# Laser isotope separation and ion yield of $^{111}$Ag through the reduced density matrix

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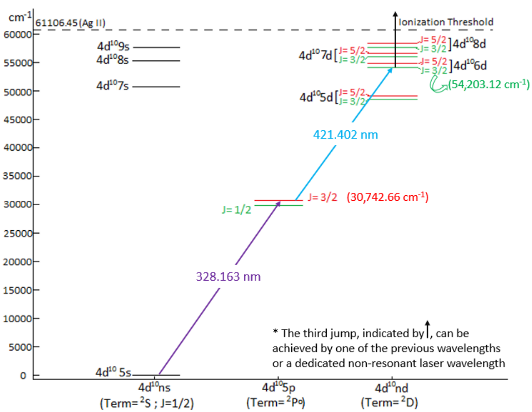
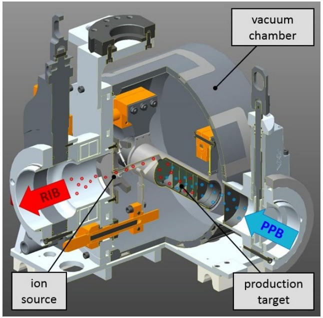
October 4, 2024

# Overview

# Aims

The **SPES Laser Ion Source (LIS)** plays a fundamental role in the **selective ionization** of exotic species. An element of particular interest is **silver**, as its isotope $^{111}$**Ag** is being studied for medical applications by **ISOLPHARM**.




This study investigates the ionization **efficiency** and **selectivity** for $^{111}$Ag after the offline tests. [Khwairakpam et al. (2023)]

# Overview

# Density matrix formalism

The interaction of an atom with its surroundings, including light and other atoms, can be modeled by the **density matrix**, whose **diagonal elements** correspond to the **level populations**:

$$
\begin{aligned}
\dot{\rho}_{11}(t) &= - i\Omega_{21}(t)\cdot\rho_{12}(t) + i\Omega_{12}(t)\rho_{21}(t) + 2A_{21}\cdot\rho_{22}(t)\\
\dot{\rho}_{22}(t) &= - i\Omega_{12}(t)\cdot\rho_{21}(t) + i\Omega_{21}(t)\cdot\rho_{12}(t) - i\Omega_{32}(t)\cdot\rho_{23}(t) + i\Omega_{23}(t)\cdot\rho_{32}(t)\\
&\quad + 2A_{32}\cdot\rho_{33}(t) - 2A_{21}\cdot\rho_{22}(t)\\
\dot{\rho}_{33}(t) &= - i\Omega_{23}(t)\cdot\rho_{32}(t) + i\Omega_{32}(t)\cdot\rho_{23}(t) - 2\big(A_{32}+\gamma_{\mathrm{i}}(t)\big)\cdot\rho_{33}(t)\\
\dot{\rho}_{\mathrm{ion}}(t) &= 2\gamma_{\mathrm{i}}(t)\cdot\rho_{33}(t)
\end{aligned}
$$

with $\mathrm{Im}(\rho_{ii}(t)) = 0\ \forall t$. $\Omega_{ij}(t)$ is the **Rabi frequency** of the $i \to j$ transition, $A_{ji}$ the **decay rate** (*Einstein A-value*) and $\gamma_{\mathrm{i}}(t)$ the **non-resonant ionization rate**.

- Due to the laser pulse, $\Omega_{ij}(t)$ and $\gamma_{\mathrm{i}}(t)$ have Gaussian shapes.

# Density matrix formalism

The **non-diagonal elements** represent instead the **coherences**:

$$\dot{\rho}_{12}(t) = i\Omega_{12}(t)\cdot(\rho_{22}(t)-\rho_{11}(t)) - i\Omega_{32}(t)\cdot\rho_{13}(t) - \left(i\Delta_1 + A_{21} + A_{32} + 2\gamma_{L1}\frac{\beta_1^2}{\Delta_1^2+\beta_1^2}\right)\cdot\rho_{12}(t)$$

$$\dot{\rho}_{23}(t) = i\Omega_{23}(t)\cdot(\rho_{33}(t)-\rho_{22}(t)) - i\Omega_{21}(t)\cdot\rho_{13}(t) - \left(i\Delta_2 + A_{21} + A_{32} + \gamma_{\mathrm{i}}(t) + 2\gamma_{L2}\frac{\beta_2^2}{\Delta_2^2+\beta_2^2}\right)\cdot\rho_{23}(t)$$

