Table-top two-color soft X-ray laser from Ni-like Mo plasma

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ABSTRACT

Two-color laser pulses in a laboratory setup are interesting for enabling a number of advanced spectroscopy techniques. The generation by means of Ni-like laser-produced plasmas is promising for scaling down the wavelength towards the soft X-ray. The occurrence of a two-color signal has been documented in the 1990s and a detailed atomic physics study interpreted the pulse as due to concomitant collisional pumping (color-1) and self-photoexcitation (color-2). If this framework is on the atomic physics basis valid, its kinetic and hydrodynamic quantitative aspects need to be better understood. In particular, experimental observations leave room for a number of open points. Here we propose an amplified Raman scattering (ARS) scheme, to explain the growth of color-2 and its sensitivity to the plasma irradiation scheme.

Keywords: plasma, XUV, two-color, spectroscopy, collisional, photopumping, Raman

1. INTRODUCTION

Short-wavelength pulses with bimodal spectra ("two-color") are very insightful for spectroscopic applications, for instance to investigate the structure and dynamics of chemical processes^{1,2}. In particular, when two coherent wavelengths with a significant spectral separation are available, a number of chemical processes can be probed, by means of pump-probe techniques, differential absorption, stimulated scattering, and so on. Advanced spectroscopies are available at beamlines, and research is done to enable them in the home-lab. Unfortunately, advanced spectroscopies profiting from two-color pulses are still not very developed even at beamlines, due to the difficulty to generate such signals.

Just recently, SLAC researchers demonstrated pairs of X-ray laser pulses through self-amplified spontaneous emission (SASE) in a X-ray Free-Electron Laser, with wavelengths at ca. 0.81 nm (ca. 1530.7 eV) and ca. 0.83 nm (ca. 1493.8 eV), i.e. a minimal spectral separation. Also the SACLA beamline demonstrated two-color X-ray pulses at ca. 0.09 nm (ca. 13'776 eV) and ca. 0.12 nm (10'332 eV). Masoudnia et al.³ have summarized the three beamline architectures that are functional to generate two-color pulses, i.e. (i) *double split undulator* mode, (ii) *gain-modulated undulator* mode, and (iii) *double-bunch undulator* mode. Spectral bandwidth tolerances in both the source, whether the beam is seeded or not, and the detection, whether it is a high-res or a FT spectrometer, are crucial in making such effects characterizable. However promising the results, short-wavelength free-electron lasers are currently available in very few sites⁴. The spectroscopic potential demands that such special pulses are made available in home-lab tabletop sources, to be fully exploited 24/7.

Nilsen et al.⁵⁻¹⁰ predicted and experimentally demonstrated¹¹ that laser-produced micro-*plasmas* are able to produce two-color pulses by means of amplified spontaneous emission (ASE). In the collisional excitation scheme, leading to color-1, the upper laser level cannot radiatively relax back to the ground state and is indeed metastable¹². The lowest excited level in Ni-like ions¹³ is the $3d^9 4s\ ^3D_3$ that can decay to the ground state only by magnetic octupole (M3) transition. For Mo, its lifetime was reported to be 4'484ms. Electric-dipole forbidden transitions dominate a large region of the M-shell X-ray emission of Ni-like ions. Levels that decay by E1 transitions feature lifetimes in the ps time-scale, while the levels that decay by M1 or E2 transitions show lifetimes in the ns or μ s range. Träbert *et al*¹² also showed that $3d^9 4d$ cannot decay easily towards $3d^{10}$ levels, and their predominant decay branch leads to the $3d^9 4p$ level.

The observation of two laser colors has been widely interpreted as due to two different "population inversion" mechanisms, namely through

As a matter of fact, the upper laser level $|u_2\rangle$ can be populated by collisional excitation and photoexcitation at the same time. The inversion is presumably accomplished by destroying the lower laser state $|l_2\rangle$ by collisional mixing mainly⁵. Different authors agree that the thick plasma requires radiative pumping at ca. 350eV (3.53nm)^{3,7}. Such soft X-ray pump may be calculated as the *peak radiation* for a black-body emitter at a temperature of 70eV, using Wien's law. A 70eV plasma is not a difficult condition to attain and that is why the self-photopumped mechanism is a qualitatively plausible interpretation for sustaining the 4f population leading to color-2.

