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Abstract

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I. SECTION1

Recent news that X-ray free electron laser at SLAC (Stanford) became operational at 0.15 nm provided tremendous excitement for scientists in various disciplines. Very large efforts made by many researchers, engineers and technicians over a dozen of years at a very substantial cost is finally paying off. The very large cost of such a large "scientific tool" did not diminish the importance of the achievement, which is expecting to have a profound impact on the studies in many fields such as crystallography, and condense matter, in general, and high resolution microscopy of biological elements, etc. in particular. However, very large cost and size, resulting in low flexibility regarding the use of this important device has forced researchers to search for a more portable, less expensive XUV and X-ray devices, which can be used in individual researchers laboratories for preparatory work before going into these larger facilities.

In particular, a portable X-ray laser operating in a transient regime at 13.5 nm has been demonstrated in Suckewer's group [1] (see Fig. **??**). The laser uses H-like Li ions as an active medium which are excited by ionization-recombination process in a microcapillary. The excitation mechanism is the following. First, a strong 250 fs laser pulse (power density 10^{17}W/cm^2) creates nonequilibrium plasma of Li^{3+} ions and electrons. Electron density is $10^{19} - 10^{20}$ cm^{-3}. The ion-electron recombination occurs on a time scale faster then 1 ps which creates Li^{2+} (H-like) ions in highly excited states (large principle quantum number n). By collisions atoms are transferred to states with smaller n on a few ps time scale. For proper density population inversion between the level $n = 2$ and the ground state ($n = 1$) is created for $10 - 100$ ps. This results in lasing on the wavelength of the $n = 2$ to $n = 1$ transition ($13.5nm$) which is measured by a spectrometer. Experimental setup and the measured spectrum is shown in the figure **??**.

As demonstrated in the figure **??**, a low power 2 Hz Nd/YAG laser (100 mJ, $5ns$) was focused with an $f/6$ lens on the entrance of microcapillary. Microcapillaries of lengths from 1 to 5 mm and diameters of 0.3 mm were made in solid LiF by drilling appropriate holes. After a few hundred nsec delay (varied between 300 and 1000 ns), the Ti:sapphire 250 fs laser was fired, whose energy is $50 - 60$ mJ in a repetition rate 2 Hz. This femto-second laser was tightly focused with the same $f/6$ lens onto the plasma at the entrance of the microcapillary providing a power density close to $2 \times 10^{17}\ W/cm^2$. The Nd/YAG laser beam

was directed to the microcapillary by a near 100reflectivity mirror, M_1, and the femto-second laser beam was directed by directed by a mirror, M_2 which is transparent for the 1.06 μm wavelength of the Nd/YAG laser. From the output spectra, we could see the soft X-ray lasing.

In the present research we are trying to make a laser in a similar way, however, instead of ions, we want to use neutral He atoms which are easier to confine then plasma. Fig. **??** shows He energy level diagram and radiative decay times for various transitions. We want to make a laser operating at the 2 ^{1}P $\rightarrow$ 1 ^{1}S transition (58 nm). So, we need to create population inversion between those levels. We are planning to do it in two stages. First, we populate the triplet 2 ^{3}S state by ionization-recombination processes similar to what has been done in the Li-ions experiment. Then we transfer population into the singlet 2 ^{1}P state of He by sending short optical laser pulses which yields population transfer via the triplet 3 ^{3}D state.

Next I discuss these processes in details.

II. PREPARATION OF ORTHOHELIUM

First we inject an ultrashort high power laser pulse to ionize the He gas. Then we turned off the laser and rapid recombination and de-excitation follow such that the lowest states of He atoms are prepared according to their statistical weights. Hence for the sake of simplicity we take the relative population of the triplet 2 ^{3}S and singlet 1 ^{1}S states to be 3 to 1.

Let me consider the physics of the laser plasma as produced in our Princeton lab. We envision a laser plasma created by Keldysh tunneling with a non Boltzmann distribution of neutral excited atoms. This involves $He^{++} \rightarrow He^{+} \rightarrow$ He electron capture via three-body recombination. Three-body recombination for H-like ions is approximately proportional to the forth power of the principal quantum number n^4 and to the square of the electron density as ${N_e}^2$. Hence for sufficiently high initial electron density three-body recombination will dominate radiative decay.