$$\dot{\rho}_{13}(t) = i\Omega_{12}(t)\cdot\rho_{23}(t) - i\Omega_{23}(t)\cdot\rho_{12}(t) - \left(i(\Delta_1+\Delta_2) + A_{21} + A_{32} + \gamma_{\mathrm{i}}(t) + 2\gamma_{L1}\frac{\beta_1^2}{\Delta_1^2+\beta_1^2} + 2\gamma_{L2}\frac{\beta_2^2}{\Delta_2^2+\beta_2^2}\right)\cdot\rho_{31}(t)$$

and $\dot{\rho}_{ji}(t) = \dot{\rho}_{ij}^*(t)$. $\Delta_i$, $\gamma_{Li}$ and $\beta_i$ are the $i$-th laser **detuning**, **Lorentzian FWHM** and **cut-off** according to the **phase diffusion model**. [Shore (1990); Suryanarayana and Sankari (2024)]

# Rabi frequency

For the **Rabi frequency**, *i.e.*, the oscillation frequency of the probability amplitudes of two levels, the following expressions can be used for **odd and even isotopes**:

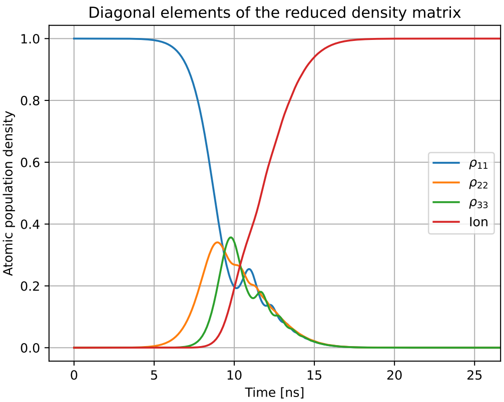
$$\Omega_{ij}^{\text{odd}}(t) = 2\sqrt{\frac{3gA_{ji}\lambda^3 p(t)}{8\pi hc} \cdot (2F+1)(2F'+1)} \cdot \begin{Bmatrix} J & F & I \\ F' & J' & 1 \end{Bmatrix}$$

$$\Omega_{ij}^{\text{even}}(t) = 2\sqrt{\frac{3gA_{ji}\lambda^3 p(t)}{8\pi hc}} \cdot \begin{pmatrix} J & 1 & J' \\ -m & q & m' \end{pmatrix}$$

Here, $g$ is the upper level degeneracy, $\lambda$ is the transition wavelength, $p(t)$ is the power density, $q = 0$ and $|q| = 1$ represent linear and circular polarization, $()$ and $\{\}$ are the Wigner 3-$j$ and 6-$j$ symbols and $F$ is the hyperfine quantum number determined by $\vec{F} = \vec{I} + \vec{J}$.

# Example

Study of a $^{91}Zr$ photo-ionization scheme in Doppler-free conditions. [Kumar et al. (2003)]



# Overview

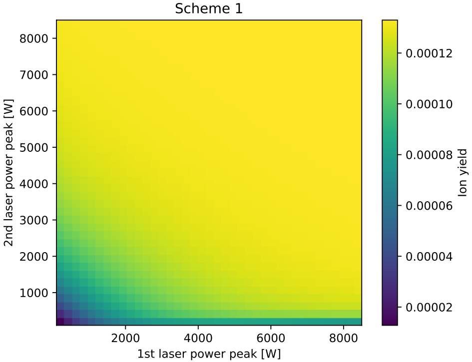
# Parameters

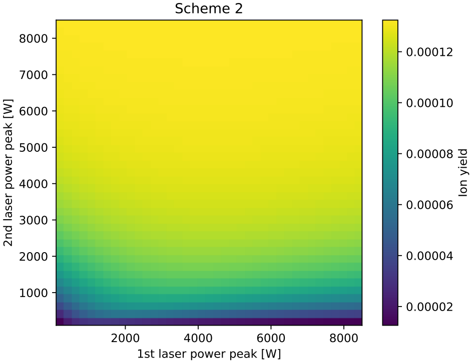
The model **parameters** were chosen as follows:

