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MODIFIED MAXWELL-BLOCH EQUATIONS FOR ASE IN X-RAY LASERS

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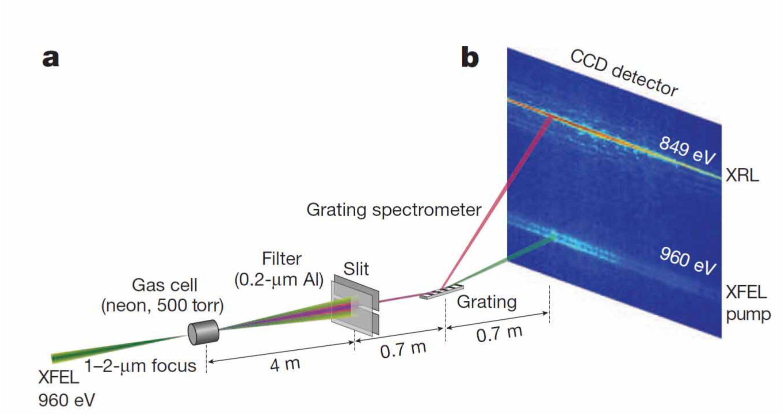
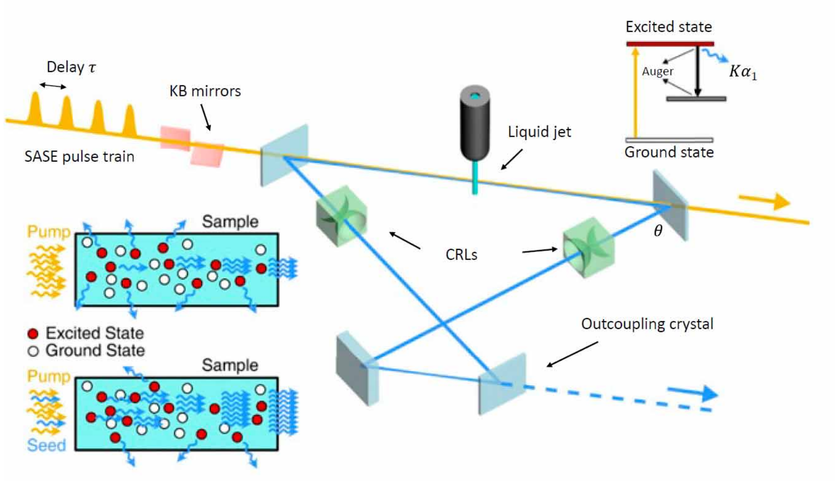
XFEL-PUMPED X-RAY LASER





- **Single-pass amplified spontaneous emission (ASE), super-fluorescence, or X-ray laser (XRL)**
  - N. Rohringer, et al., Nature, 481, 488 (2012): EuXFEL, ASE, Ne, $\hbar\omega$ =849 eV
  - H. Yoneda, et al., Nature, 524, 446(2015): SACLA, ASE and seeded, Cu, $\hbar\omega$ =8 keV
  - T. Kroll, et al., PRL 120, 133208 (2018), LCLS, transition metal, spectroscopy
- **Multi-pass X-ray laser oscillator (XLO) for improved spectrum and stability**
  - A. Halavanau, et. al., PNAS, 117, 15511, 2020
  - A. Halavanau, WE2C1

AMPLIFIED SPONTANEOUS EMISSION (ASE) OR SUPER-FLUORESCENCE

- **In optical regime, Maxwell-Bloch Equations (MBEs) describe amplification in optical medium**
  - M. Gross and S. Haroche, Phys. Report 93, 301 (1982)
- **With an ad-hoc spontaneous emission (SE) term added, the MBEs can describe ASE.**
- **However, ad-hoc SE models so far in use, exhibit wrong time-delayed peak (rather than exponential decay)**
- **Correct, quantum treatment of SE is critical for X-ray ASE due to ultrashort SE lifetime**
- **Analysis of 1D X-ray ASE based on QM was performed previously**
  - **Correlation functions and *factorization assumption***
  - ***Does not include the seed field***
  - A. Benediktovitch, V. P. Majety, and N. Rohringer, PRA99, 013839 (2019)
- **We have performed a 3D quantum analysis, and derived "modified MBEs"**
  - **SE is accurately represented by adding a noise term to atomic coherence**
  - **3D with paraxial approximation**
  - **Include seed field**
- **The theory provides an efficient framework for ASE and XLO simulation**
  - J.-W. Park, K.-J. Kim, R.R. Lindberg, ArXiv:2305.04653

