Fundamental Atomic Process in Source Development for Beyond EUV Lithography and “Water Window” imaging

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Beyond 13.5 nm lithography (BEUVEL)

New sources would be needed at 6.\(\times\) nm for the future lithography beyond 13.5 nm.

The precise value of \(\times\) will be determined by the source and multilayer mirror combination that provides optimum efficiency and in-band EUV yield and its determination is a priority.
Energy levels and transitions of Gd

Energy-level diagram.

(1) Gd XII – Gd XVIII: 5d \(4d^{-1}\) 5g and 4f.

(2) Gd XIX – Gd XXVIII: 4p\(^{-1}\) 4f 5f 5p and 4d
Most important ion stages?

The most important transitions can be inferred from studies of W spectra.

They occur in Ag-like, Pd-like and Rh-like $W^{27+}$-$W^{30+}$. Sugar et al JOSA 10, 1321 (1993)

Gd XVIII-XX, Tb XIX - XXI

In all reported studies the emission is dominated by Ag and Pd-like lines i.e. the spectra containing fewest lines where the emission is not divided amongst many transitions.

i.e. ions with $4d^{10}4f$, $4d^{10}$ and $4d^9$ ground states
Power density required?

Steady-state CR model

\[ f_z = \frac{n_{z+1}}{n_z} = \frac{S(z)}{\alpha_r(z+1) + n_e \alpha_{3b}(z+1)} \]

- \( n_z \) = density of ion \( z \)
- \( n_e \) = electron density
- \( S \) = collisional ionisation rate coefficient
- \( \alpha_r \) = radiative recombination rate coefficient
- \( n_e \alpha_{3b} \) = three-body recombination rate coefficients and \( T_e \) = electron temperature

D. Colombant and G. F. Tonon, J. Appl. Phys. 44 (1973) 3524

The laser power density required lies in the range

- \( 2 \times 10^{12} \) - \( 10^{13} \) W cm\(^{-2} \) @\( \lambda = 1.06 \) μm
- \( 2 \times 10^{11} \) - \( 10^{12} \) W cm\(^{-2} \) @\( \lambda = 10.6 \) μm

80 – 130 eV !!
The synthetic spectra

Gd: $x=0.76$, optimum temperature $\sim 110$ eV
Tb: $x=0.51$, optimum temperature $\sim 120$ eV
Perfect mirror:
LaN/B: maximum reflectance at 6.66 nm
LaN/B₄C: maximum reflectance at 6.63 nm
Position of peak emission and purity

Calculated position of peak emission from Tb (left) and Gd (right) spectra as a function of electron temperature. Plots of spectral purity as a function of electron temperature are also included for three 0.6% reflectivity bands: one centered at the wavelength of peak emission and the other two as indicated.
Influence of dielectronic recombination

Theoretical investigation of dielectronic recombination of Sn^{12+} ions

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What is DR process

Incident electron

bound electrons

Initial state

Resonant capture

Radiative transition

\[ e + A^{q+}(i) \rightarrow [A(q-1)+]^{**}(j) \rightarrow [A(q-1)+]^*(k) + hv \]

\[ [\text{Zn}] 4p^64d^9 + e \rightarrow [\text{Zn}] 4p^64d^8n'nl, \ n' = 4f, 5l, 6l, \]

\[ + 4p^54d^{10}nl + 4p^54d^8n'l'n'l', \ n' = 4f, 5l, 6l, \]

\[ [\text{Zn}] 4p^64d^8n'l'nl + 4p^54d^{10}nl + 4p^54d^9n'l'n'l' + hv, \]

\[ \rightarrow [\text{Zn}] 4p^64d^9 nl + 4p^64d^8 n'l'n''l'' + hv, \]

\[ \downarrow \text{RS NRS} \]

\[ [\text{Zn}] 4p^64d^9 + e \]
Dielectronic recombination of Pd-like gadolinium

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As research and development of extreme ultraviolet lithography (EUVL) sources at 6.7 nm (which will be based on emission from ionized gadolinium) has already begun, reliable atomic data are required in order to determine the optimum plasma conditions. However, the complexity of the atomic structure means that ab initio level-resolved dielectronic recombination (DR) calculations are currently unavailable for the ions of interest. Here we report the first detailed calculation of the DR rate coefficients for the ground state and first excited states of Pd-like gadolinium. Energy levels, radiative transition probabilities, and autoionization rates of Ag-like gadolinium for \([\text{Kr}]4d^94f\;\text{nl}\), \([\text{Kr}]4p^54d^{10}4f\;\text{nl}\), \([\text{Kr}]4d^95s\;\text{nl}\), and \([\text{Kr}]4d^96l\;\text{nl}\) \((n \leq 18)\) complexes were calculated using the flexible atomic code (FAC). It was found that inclusion of \(4p^54d^{10}4f\;\text{nl}\) configurations has significant influence on the total DR rate coefficient. The DR rate coefficients obtained here are compared with radiative recombination and three-body recombination coefficients. The results show that the DR rate coefficient is almost an order of magnitude higher than the coefficients for the other two recombination processes combined at plasma electron temperatures around 110 eV, which suggests that the DR process should be included in theoretical modeling for Pd-like gadolinium in EUVL source plasmas.

