \]

Obtain charge state selected spectra

In Sn XV, ground state is \( 4p^6 \), expect \( 4p - 4d \) \( ^1S_0 - ^1P_1 \) transition, one strong line. Observe a UTA, due to excited to excited state (\( 4d - 4f \)) transitions CX spectra dominated by satellite emission
(D'Arcy et al. PRA 79, 042509, 2009)
EBIT spectra at higher electron accelerating voltages give spectra from higher ion stages, however, they are generally very weak and recorded at low resolution. Good for strongest lines. Need for high resolution LPP or vacuum spark spectra in tandem with low resolution EBIT data for a complete analysis.
Atomic rate data required for plasma modelling

Detailed investigation of electron-impact single-ionization cross sections and plasma rate coefficients of N-shell tin ions

A Borovik Jr, M F Gharaibeh, P M Hillenbrand, S Schippers, and A Müller

Figure 1. Overview over the present single-ionization cross sections of Sn$^{4+}$ through Sn$^{13+}$ ions. The cross-section scale is in units of $10^{-17}$ cm$^2$ and the cross section for each next ion stage is shifted downwards by $1 \times 10^{-17}$ cm$^2$ and multiplied by a certain coefficient provided on the graph. The thin kinked polygon connects the ground-state ionization thresholds marked by short vertical bars.

Figure 2. The present single-ionization cross section of Sn$^{4+}$ (open circles, absolute cross-section data; grey thin solid line, energy scan) compared to the results of the present CADW calculations. The individually shaded areas show contributions of processes involving the indicated subshells. DI and EA denote direct ionization and excitation-autoionization processes, respectively. Th

Only experimental data
Crossed beam experiment.
Ions produced by an ECR source
Photoabsorption by Dual Laser Plasma Method

Experiment:

Dual Laser Plasma Method.

1 mm line LPP on target probed by EUV continuum emission from a Sm LPP at different time delays $\Delta \tau$ between pulses.
Photoabsorption of Sn, Sn$^+$ and Sn$^{3+}$, and comparison with calculation

**Theory:**
TDLDA Code **David**

**Result:**
\[ \sigma = 10 \pm 2 \text{ Mb} \]
Opacity Effects in $\Delta n = 0$ n=4-n=4 arrays

- Spectra from metal targets dominated by continuum emission
  - Some strong emission and absorption lines
  - Large **contribution from satellite lines**

- Spectra of low density targets dominated by an intense Unresolved Transition Array (UTA), with greatly reduced continuum emission
  - Emission largely mirrors line strength distribution
LHD spectra of Sn

- Sn injected using a TESPEL.
- Spectra dominated by resonant line emission to the ground state.
- Absorption free, emission mirrors \( gA \)-values.

(Suzuki et al JPB 4, 074027, 2010)
Effect of Laser Wavelength: CO$_2$ vs. Nd:YAG

1-D Hydro modeling – demonstrated increase in CE for CO$_2$

CE for CO$_2$ ~ 2x Nd:YAG value.  

Temperature depends on laser power density (Φ).  
\[ T_e (eV) \approx (\lambda^2 \Phi)^{3/5} \]

Average charge \(\approx 0.67 \left( A T_e \right)^{1/3}\)

\[ n_{ec} \approx 10^{21} \lambda^{-2} \text{ (cm}^{-3}\text{)} \]

1,064 nm:  \( n_{ec} \approx 1 \times 10^{21} \text{ cm}^{-3}\)

10,600 nm:  \( n_{ec} \approx 1 \times 10^{19} \text{ cm}^{-3}\)

- Less energy, better CE
- Improved opacity
- Lower power density required

Optical depth \(\alpha \propto \text{pulse duration} \times (\text{Intensity})^{5/9} \times (\lambda)^{-4/3}\)

Increases with pulse duration, decreases with laser wavelength.  
*(Ando et al. APL 89, 151501, 2006, Sunahara et al Plasma and Fusion Res.3,43 2008)*
Recent results for Sn with short pulse CO$_2$ Laser

CO$_2$ pulse produced by TEA system
Pulse shortened by Ge semiconductor plasma shutter.
Shortened pulse then amplified

Amano et al. JJAP. Submitted 2018
Temporal profiles of the CO₂ laser pulses before (a) and after pulse slicing by the Ge semiconductor shutter by one control 150-ps pulse (b) and two control 150-ps and 10-ns pulses (c). (d) The CO₂ laser beam profile at a pulse duration of 3 ns after the amplifier.