X-RAY ASE: QUANTUM THEORY

- **2-level atoms coupled to EM field via dipole moment μ** (Scully & Zubary, Quantum Optics (CUP, 1997))
- **Ground and excited states, $|g\rangle^a$ and $|e\rangle^a$, with energy difference $\hbar\Omega$**
  - Atomic operators: $\sigma_+^a = |e\rangle^a |g\rangle^a$, $\sigma_-^a = |g\rangle^a |e\rangle^a$, $\sigma_z^a = |e\rangle^a |e\rangle^a - |g\rangle^a |g\rangle^a$
- **Heisenberg equation for the positive freq. part of EM field at $r_a = (x_a\ , z_a)$ at retarded time $\tau = t - \frac{z_a}{c}$)) after factoring out the fast oscillation $e^{i\Omega\tau}$**

$$\tilde{E}_+^{(a)}(\tau) = \tilde{E}_{+,in}^{(a)}(\tau) + \frac{3i\hbar\Gamma_{\rm sp}}{8\pi\mu} \sum_{z_b<z_a} \mathcal{G}(\mathbf{r}_a - \mathbf{r}_b)\tilde{\sigma}_-^{(b)}(\tau); \quad \mathcal{G}(\mathbf{r}) = \int_{-\infty}^{\infty} d^2\phi e^{i\frac{\Omega}{c}\left(\phi\cdot\mathbf{x} - \frac{\phi^2}{2}z\right)} = \frac{\lambda_\Omega}{iz} e^{i\frac{\pi \mathbf{x}^2}{\lambda_\Omega z}}$$

  - $\tilde{E}_{+,in}$: Free-propagating seed field
  - Spontaneous and stimulated emission associated with down transition σ_-^b
  - $\mathcal{G}(\boldsymbol{r})$:Paraxial Green's function, replaced by a constant effective angle Δo in 1D theory
  - Equations for atomic operators

$$\frac{d\tilde{\sigma}_-^{(a)}}{d\tau} = -\frac{\Gamma_{\rm sp}}{2}\tilde{\sigma}_-^{(a)}(\tau) - \frac{i\mu}{\hbar}\sigma_z^{(a)}(\tau)\tilde{E}_+^{(a)}(\tau),$$

$$\frac{d\sigma_z^{(a)}}{d\tau} = -2\Gamma_{\rm sp}\tilde{\sigma}_+^{(a)}(\tau)\tilde{\sigma}_-^{(a)}(\tau) + \frac{2i\mu}{\hbar}\left[\tilde{\sigma}_+^{(a)}(\tau)\tilde{E}_+^{(a)}(\tau) - \tilde{E}_-^{(a)}(\tau)\tilde{\sigma}_-^{(a)}(\tau)\right]$$

- **The damping term through the $a = b$ term excluded in E_+^a (Wigner-Weisskopt).**

MAXWELL-BLOCH EQUATIONS: CLASSICAL

- **Replace operators by their c-number averages using the density matrix**

$$\left\langle \tilde{\sigma}_-^{(a)} \right\rangle = \mathrm{Tr}\left(\tilde{\sigma}_-^{(a)} \rho^{(a)}\right) = \rho_{eg}^{(a)}; \left\langle \tilde{\sigma}_+^{(a)}(\tau)\tilde{\sigma}_-^{(a)}(\tau) \right\rangle = \rho_{ee}^{(a)}(\tau); \left\langle \sigma_z^{(a)}(\tau) \right\rangle = \rho_{ee}^{(a)}(\tau) - \rho_{gg}^{(a)}(\tau) \equiv \rho_{\mathrm{inv}}^{(a)}(\tau)$$