- $A_{21} = 1.380 \times 10^{-1}\,\text{ns}^{-1}$, $A_{32}^{J=5/2} = 2.606 \times 10^{-2}\,\text{ns}^{-1}$, $A_{32}^{J=3/2} = 4.362 \times 10^{-3}\,\text{ns}^{-1}$ [Smith et al. (2001)]
- Cut-off $\beta_i = 2 \cdot \gamma_{\mathrm{L}i}$
- Laser time pulse $\tau_{FWHM} = 30\,\text{ns}$ with total duration $T = 2 \cdot \tau_{FWHM}$
- Non-resonant ionization rate $\gamma_{\mathrm{i}}(t) \equiv \sigma\Phi(t) = \sigma \cdot \frac{p(t)}{h\nu}$ with $\sigma = 10^{-16}\,\text{cm}^2$
- Laser spot radius $R = 0.5\,\text{mm}$
- Wigner symbols: positive averaged

Isotopic selectivity and ion yield were studied varying $\gamma_{\mathrm{L}i}$ and the power peaks $P_i$.

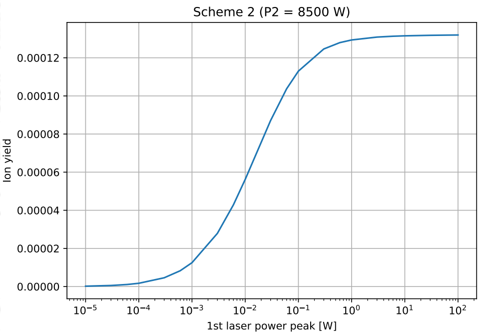
# Ionization efficiency

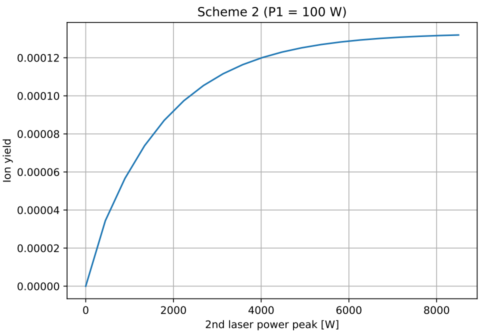



$\gamma_{L1} = \gamma_{L2} = 1\,\text{GHz}$ Doppler-free. $\gamma_{L1} = 1.5\,\text{GHz}$ and $\gamma_{L2} = 1.15\,\text{GHz}$ Doppler-broadened [Mariotti (2024)]: no difference, Doppler broadening appears negligible with respect to **power broadening**.

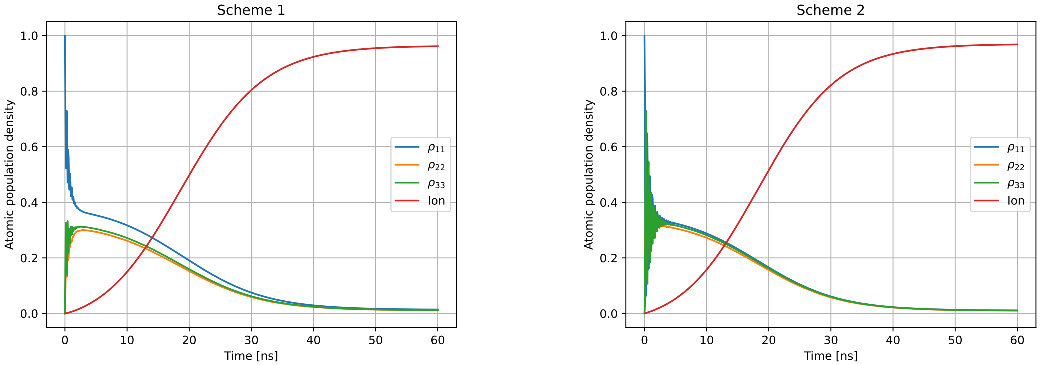
# Ionization efficiency




A good compromise between efficiency and photon economy is scheme 2, $P_1 = 100\,\mathrm{W}$, $P_2 = 6000\,\mathrm{W}$, giving a $1.287 \times 10^{-4}$ ion yield (normalized to LIS volume).