CE ~6% obtained with 8 ns pulse.
Conversion efficiency dependence on CO$_2$ laser intensity for single (dashed) and double (solid) irradiation by a 10 ns pulse. The interpulse delay was 180 ns (Nishihara et al Phys. Plasmas 15, 056708 2008)

Max CE @ 60 ns delay close to wedge centre
Nd:YAG, E~ 170 mJ, Φ = 1.5x10$^{11}$ Wcm$^{-2}$
CO$_2$: E~ 200 mJ, Φ = 4x10$^{9}$ Wcm$^{-2}$
CE = 3.33±0.16%
For CO$_2$ only, CE = 4.85±0.10%
Allowing for overfilling of plasma by CO$_2$
CE approximately 7%

Since CE ~6% obtained with single 8 ns pulse > 6% should be achievable with careful dual pulsing
4d-4f UTA at shorter wavelengths: 6.x nm

Optimum temperature for an optically thin Gd plasma ~110 eV. Maximum intensity at 6.76 nm due to $4d^{10}1S_0 - 4d^94f^1P_1$ line.

Hybrid UTA-line source

The most important transitions occur in Ag-like and Pd-like: Gd XVIII-XIX, Tb XIX & XX
i.e. Ions with $4d^{10}4f$ and $4d^{10}$ and ground states

$3 \text{ J in 20 ns, } \lambda = 1.06 \mu m$
$\Phi = (5-8) \times 10^{11} \text{ Wcm}^{-2}$

CE improves as concentration decreases 
Otsuka et al. APL 97, 111503 (2010)

CE Experiment at Gekko XII

12 beams from Gekko XII, $\lambda=1.053\,\text{mm}$ $E=1\,\text{J}$, $E(\text{Total})=12\,\text{J}$, $\tau=1.3\,\text{ns}$. Quasi1-D expansion for each beam. (Yoshida et al Appl. Phys. Exp. 7, 086202 2014)

CE = 0.8%

CE lower than Sn as higher fraction of laser energy goes into plasma heating/ionization
\[ \Delta n = 0 \text{ 4-4 UTAs in the water window region, Au, Pb and Bi (LHD Spectra)} \]

- Spectra dominated by resonant line emission to the ground state
- Only Ag-, Pd- and Rh-like ions give line emission
- Absorption free (Ohashi et al. JPB 48 (2015) 144011)

**All require Te > 500 eV for generation**
LPP Spectrum of Pb

\[ 4d^n - 4d^{n-1}f \]
\[ 4f^n - 4f^{n-1}g \]
\[ 4d^n - 4d^{n-1}p \]
\[ 4p^n - 4p^{n-1}4d \]
\[ 4p^64d^{N+1} - 4p^54d^{N+2} \]
\[ 4p^64d^{N+1} - 4p^64d^N4f \]
Dual Plasma Irradiation of Solid Bi

Pulse separation time dependence of the SXR emission from dual-laser-produced plasmas for pre-pulse durations of 10 ns (a) and 150 ps (b).

Pulse separation time dependences of the number of photons in the water-window SXR spectral region (2.34 – 4.38 nm) at pre-pulse durations of 10 ns (c) and 150 ps (d)

Relative spectral intensity enhancement \[ \frac{I(l) - I_0(l)}{I_0(l)} = \frac{\Delta I(l)}{I_0(l)} \] for pre-pulses of 10 ns duration (e) and 150 ps duration (f).