- $\mathcal{E}_+^a \equiv \left\langle E_+^a \right\rangle$

→ **Maxwell-Bloch Equations (MBEs)**

$$\mathcal{E}_+^{(a)}(\tau) \equiv \left\langle \tilde{E}_+^{(a)}(\tau) \right\rangle = \mathcal{E}_{+,in}^{(a)}(\tau) + \frac{3i\hbar\Gamma_{\mathrm{sp}}}{8\pi\mu} \sum_{z_b<z_a} \mathcal{G}(\mathbf{r}_a - \mathbf{r}_b)\rho_{eg}^{(b)}(\tau)$$

$$\frac{d\rho_{ge}^{(a)}}{d\tau} = -\frac{\Gamma^{(a)}}{2}\rho_{ge}^{(a)}(\tau) - \frac{i\mu}{\hbar}\rho_{\mathrm{inv}}^{(a)}(\tau)\mathcal{E}_+^{(a)}(\tau).$$

$$\frac{d\rho_{ee}^{(a)}}{d\tau} = r_e^{(a)}(\tau) - \Gamma_{ee}^{(a)}(\tau)\rho_{ee}^{(a)}(\tau) + \frac{i\mu}{\hbar}\left[\mathcal{E}_+^{(a)}(\tau)\rho_{ge}^{(a)}(\tau) - \mathcal{E}_-^{(a)}(\tau)\rho_{eg}^{(a)}(\tau)\right]$$

$$\frac{d\rho_{gg}^{(a)}}{d\tau} = r_g^{(a)}(\tau) + (\Gamma_{\mathrm{sp}} + \gamma_n)\,\rho_{ee}^{(a)}(\tau) - \gamma_g^{(a)}(\tau)\rho_{gg}^{(a)}(\tau) - \frac{i\mu}{\hbar}\left[\mathcal{E}_+^{(a)}(\tau)\rho_{ge}^{(a)}(\tau) - \mathcal{E}_-^{(a)}(\tau)\rho_{eg}^{(a)}(\tau)\right]$$

Incoherent rates

$$\Gamma^{(a)}(\tau) \equiv \Gamma_{ee}^{(a)}(\tau) + \gamma_g^{(a)}(\tau) + q^{(a)}(\tau), \quad \Gamma_{ee}^{(a)}(\tau) \equiv \Gamma_{\mathrm{sp}} + \gamma_e^{(a)}(\tau) + \gamma_n$$

q=decoherence, $\gamma's$=depletion, $r's$=pumping

- **MBEs are a closed set of equations describing gain and saturation of due to stimulated emission.** (Review by Gross and Haroche, Phys. Report 93, 301, 1982) However, MBEs do not include spontaneous emission→ ASE cannot start.

MODIFIED MBEs WITH SPONTANEOUS EMISSION

- **Field from atom $\boldsymbol{a}$ is proportional to $\boldsymbol{\sigma_{-}^{a}}$ →The photo-current intensity $\propto \boldsymbol{S^{a}(\tau)}$ $\equiv \langle \tilde{\boldsymbol{\sigma}}_{+}^{a}(\boldsymbol{\tau})\tilde{\boldsymbol{\sigma}}_{-}^{a}(\boldsymbol{\tau})\rangle = \boldsymbol{\rho_{ee}^{a}(\tau)} = \left|\boldsymbol{\rho_{ge}^{a}}\right|^{2} + \boldsymbol{\rho_{ee}^{a}(\tau)} - \left|\boldsymbol{\rho_{ge}^{a}}\right|^{2}$**
- **The term $\left|\boldsymbol{\rho_{ge}^{a}}\right|^{2}$ is the classical approximation**: $\langle\sigma_{+}^{a}(\tau)\rangle\langle\sigma_{-}^{a}(\tau)\rangle = \rho_{ge}^{a}\ \rho_{eg}^{a} = \left|\rho_{ge}^{a}\right|^{2}$
- **The correction term $\boldsymbol{\rho_{ee}^{a}(\tau)} - \left|\boldsymbol{\rho_{ge}^{a}}\right|^{2}$ is necessary to treat $\tilde{\boldsymbol{\sigma}}_{\pm}^{a}(\boldsymbol{\tau})$ as q-numbers. This term corresponds to the spontaneous emission**
- **The spontaneous emission can be recovered by modifying MBEs by introducing a randomly phased noise amplitude to the "*atomic coherence*" $\boldsymbol{\rho_{ge}^{a}}$**