However, the collisional ionization from highly excited states is also fast, thus in order for three-body recombination rates to dominate ionization rates, the recombining plasma should have a low electron temperature T_e. For example, for $k_B T_e \approx 1\ eV$, initial electron density $N_e \approx 2 \times 10^{16}$ cm^{-3} $\approx N_{He^{++}}$ for fully ionized He^{++} in quasi-neutral plasma, the

equilibrium between recombination and ionization processes will be established after a few dozen picoseconds at a ratio of $N_e/N_{\rm He} \approx 3 \times 10^{-3}$ for $N_{\rm He} = 10^{16}$ cm^{-3}. This ratio becomes even smaller ($N_e/N_{\rm He} \approx 10^{-8}$) for $k_B {\rm T}_e \approx 0.5\ eV$. In such way one can obtain an essentially neutral gas of excited He atoms.

In order to create a fully ionized He^{++} plasma at low temperature, we consider the example of a plasma capillary 10-100 μm in diameter and a few cm long. The tunneling ionization can be used to generate the plasma [3–6]. In this way one can strip both electrons from He atoms without significantly heating the plasma, especially for ultra-short laser pulses. The laser intensity needs to be in the order of 10^{16} W/cm^2 for efficient tunneling ionization of He to He^+ and to He^{++} according to Keldysh theory [3]. For needle like plasma column such intensities can easily be obtained from a Ti/Sapphire laser at wavelength $\lambda = 0.8\ \mu$m with ~ 10 mJ energy per pulse in pulses of $50 - 100$ fs duration with ionization pulse propagating in plasma channel. Use of such short pulses is crucial to minimize plasma heating. It is important to use laser pulses shorter than collision times of electrons in order to minimize plasma heating during the ionization process.

The bottom line is that we can create a cold laser plasma which recombines to produce an excited neutral gas. In particular the metastable triplet 2 ^{3}S (8000 s radiative life time) state will be formed with a statistical weight of around 3 compared to the 1^1S state. The population in the 2 ^{3}S state is then transferred to the 3 ^{3}D state via the 2 ^{3}P levels by dark state adiabatic transfer (STIRAP).

III. POPULATION TRANSFER FROM 2 ^{3}S TO 3 ^{3}D VIA STIRAP PROCESS

Robust population transfer from the triplet 2 ^{3}S to triplet 3 ^{3}D is then made possible by Stimulated Raman Adiabatic Passage (STIRAP) [7]. In this technique one subjects the system, whose state is 2 ^{3}S at $t = 0$, to a so called counter intuitive pulse sequence with Rabi frequencies Ω_1 and Ω_2 in which the Ω_1 (2 ^{3}P $\rightarrow$ 3 ^{3}D) pulse precedes the Ω_2 (2 ^{3}S $\rightarrow$ 2 ^{3}P) pulse. This pulse sequence ideally results in a complete transfer of population to the desired state 3 ^{3}D without necessarily populating the 2 ^{3}P state in the process.

The mechanism of STIRAP is best understood in the dressed state basis in which we

introduce bright and dark states. Beginning with the dark state

$$|0\ \rangle = \frac{\Omega_1|2\ ^3\mathrm{S}\rangle - \Omega_2|3\ ^3\mathrm{D}\rangle}{\sqrt{\Omega_1^2+\Omega_2^2}} \tag{1}$$

we apply Ω_1 before Ω_2 so that $|0\rangle \cong |2^3\mathrm{S}\rangle$ during the early stages of transfer. Then we adiabatically turn on Ω_2 while turning off Ω_1, such that $|0\rangle \cong |3\ ^3\mathrm{D}\rangle$ for large times. The condition of adiabaticity implies the following estimate of the required pulse energy (see Appendix A)

$$W \gtrsim 1000\frac{\hbar c S}{\lambda^3 \gamma \tau_{\mathrm{pulse}}}, \tag{2}$$

where S is the cross section area of the pulse, τ_{pulse} is the pulse duration, λ and γ are the wavelength and the rate of the transition. For the weakest $2^3\mathrm{P}\rightarrow 2^3\mathrm{S}$ transition $\lambda = 1083$ nm and $\gamma = 10^7\mathrm{s}^{-1}$. Then for a plasma capillary of radius ~ 0.1mm and pulse duration $\tau_{\mathrm{pulse}} = 1$ ps Eq. (2) yields $W \gtrsim 20\ \mu$J. Currently pico-second lasers are commercially available with much greater energy.