Potentially brightest 4d-4f UTA?

Width of UTA:
In Sn: 4d-4f transitions extend over a large wavelength range and are weak in states up to Sn $^{9+}$. Minimum width in rare earths, ~ Ce and Pr due to complete contraction of 4f wavefunction and almost constant value of $\langle 4d | 4f \rangle$. 
UTA in Cs- Nd

Ce brightest in early work 4.5% CE into 3% Bandwidth at 5.8 nm (Dunne et al. APL 76, 34, 2000)

$\Delta n = 1$ UTAs

Periodic Table of the Elements

- **Alkali Metal**
- **Alkaline Earth**
- **Transition Metal**
- **Basic Metal**
- **Semimetal**
- **Nonmetal**
- **Halogen**
- **Noble Gas**
- **Lanthanide**
- **Actinide**

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Early target studies @13.5 nm

Δn = 1 UTA:
$\text{Xe}^{10+}$ in-band 13.5 nm emission due to $4p^64d^8 - 4p^64d^75p$ resonance transitions.

Max. CE on solid Xe target ~1%
Δn=1 transitions in 2\textsuperscript{nd} transition row

Δn=0, 3p-3d

Δn=1, 3d-4p and 3d-4f

To produce the ion stages required (up to ~23+:

$T_e \sim 300$ eV, $\Phi \sim 2 \times 10^{12}$ Wcm\textsuperscript{-2}
Multilayer mirrors in the EUV/SXR

<table>
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<th>Material</th>
<th>Wavelength (nm)</th>
<th>Reflectivity (%)</th>
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<tr>
<td>Cr/V</td>
<td>2.42</td>
<td>9</td>
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<tr>
<td>Cr/Ti</td>
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<td>TiO$_2$/ZnO</td>
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<td>Cr/Sc</td>
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<tr>
<td>Cr/Sc</td>
<td>3.14</td>
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<tr>
<td>Cr/Sc B$_4$C</td>
<td>3.15</td>
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<tr>
<td>Cr/Sc</td>
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<td>10</td>
</tr>
<tr>
<td>Cr/Sc</td>
<td>3.37</td>
<td>5.5</td>
</tr>
</tbody>
</table>

Mo/Si

13.5 nm Litho

6.0 nm Litho

Water window
Integrated Intensity in the Water Window due to different UTAs

Comparison of the time-integrated emission spectra in the soft x-ray spectral region from laser-produced plasmas of Zr (a), Nb (b), Mo (c), Au (d), Pb (e), and Bi (f).

Laser power density dependence of the number of photons emitted in the water window
Dual Laser Illumination of Mo
Plasmas created by a 10 Hz, 805 nm, 10 mJ, 65 fs Ti:sapphire Laser. 

$f = 150$ mm, energy at target: 4.5 mJ. The laser spot diameter ~50 μm,

$\Phi = 3 \times 10^{15}$ W/cm².

3d–4f transitions arrays from Ru XXI-XXIII, Rh XXI-XXIII and Pd XXI-XXIII are clearly seen in the observed spectra.

(Lokasani et al. JPB 50(14) 2018.)
Conclusions

• Still more CE can be attained at 13.5 nm. Modelling needs more atomic data. Solid state mid-IR lasers could give better beam profiles (spatially and temporally).

• Highest CE for $\Delta n = 0$ UTA in 2-3% BW around Ce or Pr. (8-8.8 nm).

• $\Delta n=1$ transitions in medium and high Z elements and $\Delta n=0$ in high Z elements can be used for water window sources.

• $\Delta n=1$ transitions require less energy for excitation than $\Delta n=0$. Also some match existing MLMs.

• Ideal source ideally depends on mirror bandwidth. For very narrow bandwidth at low wavelength H-like 1s-2p line in low Z ions best. Water/ammonia/organic liquid droplet, dual ps pulse irradiation.
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