Dielectronic recombination of Rh-like Gd and W

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Energy levels of Gd and W

Figure: The energy levels near the ionization limits for 4d complexes

Figure: The energy levels near the ionization limits for 4p complexes
The 4p complexes contribute around 25% to the total DR rate coefficients.

The contributions from NRS transitions are significantly enhanced for W when compared with Gd as a result of lowering of energy levels relative to the ionization limit.
Influence of DR process

Burgess-Merts (BM) approximation

Bauche et al. HEDP5 (2009)51
Future work: Ga/Ge?

Galinstan deserve a detailed study:
1) low melting temperature
2) current Laser assisted DPP can be directly transferred
Future work: Complex target?

Experiment: T. Faulkner

Complex target by using nano-particles or colliding plasma might be a solution!

Experiment: Takeshi Higashiguchi
Ionic fraction for complex target
The emission processes in Gd/Tb is more efficient than in Sn because the 4f wavefunction is completely contracted in the relevant ions. Strongest lines are expected from Ag-, Pd- and Rh-like ions.

- Gd: $x=0.76$, optimum temperature $\sim 110$ eV.
- Tb: $x=0.51$, optimum temperature $\sim 120$ eV.
- Dielectronic recombination process is important.

Future work:
modeling plasma hydrodynamics and radiation transport, to find both the optimum temperature and source conditions.
**Water Window:** 2.3–4.4 nm. In this region, the absorption of carbon is approximately 10 times higher than that of oxygen and water.

**Potential Commercial Uses:** x-ray microscopy, absorption spectroscopy, etc.

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**Fig. 1.** "Water window" X-ray energy range.  
**Fig. 2.** Onion skin cells, 450eV X-ray energy, 800nm pixel size.

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A. Sasov  
X-ray microscopy of living cells  
5th IEEE international symposium on biomedical imaging

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**Proposed by Prof. O’Sullivan & Prof. Endo**

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Bi as a choice ($\Delta n = 0$)

Feasibility study of broadband efficient “water window” source

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Zr as a choice ($\Delta n = 1$)

**Figure 1.** Comparison of the experimental spectrum of Zr with theoretical resonance transitions. 3d–4f and 3d–4p transitions are clearly seen for each ion with a 3d$^n$ ground configuration.

**Figure 2.** Energy-level diagram showing the level structure of 3d$^n$, 3d$^{n-1}$ (4s, 4p, 4f) and 3d$^{n-2}$ 4s (4p, 4f) configurations for each ion stage in ascending order. The ionization limit is indicated by the long solid line.

Spectator transition: $3d^{n-1}4s \rightarrow 3d^{n-2}4s4f$

Populating process

(a)

\[ \frac{dN(k)}{dt} = R^{cc}(i)N_e N(j) + R^{ee}(j,k)N_e N(i) - (R^{ee}(k,j)N_e + A^e(k,j) + A^r(k,i))N(k) \]

= 0

DC – Dielectronic capture; AI – Autoionization; EIE – Electron impact excitation; DE – De-excitation; AR – Radiative transition

(b)
Spectator satellite transition is important!

**Cyan lines:** Without satellite lines

**Red line:** With satellite lines
Influence of bandwidths and power densities

Figure: Variation in total counts as a function of multilayer reflective bandwidth for water window emission from 150-ps (a) and 10-ns (b) laser-produced plasmas of Zr.

Figure: Water window emission (total counts) as a function of power density for 150 ps (a) and 10 ns (b) laser pulses.
Further work

Emission properties of ns and ps laser-induced soft x-ray sources using pulsed gas jets

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lengths. Li et al. [35] observed a similar shift to shorter wavelengths with increasing pulse energy using a 150 ps Nd:YAG laser for plasmas induced on a solid state zirconium target. On the other hand, Li et al. note higher total emission intensities for ns laser-induced plasmas of Zr, Mo, C and N from solid targets compared to ps laser plasmas.
We have demonstrated a laser-produced plasma soft x-ray source in the water window spectral region using high-Z plasmas, like a Bi plasma as a water window source for biological microscopy.

\[ \Delta n = 1 \] transitions, like Zr, might be useful with narrow bandwidth reflector for water window source.

Dielectronic recombination satellite lines is important.
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