$$\rho_{ge}^{a}(\tau) \rightarrow \hat{\rho}_{ge}^{a}(\tau) \equiv \rho_{ge}^{a}(\tau) + \xi^{a}(\tau)$$

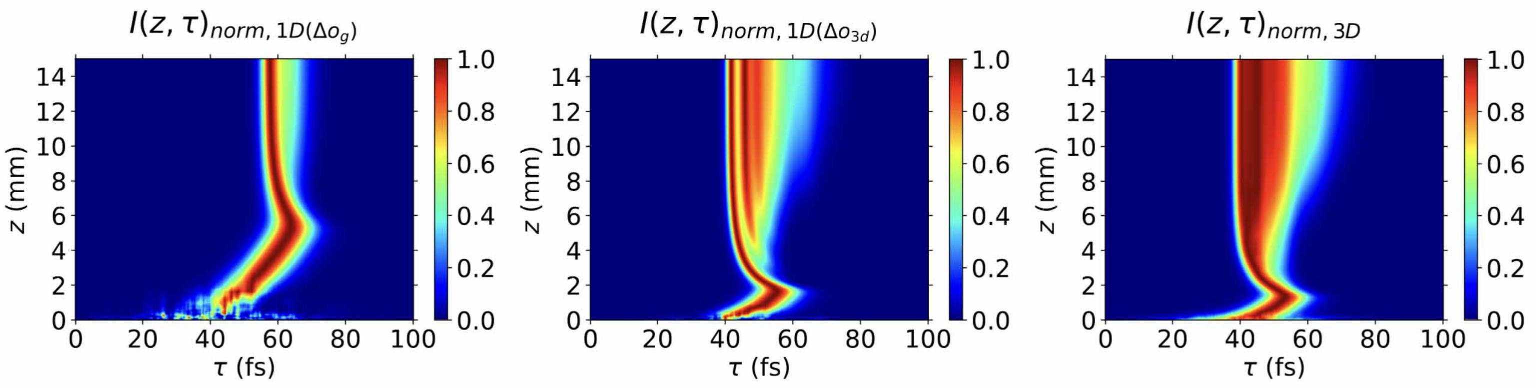
$$\langle\xi^{a}(\tau)\rangle_{en} = 0, \langle\xi^{a}(\tau_1)\xi^{*a}(\tau_2)\rangle_{en} = \left(\rho_{ee}^{a}(\tau_1) - \left|\rho_{ge}^{a}(\tau_1)\right|^{2}\right)\delta_{ab}\delta_{\tau_1\tau_2}$$