Long-range, i.e. beyond the $c\tau_{\text{pulse}}$ "horizon", optical thickness is required for the entire amplification time and space (fig. 2). Nilsen⁷ calculated strict spatio-temporal windows for the accomplishment of the self-photopumping, while Masoudnia et al.¹⁷ provided less strict ranges. It should be said that the laser input conditions are not exactly the same for the two references, and in this respect the two works provide comparable orders of magnitude for the temporal and spatial windows. Both agree that color-2 is characterized by a rather limited efficiency, which demands a more detailed explanation.

Fig. 2: Sketch of plasma structure and evolution supposed to host the collisional and the photo-pumped ASE (Plasma Zoning Model). The contour indicate density, temperature or gain. The drawing is stylized from literature results, such as Nilsen et al.⁵⁻¹⁰ and Masoudnia¹⁷.

Two major limitations to propagating the gain through the plasma are possibly refraction and transit-time⁷. Nilsen however excluded that refractive effects are important as the gradients are claimed to be small⁵, while a few other workers do indicate that this effect must be considered carefully¹⁷. Nilsen computed the gain for a number of Ni-like Mo lines (Tab. 1, in ref.⁷), with and without optical thickness considered in the simulation, showing that radiation self-trapping is essential for the gain of the self-photopumping $4f \rightarrow 3d$ line (88% reduction when the plasma is optically thin), and that it is also important for the collisional $4d \rightarrow 4p$ line (26% reduction when the plasma is optically thin). The latter suggests that (i) collisional-radiative mixing is important, and (ii) color-1 and color-2 are somehow interlinked, rather than contending pumping schemes.

Nilsen et al.⁸ have done a detailed gain length study of Mo plasma-laser with the traveling-wave excitation (TWE)¹⁸, and showed that without the traveling wave color-2 showed a weak gain. The use of the traveling-wave setup does increase the output of the color-2 in Mo by two orders of magnitude and is essential for observing strong lasing on this line. Furthermore, the $4f \rightarrow 4d$ line was never observed in a *non-lasing plasma*, i.e. without color-1. Its interpretation is thus relying on calculations using measured values of the $3d \rightarrow 4f$ photo-pumping resonance.

Kuba et al.¹⁹ investigated the performance of plasma-lasing under TWE and also a dual pulse scheme (70J long pulse for high pre-ionization, and 30J short pulse for inversion pumping) with fundamental (1ω) and second harmonic (2ω). Using TWE a 300-fold increase in intensity on color-1 and color-2 was observed. Refraction was pointed out to be an important

factor, with as high as 13mrad deflection angle over 1 cm plasma propagation. The lasing was observed from both endings of the plasma column. In order to prevent cold plasma termination, the line focus was longer than the target length. Lasing was observed on the color-1 $4d-4p$ line ($\lambda = 13.9$ nm with Ag target), but also on what has been computationally identified as color-2 $4f-4d$ ($\lambda = 16.0$ nm with Ag target). Insightful observations are reported that turn out to be crucial, namely:

- The intensity of the color-2 was correlated to the one of the color-1, in some cases with similar amplitude, but color-2 was very unstable from shot to shot, with very high sensitivity to the energy of the short pump pulse;
- The color-2 was observed only under TWE, and in particular when TWE was progressing towards the spectrometer (forward plasma ending), but not on the opposite side;
- The temporal delay between long and short pulse must be set to 250ps and the plasma lengths greater than 3.8, such that color-1 was saturated;
- The irradiation scheme must be such that 2ω for the first pulse and 1ω for the second pulse, and never with other combinations of 1ω and 2ω .

