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Practicable X-Ray Amplifier

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Inverted populations of practical interest in the x-ray region are obtainable from selective inner-shell vacancy production in intermediate-energy ion-atom collisions. A highcurrent ion beam swept at nearly the speed of light along the length of an extended foil provides an active region in synchronization with a resonant x-ray pulse traveling parallel to the foil surface. Significant amplification may be realized with presently available ion sources.

Means of creating population inversions at vacuum uv and x-ray wavelengths are now available and system designs for achieving amplified x-ray emissions are realizable.¹ The large cross sections² for selective production of atomic innershell vacancies by heavy-ion bombardment at intermediate energies (~1 keV/amu) of metal targets indicate an obvious mechanism. The physical process is thought to be an electron promotion mechanism that occurs at level crossings in the quasimolecule formed during the collision, and/or rotational excitation.³ The vacancies produced may occur in the ion or the atom or both. Thus for ions passing through a thin foil, on the downstream side of the foil a significant number of ions will contain inner-shell vacancies. By a judicious choice of collision partners as well as beam energy and foil thickness, population inversions of interest may be achieved. Moreover, by sweeping the ion beam along the length of an extended target, such population inversions may be obtained in synchronism with a traveling wave front due to radiative decay of the states produced. Such a system would be capable of amplifying by stimulated emission this wave front advancing in the direction of sweep of the beam. Systems of this nature would be of interest when operated in a single-pass mode with no cavity,⁴ or when placed in a suitable x-ray laser cavity (such as is now being investigated⁵ or may be available at the time of realization of this system).

For ion-atom combinations such that innershell energy levels of interest match, ion to atom, cross sections for the selective production of vacancies in these shells are given approximately by taking for the level-crossing radius a value equal to the sum of the radii of the two electronic shells involved.^{2, 6} As an example, the match in energy between the carbon K shell and the argon L shell allows selective vacancies to be produced in the L shell of argon. In particular, measurements⁷ show that for bombarding energies below ~80 keV effects of double L-shell excitation are not observed. Furthermore, it was found that at an ion energy of ~50 keV, the resulting x-ray spectra indicated the strong predominance of the 224-eV line due to a 3s - 2p transition. The radiative lifetime of this transition may be taken as ~ $2.8 \times 10^{-11} \text{ sec}^8$ and a fluorescence yield of 1.67×10^{-3} is reasonable.⁹

A system design is indicated in Fig. 1. A highcurrent, well-focused, heavy-ion beam, in the energy range of interest (30 to 500 keV), is deflected by a parallel-plate transmission system



FIG. 1. Schematic of x-ray traveling wave amplifier.

using a deflection pulse applied from the downstream end of the system. The pulse (on the order of tens of kilovolts) travels in the upstream direction at a linear rate of approximately the speed of light. At a well-defined constant distance behind this advancing wave front, the stream intersects the low-potential plate which houses the foil. Thus, on the back side of the foil there will exist a thin layer of ions with selected innershell vacancies. Furthermore, this region will have an area in the plane of the foil of approximately the cross section of the beam intersection with the foil and it will travel in the upstream direction at a linear rate near the speed of light. A fraction of the excited states produced (as well as perhaps many of the unexcited ions) may possibly be in an aligned condition.¹⁰ Significant line-of-sight amplification for a resonant x-ray pulse originating near the terminal end of the tube and proceeding along the back side of the foil in the upstream direction may occur and an enhanced output obtained on the chosen x-ray line.

Consider a system in which a 30-kV argon ion beam is deflected by a pulse such that the beam changes potential energy by 20 keV due to motion transverse to the original beam direction. The beam will intersect the foil a distance $2l(fV_{b0})$ $(V_d)^{1/2}$ downstream from the position of interception of the advancing wave front. Here, l is the parallel-plate separation distance, V_{b0} the undeflected beam voltage, V_d the deflection voltage, and f the fractional distance of the undeflected beam from the plate housing the foil. For V_{b0} = 30 kV, $V_d = 40$ kV, and $f = \frac{1}{2}$, the downstream distance is $\sim 1.2l = 6$ cm for a plate separation of 5 cm. The beam will intersect the foil at an angle α to the foil surface, where $\sin \alpha = (20/50)^{1/2}$, since the beam energy will be 50 keV at this point. Then $\alpha \simeq 39^{\circ}$. Thus, if a carbon foil of thickness 5 $\mu g/cm^2$ is used, the apparent thickness to the beam will be 7.79 $\mu g/cm^2$.