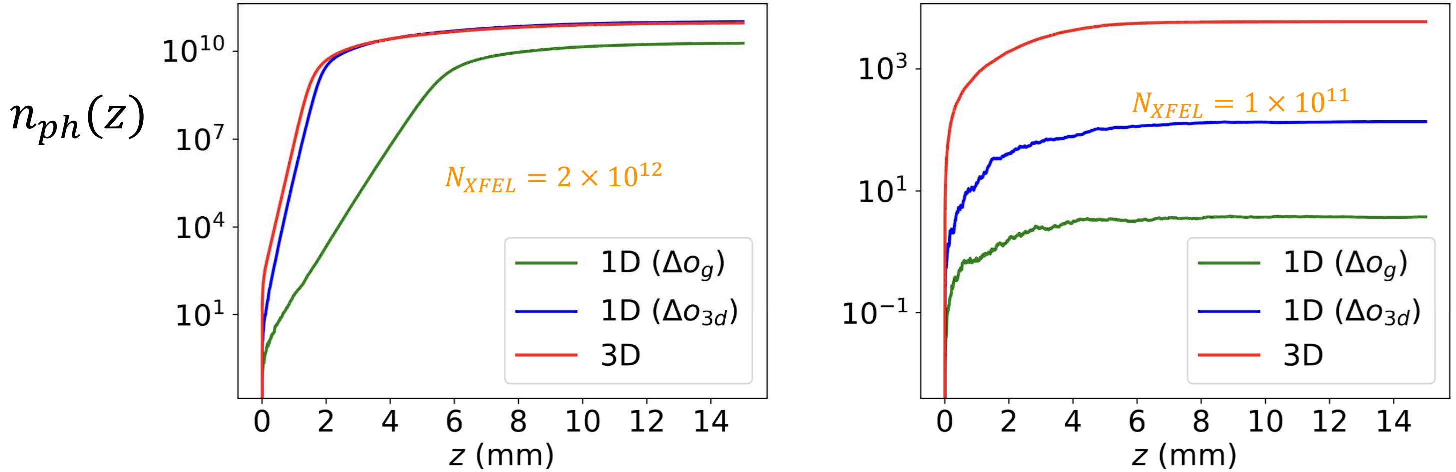
- **The spontaneous emission computed from modified MBEs agrees with quantum mechanical predictions:**
  - $S^{a}(\tau) \propto e^{-\Gamma_{sp}\tau}$ and correct photon number

- **Past attempts to add the SE noise term to the time-derivative of $\boldsymbol{d\rho_{ge}^{a}(\tau)/d\tau}$ →incorrect time-delayed peak, $\boldsymbol{\tau e^{-\Gamma_{sp}\tau}}$ as in Brownian motion**

ASE SIMULATION BASED ON THE MODIFIED MBE

$\lambda_\Omega = 1.46\, nm, \Gamma_{sp}^{-1} = 160$ fs, $\gamma_e^{-1} = 2.4\, fs, R = 2\mu m, L = 15mm$, volume density= 1.6×10^{25}, $r_g = \gamma_g = q = \gamma_n = 0$, photoionization cross section (pumping)= $0.3\, Mb$, 880 eV XFEL pulse (flattop transverse profile) $N_{XFEL} = 2 \times 10^{12}$ photons. For 1D: $\Delta o = \Delta o_g \equiv \pi R^2/L^2 = 0.56 \times 10^{-7}$ or $\Delta o = \Delta o_{3d} \equiv \lambda_\Omega^2/\pi R^2 = 1.70 \times 10^{-7}$

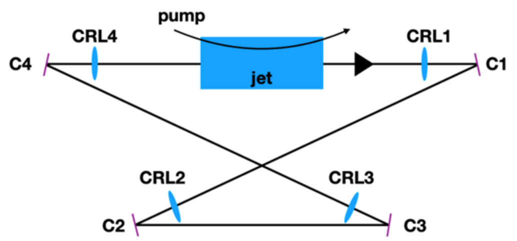




With 3D, more modes participate in ASE with larger start-up noise and steeper growth