Recently, Siegrist et al.¹⁶ reported on two-color plasma-lasing experiments where the *self-photopumped* Neon-like $3d^1P_1-3p^1P_1$ and Nickel-like $4f^1P_1-4d^1P_1$ laser transitions were invoked. The self-photopumped x-ray laser was regarded as an *alternative* scheme to electron collisional excitation in the search for an extension of available XUV and soft X-ray laser wavelengths. This implies that color-1 and color-2 are alternatives, and not interlinked. Further, one has to point out that the occurrence of the so-called “self-photo-pumped” wavelength (color-2) is “harder” (higher photon energy) than the collisional one for the Ne-like radiators, but “softer” for Ni-like case. Moreover, the color-2 gain is typically lower than color-1 although the saturation intensity is expected to be higher¹⁶. The Bern group studied the parametric dependence of self-photopumping, e.g. varying the pre-pulse delay. The latter was indicated to scale-down, for optimum operation, as a function of atomic number of the target. Two-color lasing was shown to occur for a number of “uncharted elements” including Ne-like V, Cr, Fe, and Co as well as Ni-like Ru and Pd. Time-resolved streak measurements were presented in the same work, which show that color-2 is not occurring over the entire lifetime of color-1. On the contrary, color-2 occurs, with a minimal time-lag (ca. 2ps), after and during color-1, and only in parallel to the peak of it. This observation is very insightful, although the time resolution was at the limit of the used instrumentation.

If the mechanism of the two-color process is *qualitatively* interpreted as due to a collisional (color-1) and a photo-excited (color-2) pumping, some *quantitative* aspects on the realization of the latter process remain to be clarified. In ref.¹⁷ we have discussed in detail the contribution of collisional and photo-driven pumping for color-2. Still three main “open points” deserve to be better understood, namely:

- Self-photopumping “recycles” existing energy, and it will not lead to a growth of population in the upper laser level; Assuming self-absorption supports self-photopumping, how does the color-2 stimulated emission is seeded?
- Assuming the seed of photoexcitation comes from Bremsstrahlung, the fast plasma dynamics can deplete the required radiation within the buildup time; on the other hand, if the plasma is too hot, excessive photo-excitation can progress towards photoionization;
- The observation made in Kuba et al.¹⁹ (as summarized also above) indicate a dependence on plasma structure; What is the role of TWE? What is the “interlink” between color-1 and color-2, and why the optical thickness is crucial for *both* of them?

Aim of this paper is to contribute to the process of two-color plasma-lasing, and discuss in further detail a potential mechanism that would *quantitatively* explain the occurrence of color-2, even without the strict requirement for a population inversion.

2. MATERIALS & METHODS

2.1 Experimental

A planar molybdenum (Mo) target is line-irradiated by a $\lambda=1054$ nm Nd:glass chirped pulse amplification (CPA) laser system with a pulse duration of 1.5 ps and the plasma gain-medium is created. Two pre-pulses (0.5% and 16% of main

pulse energy) and a 2.5J main-pulse are delivered at an incident angle of 40° with respect to the target normal (Fig. 3). The pre-pulses arrived 4.8ns and 0.5ns before the arrival of the main pulse. It should be remarked that is the first observation of two-color Mo-plasma laser with a pump energy below 3J. This is promising in order to enable advanced spectroscopic techniques in the home laboratory.

A flat-field grazing incidence spectrometer is used to acquire spectra of the plasma-driven XUV laser (Fig. 3). The XUV laser emission was measured on-axis using a 1'200 grooves/mm, aberration-corrected, flat-field Hitachi grating. The spectrum was recorded on a phosphor screen (P43) imaged onto a cooled CCD camera having a pixel size of $23\mu\text{m}$. The spectrometer resolves the image with a spectral resolution of 10^{-3} thanks to an "entrance pinhole" to delimit the aperture of the spectrometer.