IV. POPULATION TRANSFER FROM 3 ^{3}D TO 2 ^{1}P: LEVEL DEGENERACY PROBLEM

Once the population is transferred to the 3 ^{3}D state a strong resonant driving field is applied on the 3 ^{3}D to 2 ^{1}P transition. However, levels 3 ^{3}D and 3 ^{1}D are essentially degenerate (splitting between them is 0.2 nm) and the applied external field inevitably drives transition 3 ^{1}D to 2 ^{1}P which is more then 1000 times stronger then 3 ^{3}D to 2 ^{1}P transition (see Fig. **??**). Thus, the Rabi frequency which drives the 3 ^{1}D to 2 ^{1}P transition is much larger then those driving the 3 ^{3}D to 2 ^{1}P transition. Under such condition, practically no population can be transferred from the 3 ^{3}D state as shown in Fig. **??**. This is true no matter how strong the driving field is.

To avoid this problem it has been suggested to apply an additional resonant field which drives the 3 ^{1}D to 4 ^{1}P transition. The equivalent scheme is sketched in the insert of Fig. **??**. If the additional driving field $\Omega_{bc}(t)$ is strong enough the population can be transferred completely from 3 ^{3}D to 2 ^{1}P (see Fig. **??**). The condition for an efficient population transfer is that

the amplitude A of the additional field Rabi frequency $\Omega_{bc}(t)$ is greater then those of $\Omega_{cd}(t)$. This is demonstrated in Fig. **??** which shows population transfer as a function of A.

Result shown in the figures are obtained by numerical solution of the evolution equations for C_a, C_b, C_c and C_d which are probability amplitudes to find the system in the states a, b, c and d respectively. For resonant driving fields the evolution equations read

$$\frac{dC_a}{dt} = i\Omega_{ad}(t)C_d, \tag{3}$$

$$\frac{dC_d}{dt} = i\Omega_{ad}(t)C_a + i\Omega_{cd}(t)C_c, \tag{4}$$

$$\frac{dC_c}{dt} = i\Omega_{ad}(t)C_d + i\Omega_{bc}(t)C_b, \tag{5}$$

$$\frac{dC_b}{dt} = i\Omega_{bc}(t)C_c, \tag{6}$$

with the initial condition $C_a(0) = 1$, $C_b(0) = C_c(0) = C_d(0) = 0$. Rubi frequencies $\Omega(t)$ are given in the figures in dimensionless units so that unit of time is the inverse amplitude of $\Omega_{ad}(t)$.

Derivation of these equations is provided in Appendix B for the special case of a two level system.

V. DISCUSSION

There are several methods for producing extreme ultra-violet lasing: for example, using a capillary discharge [8], a free-electron laser [9], optical field ionization of a gas cell [10] or plasma-based recombination lasers [11]. Coherent XUV radiation can also be produced by the generation of harmonics of an optical laser in a gas or plasma medium. Our main goal is to investigate the extent to which coherence effects might be useful in this problem.

Electron excitation has been the mechanism of choice for the pumping of a wide variety of XUV lasers. Alternatively, high-intensity ultrashort (with pulse duration less then 100 fs) optical pulses can be used to pump recombination lasers [4]. In this method, intense circularly polarized light ionizes atoms via tunneling process. Then atoms recombine yielding species in excited electron states.

The three-body recombination scheme is attractive due to its potential of achieving lasing at XUV wavelengths with relatively moderate pumping requirements. Several experiments have demonstrated gain and lasing in such scheme [1, 12, 13]. Recombination mechanism

relies on obtaining ions in a relatively cold plasma which is possible due to short duration of the pump pulse. Then rapid recombination and de-excitation processes follow during which transient population inversion can be created.

In the present work we are focusing on lasing in He and He-like ions which utilizes advantages of the recombination XUV lasers and possibly the effects of quantum coherence. The later, for example, is the key for lasing without inversion wherein quantum coherence created in the medium by means of strong driving field helps to partially eliminate resonant absorption on the transition of interest and to achieve gain without population inversion. Such an effect holds promise for obtaining short wavelength lasers in the UV and $X-$ray spectral domains where inverted medium is difficult to prepare due to fast spontaneous decay.

APPENDIX A: CONDITION OF OPTIMUM STIRAP

STIRAP is optimum if during the time of overlap of the two pulses ($\Omega_1(t)$ and $\Omega_2(t)$) the effective Rabi frequency

$$\Omega_{\text{eff}}(t) = \sqrt{\Omega_1^2(t) + \Omega_2^2(t)} \tag{A1}$$

is constant [7]. This implies that population transfer is efficient if the peak values of Ω_1 and Ω_2 are equal.