As ions traverse the foil, the number of excitations per unit time occurring between x and x +dx into the foil is Ndx/λ , where $N = N_0 e^{-x/\lambda}$ $(N_0$ is the number of ions incident per unit time) and $\lambda = 1/N_f \sigma^*$ is the mean free path for innershell excitation, σ^* being the cross section for this process and N_f the atomic density of the foil. The probability that these excited ions exit the back side of the foil in such a state is $\sim \exp[-(t_b - x)/v_b \tau] \exp[-(t_b - x)/\lambda]$, the first factor being due to the exponential decrease in population due to spontaneous decay (both Auger and radiative) with a lifetime τ , the ion having a speed v_b , and

the second factor being due to subsequent atomion collisions leading to large-angle scattering and hence loss from the excited beam. The mean free path for the latter process is taken to be that for inner-shell excitation, an approximation valid because of the screened Coulomb nature of the scattering interaction and the distance of closest approach needed for inner-shell excitations.^{2b,6,11} Then the fractional excitation obtained on the exit side of the foil is $v_b \tau \lambda^{-1} \exp(-t_b/\lambda) [1 - \exp(-t_b/\lambda)]$ $v_{b}\tau$]. This ratio is maximized by taking $t_{b} = t_{bm}$ $= v_b \tau \ln(1 + \lambda/v_b \tau)$. With $N_f \simeq 1.13 \times 10^{23}$ atoms/ cm³ for carbon, $\sigma_{Ar} +_{on C} = 0.795 \times 10^{-18} \text{ cm}^2$,²a $\tau \simeq \Delta t_{Auger} = 3.84 \times 10^{-14} \text{ sec, and } v_b = 4.90 \times 10^7 \text{ cm}/$ sec at 50 keV primary beam energy, $t_{bm} \simeq 8.6 \ \mu g/$ cm² thus requiring the foil thickness specified above. The fraction of useful excitations then becomes $\sim \frac{1}{8}$. This ratio may be improved by increasing the primary beam energy (consequently decreasing λ and increasing v_b). At 50 keV, the range of Ar⁺ on C is $10.4 \pm 2.7 \ \mu g/cm^{2.12}$ Should range be a problem, an increase in energy may be in order.

The number of excited states existent at any one time is $N^* \simeq R \Delta t_{Auger}$, where R is the production rate. Because of the deflection scheme chosen, a large number of beam particles intersect the foil per unit time. In particular, the point of intersection of the beam with the foil travels a 1-m distance in a time (1 m)/c during which the number of particles intersecting the foil will be the number of beam particles per meter length, $N_{b\lambda}$, plus the number of beam particles that advance across an arbitrary position in the undeflected beam during this time, $N_{b\lambda}v_b/c$. Since $\sim \frac{1}{R}$ of these become excited ions that emerge from the foil, the useful production rate is $I_{bc}(1)$ $+v_{b}/c)/8ev_{b}$. Utilizing recently developed ion sources,¹³ 500-mA argon ion beams at 30 kV are realizable. Then $R \simeq 3.09 \times 10^{20} \text{ sec}^{-1}$ and N^* $\simeq 1.19 \times 10^7$ excited states. In the time Δt_{Auger} , the active region moves a distance $\sim 1.15 \times 10^{-2}$ mm. If the beam is focused to an ellipse of major axis 8 mm and minor axis 3 mm, the geometry of intersection will be 12.6 mm by 3 mm for an area of 29.8 mm² and will be essentially static in a time interval Δt_{Auger} . During this time, the excited ions will occupy a volume of $v_{b\perp}\Delta t_{Auger}$ $\times 29.8 \text{ mm}^2$, where $v_{b\perp}$ is the component of ion velocity perpendicular to the foil upon exiting the foil. From stopping power data,¹⁴ ions will emerge with an energy of ~ 9 keV at an average angle of 39°. Then $v_{b\perp} \simeq 1.32 \times 10^7$ cm/sec and the volume occupied by the N^* particles is ~1.51

 $\times 10^{-7}$ cm³, giving an excited density of ~0.79 $\times 10^{14}$ cm⁻³. In general, the excited density is $I_{b}c/8ev_{b0}v_{be}A_{b}$, where v_{b0} is the beam speed before deflection and v_{be} is the beam exit speed. The density of excited states may be taken as a measure of the inverted population density since most nonexcited states will contain electrons in both the upper and lower levels. Furthermore, since the coherence length of the radiation (neglecting pulse-shaping effects) is ~8.4 mm, the 12.6-mm length of active volume provided is sufficient for full amplification of the pulse.