Fig. 3: Schematic of the experimental setup used for the generation and observation of the two-color Mo-plasma X-ray laser.

2.2 Computational

A suite of public codes was used and coupled through self-written routines to investigate amplified spontaneous emission (ASE) in Ni-like Mo¹⁴. Boundary conditions were chosen to reproduce the experimental conditions, as summarized in the section 2.1. The first code is performing 1D hydrodynamic calculations of the laser-plasma expansion. With that, one can compute the electron temperature and density, as well as the gain in the spatio-temporal dimension. Given the nature of the lasing process, of ASE along the plasma column, here a translational invariance along the propagation dimension is assumed. An atomic physics code (FAC) was used to obtain collisional and radiative rate coefficients. The exact calculation of the gain and the refractive propagation of the XUV laser was done with self-written codes. All codes are described in detail in ref.³.

3. RESULTS & DISCUSSION

3.1 Observation of two-color XUV lasing process with minimal pump energy

Fig. 4 summarizes experimental results obtained from a Mo target. Our computational results are summarized in Tab. I, along with experimental data for the case of a Mo target. Fig. 5 shows the collisional rates as a function of electron

temperature, along with the *inversion temperature* (see arrows), i.e. the temperature at which excitation and relaxation rates are equal for a given transition. Above the inversion temperature, excitation dominates, which helps to grow population at the upper level. For some transitions, one cannot find any inversion temperature because the relaxation is always larger than the excitation, and such transitions do not experience any population development.

Fig. 4: Experimental data showing (a) the spectrograph image, (b) the image projection along the wavelength axis, and (c) the angle resolved image as centered on the color-1 peak.

Tab. I: Main parameter values as obtained from calculation or experiment. See text for discussion.

Denomination (Transition)	4d-4p (color-1)	4f-4p (color-2)
Far field beam centroid [mrad]	0.0	Mo: 3.2
Pointing stability [mrad]	Mo: 1.4;	Mo: 1.4;
Beam refractive divergence [mrad]	Calc. 6.2 (exper. Mo: 10.2)	Calc. 9.8 (exper. Mo: 8.3)
Electron density at max gain [cm^{-3}]	$1.2 \cdot 10^{20}$	$2.1 \cdot 10^{20}$
Electron temperat. at max gain [eV]	295	350
Inversion electron temperature [eV]	270	320
Elevation of gain peak [μm]	76	70
Rel. time of gain peak [ps]	0	0.2
Gain peak [cm^{-1}]	230	55
Gain lifetime [ps]	5.7	1.1
Gain thickness [μm]	32.8	18.2
Gain buildup time [ps]	0.7	0.9
Gain buildup length [μm]	11.5	6.3
Intensity [cts]	Mo: 600	Mo: 35
Rel. bandwidth $\Delta\lambda/\lambda \cdot 10^{-3}$	Mo: 9	Mo: 6

Fig. 5: Collisional rates for a selection of six transitions in the Ni-like Mo. The arrows indicate so-called “inversion temperatures”. Above such temperatures the upward transitions exceed the relaxation rates, which contributes to the upper population growth. The level 2 is 2.1 (see introduction).

The inversion values indicate that the upper level are populated from the ground level $|g\rangle$ for typical temperatures of a few hundred eV, while population from non-ground level require much higher temperatures above the keV. The population of the $|l_1\rangle$ level at 230eV (clearly for the present case of a Mo target) can degrade the lasing process, because (i) populates the low laser level thus reducing the stimulated emission rate, and (ii) subtracts electrons from the feed to the upper laser level. This temperature is thus to be intended as a bottom limit for the lasing process to be sustained.

For color-1's $|u_1\rangle$ state, the rate is low ($3 \cdot 10^{-12} \text{ cm}^3/\text{s}$), while the inversion dominates above 270eV for the considered case of a Mo target. Considering the thermal profile induced by the main pump of the laser-plasma, a low inversion temperature indicates a more rapid onset of inversion, than the “contending” color-2 process. For the latter, i.e. up to $|u_2\rangle$, the inversion temperature is at 320eV, i.e. 50eV higher than the “contending” color-1 process.