Because transition $2^3\text{P}\rightarrow 2^3\text{S}$ is about 6 times weaker then transition $3^3\text{D}\rightarrow 2^3\text{P}$, the $2^3\text{P}\rightarrow 2^3\text{S}$ transition determines the required energy per pulse. One can estimate the energy from the condition

$$\Omega\tau = 2\pi, \tag{A2}$$

where τ is the pulse duration,

$$\Omega = \frac{\wp E_0}{\hbar} \tag{A3}$$

is the Rabi frequency for the $2^3\text{P}\rightarrow 2^3\text{S}$ transition, E_0 is the amplitude of the electric field and $\wp$ is the electric-dipole transition matrix element.

Condition (A2) is essentially requirement of adiabaticity ($\Omega\tau \gg 1$). Eqs. (A2) and (A3) yield

$$E_0 = \frac{2\pi\hbar}{\wp\tau}. \tag{A4}$$

Energy density in electromagnetic wave is

$$u = \frac{1}{2}\epsilon_0 E_0^2, \tag{A5}$$

so that the total energy in pulse reads

$$W = uc\tau S = \frac{1}{2}\epsilon_0 E_0^2 c\tau S = 2\pi^2 \epsilon_0 cS\frac{\hbar^2}{\wp^2\tau}, \tag{A6}$$

where S is the cross section area of the pulse. The $2^3\mathrm{P}\rightarrow 2^3\mathrm{S}$ transition rate γ can be estimated as

$$\gamma = \frac{k_0^3\wp^2}{2\pi\epsilon_0\hbar} \quad \text{which yields} \quad \wp^2 = \frac{2\pi\epsilon_0\hbar\gamma}{k_0^3}. \tag{A7}$$

Substitute Eq. (A7) into Eq. (A6) gives finally

$$W = 8\pi^4\frac{\hbar cS}{\gamma\tau\lambda^3}, \tag{A8}$$

where λ is the wavelength of the transition.

APPENDIX B: DERIVATION OF EVOLUTION EQUATION FOR A TWO-LEVEL SYSTEM

Here we consider a two level atom (a is excited and b is the ground state). States a and b are described by wavefunctions ψ_a and ψ_b. We assume that atom interacts with an applied electromagnetic field which is polarized along the $x-$ axis

$$\vec{E}(t) = \hat{x}E_0\cos(\nu t). \tag{B1}$$

Hamiltonian of the system reads

$$\hat{H} = \hat{H}_0 + \hat{V}, \tag{B2}$$

where $\hat{H}_0$ is the Hamiltonian of the free atom

$$\hat{H}_0\psi_a = \hbar\omega_a\psi_a, \qquad \hat{H}_0\psi_b = \hbar\omega_b\psi_b \tag{B3}$$

and $\hat{V}$ corresponds to the atom-field interaction in the dipole approximation

$$\hat{V} = -e\vec{E}(t)\cdot\vec{r} = -exE_0\cos(\nu t), \tag{B4}$$

$\vec{r}$ is the electron position in the atom and e is the electron charge.

Matrix elements are given by

$$< \psi_a|\hat{H}|\psi_a >=< \psi_a|\hat{H}_0|\psi_a > + < \psi_a|\hat{V}|\psi_a >= \hbar\omega_a, \tag{B5}$$

$$< \psi_b|\hat{H}|\psi_b >=< \psi_b|\hat{H}_0|\psi_b > + < \psi_b|\hat{V}|\psi_b >= \hbar\omega_b, \tag{B6}$$

$$< \psi_a|\hat{H}|\psi_b >=< \psi_a|\hat{H}_0|\psi_b > + < \psi_a|\hat{V}|\psi_b >=< \psi_a|\hat{V}|\psi_b >= -eE_0\cos(\nu t)\int d^3r\psi_a^* x\psi_b. \tag{B7}$$

We introduce the following notation for the dipole matrix element between levels a and b

$$\wp = e\int d^3r\psi_a^* x\psi_b \tag{B8}$$

so that

$$< \psi_a|\hat{H}|\psi_b >= -\wp E_0\cos(\nu t). \tag{B9}$$

Under the applied electromagnetic field the atomic wavefunction ψ evolves so that ψ is a superposition of ψ_a and ψ_b

$$\psi = C_a(t)\psi_a + C_b(t)\psi_b \tag{B10}$$

with time-dependent coefficients $C_a(t)$ and $C_b(t)$ which we need to find. $C_a(t)$ and $C_b(t)$ have meaning of probability amplitudes to find atom in the state a and b respectively.