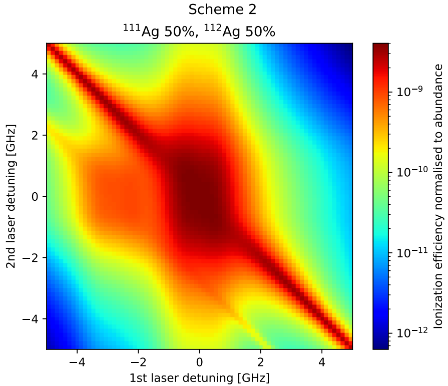
# Ionization efficiency

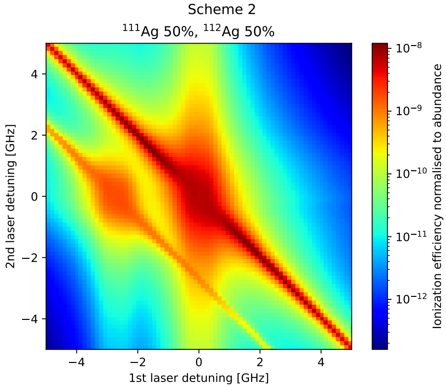



Population dynamics within $T$ (SciPy solver: RK45).

# Isotopic selectivity

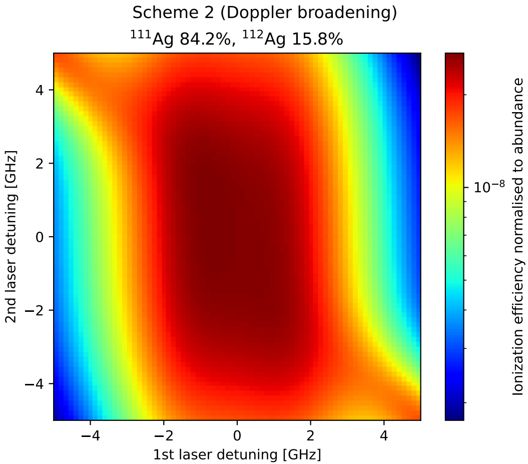
$^{111}$**Ag**: same nuclear spin $I = 1/2$ as stable $^{107,109}$Ag.
$^{112}$**Ag**: possible SPES contaminant; **isotopic shift** $\sim 2767.5\,\mathrm{MHz}$ [Jading et al. (1997)].




$\gamma_L = 500\,\mathrm{MHz}$ (left), $\gamma_L = 250\,\mathrm{MHz}$ (right); $P_1 = 1\,\mathrm{mW}$, $P_2 = 1\,\mathrm{W}$.

# Isotopic selectivity




More realistic case: **isotopic abundances** after a 6 h irradiation cycle at SPES [Mariotti (2024)], **Doppler broadening**. Here, $P_2 = 10\,\mathrm{W}$.

# Degree of enrichment

Table 1: Degree of enrichment $\xi$ and ionization efficiency $\varepsilon_{111}$ of some online configurations using scheme 2 (abundance $a_{111} = 0.842$, $\Delta_1 = \Delta_2 = 0$, $\xi \equiv a_{111}\varepsilon_{111} / \sum_i a_i\varepsilon_i$).

| $P_1$ [mW] | $P_2$ [W] | $\gamma_{L1}$ [GHz] | $\gamma_{L2}$ [GHz] | Doppler | $\varepsilon_{111}$ | $\xi$ |
|---|---|---|---|---|---|---|
| 1 | 10 | 1.50 | 1.15 | yes | $3.13 \times 10^{-8}$ | 0.979 |
| 1 | 10 | 1.0 | 1.0 | no | $4.51 \times 10^{-8}$ | 0.992 |
| 1 | 1 | 0.5 | 0.5 | no | $7.94 \times 10^{-9}$ | 0.999 |

- If $a_{111} = 0.5$, the Doppler-broadened configuration gives $\xi = 0.899$
- Efficiency seems very low, but it can be acceptable considering the **high repetition rate** online (10 kHz) and irradiation cycles lasting some hours

# Overview

# Next steps

- Repeating this study with **other medical radionuclides and radiotracers** obtainable at SPES
- Next candidates will be **Sc, I and Mg isotopes**
- Theoretical or experimental **theses** or **internships** available for Physics and Chemistry students

Thank you for your kind attention!

# References I

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