The measured Mo color-1 intensity was approximately 18-fold stronger than color-2, despite a 2.5 order of magnitude weaker collisional rate, which suggests that the collisional gain of the color-2 is more drastically limited in duration, spatial distribution, and Stokes efficiency. The idea that the color-2 is largely sustained by radiative pumping, whose maximum rate was calculated to be $1.5 \cdot 10^{-13} \text{ s}^{-1}$, requires optical thickness (self-absorption) and rapid pumping, since, as said above, the $|u_2\rangle$ state is not metastable (actual lifetime $\tau = 67\text{fs}$).

It is noteworthy to mention that our hydrodynamic calculation shows that in the lasing region, the population in the ground level is approximately 3.4 times higher than the population in the $|u_2\rangle$ level. However, there is a gain because the population of the $|u_2\rangle$ level is greater than the population of the depleted $|l_2\rangle$ level.

Fig. 6 summarizes the calculated temporal and spatial distribution of the lasing gain for the two colors from a Mo target. Temporally we determined a “gain *time-voxel*” (*integral of gain temporal profiles*) of ca. $22 \cdot 10^3 \text{ cm}^{-1} \text{ ps}$ (color-1) and ca. $250 \text{ cm}^{-1} \text{ ps}$ (color-2), which makes a ratio of ca. 88. Spatially we determined a “gain *space-voxel*” of ca. $6 \cdot 10^6 \text{ cm}^{-1} \mu\text{m}$ (color-1) and ca. $1 \cdot 10^6 \text{ cm}^{-1} \mu\text{m}$ (color-2), which makes a ratio of ca. 6.6. This indicates that the purely collisional color-1 process benefits of a 580-fold wider “spatio-temporal window”, which largely overrules the 2.5 orders of magnitude weaker collisional rate (Fig. 5).

Fig. 6: Calculated ASE gain as a function of (a) time and (b) height above the target, for the color-1 (monopole) and color-2 (dipole).

In Masoudnia et al.¹⁷, we showed a substantial effect of refraction and attributed the different color-1 and color-2 divergences to the fact that color-2 was produced deeper inside the thicker plasma, thus experiencing 20% more refraction. Scattering effects and walk-off are considered in sect. 3.2. The maximum of gain, for both colors, was calculated to occur shortly after the turning point, i.e. the deepest penetration of the pump pulse, where the density exceeds 10^{20} cm⁻³.

3.2 Potential Mechanism for Color-2 Lasing, compatible with the open points

As discussed so far, the color-2, observed in a number of specific circumstances, is *atomically* interpreted as due to 4f self-photoexcitation, while a more *plasma-wise* understanding is necessary. Here it is presented a complementary explanation, which fulfills all open points, but that is still subject of detailed experimental validation.

Fig. 7 was prepared with the Mo-XV (Ni-like) NIST data base, as compiled by Sugar *et al.*¹⁴ from a number of references. The Mo-XV (Ni-like) stage is generated with 544eV ionization energy, and is bounded by the Cu-like stage with ionization energy of 302eV, and the Co-like limit with 570eV, as shown in Fig. 1. Various authors compiled a consistent data base of lines, while pointing out that there is a “*severe blending*” of the 4p-4d transition, which made only the ³G₅ level of 3d⁹4d clearly attributable. In Fig. 7 we manually added (gray lines) the known XUV laser 4d levels, even though not reported in the NIST Database.