Schrödinger equation for the atomic wavefunction ψ reads

$$i\hbar\frac{\partial\psi}{\partial t} = \hat{H}\psi. \tag{B11}$$

Substituting Eq. (B10) into Eq. (B11) we obtain

$$i\hbar\dot{C}_a(t)\psi_a + i\hbar\dot{C}_b(t)\psi_b = C_a(t)\hat{H}\psi_a + C_b(t)\hat{H}\psi_b. \tag{B12}$$

Multiplying both sides of Eq. (B12) by ψ_a^* and integration over the volume we find

$$i\hbar\dot{C}_a(t) = C_a(t) < \psi_a|\hat{H}|\psi_a > + C_b(t) < \psi_a|\hat{H}|\psi_b > \tag{B13}$$

or

$$i\hbar\dot{C}_a(t) = \hbar\omega_a C_a(t) - \wp E_0\cos(\nu t)C_b(t). \tag{B14}$$

Multiplication of Eq. (B12) by ψ_b^* and integration over volume yields

$$i\hbar\dot{C}_b(t) = \hbar\omega_b C_b(t) - \wp E_0\cos(\nu t)C_a(t). \tag{B15}$$

Dividing both sides by $i\hbar$ and incorporating decay rates γ_a and γ_b of the states a and b due to spontaneous emission and collisions Eqs. (B14) and (B15) reduce to

$$\dot{C}_a = -i\omega_a C_a + i\Omega\cos(\nu t)C_b - \frac{\gamma_a}{2}C_a, \tag{B16}$$

$$\dot{C}_b = -i\omega_b C_b + i\Omega\cos(\nu t)C_a - \frac{\gamma_b}{2}C_b, \tag{B17}$$

where

$$\Omega = \frac{\wp E_0}{\hbar} \sim 10^8 \div 10^9 \text{ Hz}.$$

Eqs. (B16) and (B17) determine $C_a(t)$ and $C_b(t)$ and, thus, give a complete description of the system evolution. Introducing new functions

$$C_a = \exp\left(-\left[i\omega_a + \frac{\gamma_a}{2}\right]t\right)\tilde{C}_a, \tag{B18}$$

$$C_b = \exp\left(-\left[i\omega_b + \frac{\gamma_b}{2}\right]t\right)\tilde{C}_b, \tag{B19}$$

Eqs. (B16) and (B17) reduce to

$$\frac{d\tilde{C}_a}{dt} = i\Omega\cos(\nu t)\exp\left(\left[i\omega + \frac{\gamma_a - \gamma_b}{2}\right]t\right)\tilde{C}_b, \tag{B20}$$

$$\frac{d\tilde{C}_b}{dt} = i\Omega\cos(\nu t)\exp\left(-\left[i\omega + \frac{\gamma_a - \gamma_b}{2}\right]t\right)\tilde{C}_a, \tag{B21}$$

where

$$\omega = \omega_a - \omega_b.$$

Taking into account that

$$\cos(\nu t) = \frac{1}{2}\left(e^{i\nu t} + e^{-i\nu t}\right)$$

we find

$$\frac{d\tilde{C}_a}{dt} = i\frac{\Omega}{2}\left[\exp\left(\left[i(\omega+\nu) + \frac{\gamma_a - \gamma_b}{2}\right]t\right) + \exp\left(\left[i(\omega-\nu) + \frac{\gamma_a - \gamma_b}{2}\right]t\right)\right]\tilde{C}_b, \tag{B22}$$

$$\frac{d\tilde{C}_b}{dt} = i\frac{\Omega}{2}\left[\exp\left(-\left[i(\omega+\nu) + \frac{\gamma_a - \gamma_b}{2}\right]t\right) + \exp\left(-\left[i(\omega-\nu) + \frac{\gamma_a - \gamma_b}{2}\right]t\right)\right]\tilde{C}_a. \tag{B23}$$

Next we omit the fast oscillating terms (containing $\omega + \nu$). Such simplification is known as the Rotating Wave Approximation (RWA). In the RWA Eqs. (B22) and (B23) reduce to

$$\frac{d\tilde{C}_a}{dt} = i\frac{\Omega}{2}\exp\left(i\Delta t\right)\tilde{C}_b, \tag{B24}$$

$$\frac{d\tilde{C}_b}{dt} = i\frac{\Omega}{2}\exp\left(-i\Delta t\right)\tilde{C}_a, \tag{B25}$$

where

$$\Delta = \omega - \nu - i\frac{(\gamma_a - \gamma_b)}{2}$$

is the detuning of the frequency of the applied field ν from the atomic transition frequency $\omega = \omega_a - \omega_b$.

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