ASE settles into a steady state

XLO SIMULATION

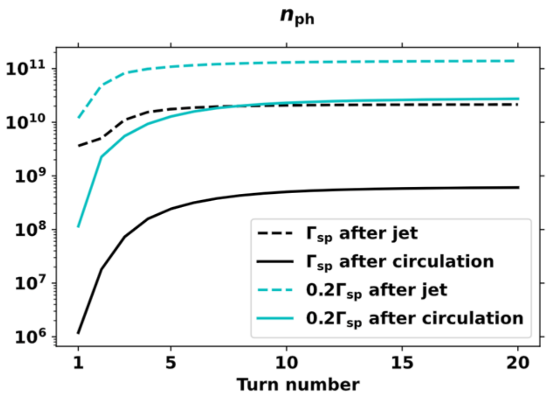


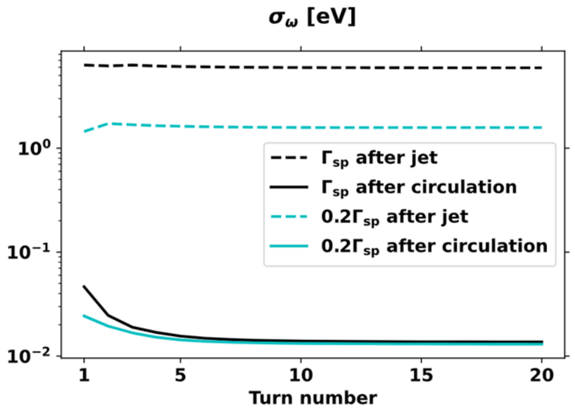
$L_{cav} = 1.6\text{m}$
$f = 0.0623\text{m}$
$\sigma_r = \sigma_{r_{pump}} = 0.3\mu m$

1mJ pump

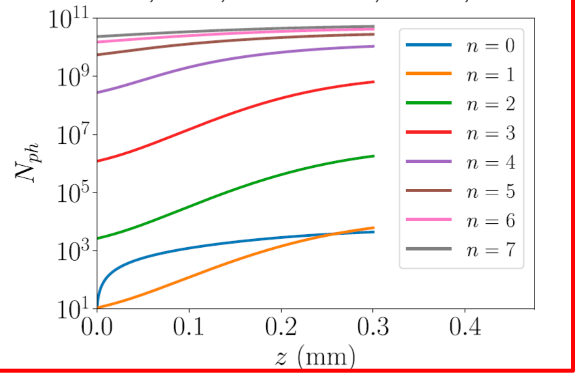
20fs, 1 mJ pump
$L_{jet} = 0.3\text{mm}$, $n_v = 4.2 \times 10^{27}/m^3$, $\sigma_{abs} = 0.324\text{Mb}$, $E_{\text{pump}} = 9\text{keV}$, $E_{\text{rad}} = 8\text{keV}$ (A. Halavanau et al, PNAS 117, 15511, 2020)
$\Gamma_{\text{sp}}^{-1} = 1.1\text{fs}$ (A. Benediktovitch et al, arXiv:2303.00853)
$\Gamma_e^{-1} = 0.693\text{fs}$, $\Gamma_g^{-1} = 1.18\text{fs}$ (M. O. Krause and J. H. Oliver, Phys. Chem. Ref. Data 8, 329)

Silicon 444's cut: $\Delta\omega = 48.1\text{meV}$, $\Delta\phi_x = 31.6\mu\text{rad}$





Halavanau, et al, PNAS 117,15511, 2020



- XLO performance suffers if the spontaneous emission rate is too large
  - ASE has a smaller BW when the SE lifetime is shorter
    - In the above example, $\hbar\Gamma_{sp}$~ 1 eV >> $\Delta\omega_{refl}$→ large loss due to crystal filtering
- The previous work assumed that the spontaneous emission was negligible after the first pass
  - See, however, WE2C1

SUMMARY AND CONCLUSIONS

- We have developed quantum theory of XFEL-pumped ASE including seed field and 3D effect in paraxial approximation
- Assuming factorization: $\left\langle \sigma_j^a(\tau)\sigma_k^b(\tau)\right\rangle = \langle\sigma_k^a(\tau)\rangle\left\langle\sigma_k^b(\tau)\right\rangle$,$a \neq b$, the theory can be cast in the form of a modified MBEs, in which a noise term is added to the atomic coherence
- An efficient simulation code was developed based on the modified MBEs.
  - The effective initial noise is higher in 3D than in1D
- Since seed field is included, a multi-pass XLO can be simulated
- The performance for hard X-ray XLO may be limited due to ultrafast spontaneous emission

- ***We thank Nina Rohringer, Andrei Benediktovitch, Linda Young, and Kai Li for discussion***