The strong 4d-4p color-1 emission is propagating in a high-density thick plasma-medium, where concomitantly the 3d-4f transition is being self-absorbed, supporting the 4f population. In the optically thick plasma, the color-1 may interact with the 4p population and scatter. Elastic self-resonant scattering of the *highly monochromatic* color-1 would maintain the population inversion of the color-1 (Rabi flopping scheme), and is a trivial case. *Inelastic* scattering, however, over a quasi-resonant “virtual state”, may create Stokes radiation that matches with color-2. The inelastic loss is highlighted in Fig. 7 at the 3d⁹ 4p ensemble, which is approx. 10eV. The Stokes branch may then be very close to the energy of color-2, which would support a *stimulated Raman scattering* process. This fast second-order process would happen in the very thick plasma-region, and would support color-2 amplification even *without the requirement of population inversion*, as typical for Raman amplifiers²⁰. The Raman process given in jj-coupling is $(5/2, 1/2)_2 \rightarrow \text{virtual state} \rightarrow (1/2, 1/2)_2$, which with $\Delta J=0$ indicates a transfer of energy from the j_1 core to the j_2 electron. The *selection rules* for electronic Raman effect are discussed in Kiel et al²². Even in cases where the electric dipole transitions are strongly forbidden (weak oscillator strengths), the electronic Raman effect is quite competitive with absorption processes. It should be reminded

that the Raman process scales with $1/\lambda^4$ and thus in the XUV domain Raman is surely quantitatively significant, especially since the ASE linewidth is very narrow²³.

Such process would require a strong color-1 “pump” and the concomitant photo-pumping in the same plasma domain, as in TWE, to ensure (i) hot and dense conditions over a length-scale *larger* than the light propagation “horizon” in the given pulse lifetime ($\sim$ ps); (ii) to drive the Amplified Raman Scattering (ARS) along with color-1’s ASE.

The characteristic “horizon” length-scale can be determined in TWE as $\Delta x \sim c_p \tau / \cos \theta$ where c_p is the speed of light in the plasma medium, τ is the pulse duration, and θ is the grazing angle. For 1.5ps one can obtain a few hundred microns, which is in agreement with the prediction summarized in Fig. 2 and Fig. 6.

Fig. 7: Compilation of NIST Database levels for Mo-XV above the ground Ni-like level. Tentatively a Raman-like process is sketched, and with the inelastic loss close to 10eV, one can predict Amplified Raman Scattering (ARS) to support amplification of color-2, which does not require population inversion. ARS requires color-1 saturation, and links color-2 to color-1.

If a highly monochromatic²¹ pump field at frequency $\omega_{\text{col},1}$ interacts with a scattering medium (thick plasma), the *spontaneous* Raman scattering of photons from metastable levels, i.e. without considering coupling with color-2, can be understood as an entropic decay of highly coherent states, within energy and momentum conservation. The former implies a “conversion of color”, the latter implies a walk-off of color-1 and color-2, i.e. two beam have different pointing of the centroid as observed here (as observed in this work, Tab. I). Detailed experimental investigations are however necessary, and are the subject of our ongoing work.

4. CONCLUSIONS

Ni-like plasmas have proven to scale down the laser wavelength of laboratory system to the extreme ultraviolet, at the limit of the soft X-rays. Further, they have demonstrated to generate two-color pulses, which are very interesting for

spectroscopic application. The interpretation of the second color was addressed in the 1990s, considering a self-photo-pumping mechanism. The atomic physics of such framework is supportive of such interpretation, but kinetics, population inversion, and hydrodynamic considerations are only qualitatively addressed. In this work a complementary model is presented, which does not conflict with previous contributions on self-photoexcitation, and extends their reach in more quantitative aspects. In brief, the occurrence of Raman scattering in the thick high-density plasma, may generate a Stokes signal that is degenerate with the weak color-2. Hence, Amplified Raman Scattering (ARS) may support the growth of color-2, especially when color-1 is saturated, even without 4f-4d population inversion. Color-1 and color-2 are thus interlinked, and propagate with a walk-off, to fulfill momentum conservation. The hydrodynamic structure of the plasma as related to the pumping scheme, e.g. TWE, forward or backward TCE, etc. turns crucial to determine the efficiency of the ARS process. The initial “noise” of color-2 thus generated, could also seed the self-photopumped process in a feedback loop.

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