Since angular scattering approximates that due to a screened Coulombic interaction and the probability of inner-shell vacancy production rises rapidly from zero to a fixed high value as the distance of closest approach decreases,^{2, 6, 11} vacancy-producing collisions are estimated to scatter ions into a cone at $\theta \sim 5^\circ$ with a $d\theta$ of 3° to 4° . With the beam incident at 39.2°, one segment of the scattered cone lies within an angular spread from 30.7° to 34.2° giving a spread in ion velocity parallel to the foil of 6.81×10^5 cm/sec. This results in an inhomogeneous broadening of 1.23 $\times 10^{12}$ Hz. Then $\Delta v_{D\theta} / \Delta v_N \sim 34.5$. The fraction of excited particles within an angular spread of 3.5° from each other is $\sim \psi/\pi$, where $\cos\psi = \sin\theta/\sin(\theta)$ $+d\theta$), and thus $\psi/\pi \sim \frac{1}{3}$. Accordingly, if the Doppler spread calculated above is used, the effective population inversion must be decreased by this factor, giving 2.63×10^{13} cm⁻³.

Under these conditions, the negative absorption coefficient becomes 5.42/m for a gain of 23.5 dB/ m. It is of interest to enumerate some of the effects neglected above. Although the main branching ratio implied by fluorescence yield was considered, branching due to competing radiative decay modes was neglected. However, previous $work^7$ indicates that a single intercombination may predominate. Also, since the branching ratio is due primarily to fluorescence yield, argon represents a highly unfavorable choice.⁸ Furthermore, orientation effects known to exist¹⁰ in the excitation scheme employed have been ignored. Although straggling effects on the ion energy spread have been neglected, the radiative linewidth used is perhaps low since the radiative half-life was chosen as large as is reasonable. Even though the extent to which an excited system may release energy through cooperative decays to produce superradiance is now being actively investigated,¹⁵ the existence of such events represents an attractive possibility. Finally, the best combination of collision partners is most likely

not the case considered and can only be determined over a period of time and with much experimentation. An attractive candidate appears to be argon ions on titanium.^{2c}

For neon ion-copper atom collisions, level matching between the K shell of Ne⁺ and the L shell of Cu results in high cross sections¹⁶ for selective vacancy production in these shells. Furthermore, evidence is accumulating that for K vacancy production, the vacancies are almost exclusively produced in the lower-Z collision partner.^{2d, 2e} Additionally, in this case the fluorescence yield is at least in order of magnitude higher than for the argon L shell.^{8, 17} Thus a significant output of 874-eV photons may be more easily realized than in the case detailed above.

Valuable contributions to this work were made by J. M. Joyce, J. L. Cox, Jr., and J. J. Kim.

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Generation of Vacuum Ultraviolet Radiation in Phase-Matched Cd Vapor*

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We report the generation of 1773-, 1520-, and 1182-Å radiation by frequency tripling and summing in a phase-matched mixture of Cd and Ar. For the third-harmonic process, $5320 \rightarrow 1773$ Å phase matching occurs for Cd and Ar atoms in the ratio 1:25, and $\chi^{(3)} \approx 2 \times 10^{-34}$ esu/atom. The energy conversion efficiency to 1773 Å was about 10^{-4} , yielding a peak picosecond power of 7 kW.

Because of the increased difficulty of obtaining laser oscillations in the vacuum-ultraviolet (vuv) and soft x-ray regions of the electromagnetic spectrum,^{1,2} it is of interest to consider other techniques which are capable of generating coherent radiation in these spectral regions. One possible approach is to use cascaded frequency triplers to convert the very high peak power now available at 1.06 μ m to much shorter wavelengths. In this Letter we report the generation of 1.06- μ m harmonic vuv radiation at 1773, 1520, and 1182 Å.

To obtain radiation at the above frequencies we employed the technique of phase-matched harmonic generation and frequency summing in a mixture of a metallic vapor and an inert gas. This technique was first suggested by Harris and Miles³ and experimentally demonstrated by Young *et al.*⁴ In these first experiments a mixture of Rb vapor and Xe was used for the third-harmonic process 1.06 μ m \rightarrow 3547 Å. To extend this technique to the vuv we employed a mixture of Cd and Ar. The choice of Cd was indicated for two reasons: First, its nonlinear susceptibility in the vuv should be enhanced by its strong atomic transitions which extend from the fundamental resonance line at 2288 Å to the beginning of the continuum at 1378 Å. Second, as is the case for the alkali metals, Cd has a relatively small absorption cross section in the spectral region just above its ionization potential ($\sigma = 7 \times 10^{-20}$ cm² at $\lambda = 1182$ Å).⁵ To obtain efficient harmonic or sumfrequency generation it is necessary that the driving dipole polarization wave travel at the same velocity as the electromagnetic wave which it is desired to generate. This phase-matched condition is obtained by correctly choosing the ratio of Cd to Ar atoms.

We describe three nonlinear processes. These are (1) tripling of 5320 Å to yield 1773 Å, (2) summing of 1.064 μ m with 3547 Å to yield 1520 Å, and (3) tripling of 3547 Å to yield 1182 Å. The exper-