PHYSICAL REVIEW A VOLUME 12, NUMBER 2 AUGUST 1975

Feasibility of coherent x-ray production by x-ray pumping

Paul L. Csonka*†
Institute of Theoretical Science and Department of Physics, University of Oregon, Eugene, Oregon 97403

Bernd Crasemann‡
Department of Physics, University of Oregon, Eugene, Oregon 97403
(Received 15 July 1974; revised manuscript received 17 March 1975)

It is suggested that coherent soft x rays can be produced by inverting the electron population in a suitable target, such as Li, through irradiation with x rays generated by fast electrons traversing an electromagnetic field (as in a storage ring). Conditions to be satisfied by target and radiation parameters are stated, and examples given.

I. INTRODUCTION

Coherent x-ray sources would provide us with an important tool in basic research and for industrial applications. Coherent x rays could be produced through stimulated emission from a working substance containing an inverted electron population, e.g., from an assemblage of atoms with K-shell vacancies. Laser photons have been used for pumping,[1] with varying success. Pumping with x rays of energy just above the K edge[2] would offer the advantage of high efficiency because (i) the photoelectric cross section is large, and (ii) the pumping radiation would be used directly, rather than through an intermediary plasma.

X rays radiated by fast electrons passing through an electromagnetic field could be suitable for pumping. The electrons may be circulating in a storage ring; the field may be the regular field of the ring or an intense magnetic field inserted in the path of the electrons to cause a sudden "bend" or "wave" in their orbit,[3,4] or a radiation field shaped by an optical element called the "template" causing fast electron oscillations and associated x-ray emission.[5]

In the present paper, we examine the conditions that must be satisfied for coherent x rays to be produced through pumping with x rays, specifically with synchrotron-radiation pulses. We discuss the suitability of Li and LiH as the active medium and show that it may be possible to pump LiH adequately with photon pulses that exceed by 2 to 4 orders of magnitude in intensity those now attained in the Stanford Synchrotron Radiation Project (SSRP), for example.

II. GENERAL CONSIDERATIONS

We consider a target (Fig. 1) containing "active atoms" (the working substance) as well as other "inert atoms." The pumping x-ray beam travels parallel to the z axis, entering the target at $z=0$. We require that several conditions be satisfied.

(a) The coherence length of the emitted x rays (i.e., the distance they can travel before the various Fourier components in the beam begin to get significantly out of phase) must exceed the dimensions of the target; otherwise x rays produced in different parts of the target cannot be coherent. When the index of refraction is ~1 and the target dimensions are chosen as indicated at the end of this paper, then this restriction is always satisfied.

When an approximately plane wave with wavelength λ_0 travels along the z axis and the extension of the wave front along the x and y axes is a_x and a_y, respectively, then the energy density of the wave at the z axis will decrease appreciably after the wave has traveled a distance $\sim a_x^2\lambda_0^{-1}$ or $a_y^2\lambda_0^{-1}$, whichever is smaller. Therefore, focusing such a wave on a spot at $z=\frac{1}{2}d_z$, we require

$$d_z \leq \min(a_x^2/\lambda_0, a_y^2/\lambda_0) . \qquad (1)$$

(b) Photons of the stimulated-emission energy E_0, traveling in the z direction, are absorbed with a total cross section σ_{ti} by inert atoms, of which there are ρ_i per unit volume, with σ_{ta} by active atoms in their ground state (ρ_a per unit volume), and with σ_{tK} by active atoms containing a K vacancy (of which there are ρ_K per unit volume):

$$dN(E_0)/dz = -(\rho_i\sigma_{ti}+\rho_a\sigma_{ta}+\rho_k\sigma_{tK}) , \qquad (2)$$

where all cross sections are evaluated for the energy E_0. If σ_s is the stimulated-emission cross section, $N(E_0)$ will increase as

$$dN(E_0)/dz = \rho_K\sigma_s , \qquad (3)$$

and there will be a net increase if Eq. (3) exceeds Eq. (2). If almost all active atoms contain a K vacancy ($\rho_a \approx 0$), this condition is

$$\rho_K\sigma_s = (\rho_i\sigma_{ti}+\rho_K\sigma_{tK})\beta, \quad \beta > 1 . \qquad (4)$$

(c) The number of x-ray photons emitted per unit target volume in a short time interval Δt by spontaneous K-vacancy decay is $\rho_K \Delta t/\tau_K$. Each of these photons can stimulate the emission of more photons, which in turn can cause further induced emission, etc. Photons from spontaneous decays are emitted randomly in all directions. A few, with momentum parallel to z, have a chance to produce a strong coherent x-ray pulse by deexciting atoms all along the target. Most, however, leave the target "sideways" after traveling only a short distance. They are undesirable, because by deexciting some atoms they reduce ρ_K. The number of these undesirable deexcitations must be kept small. If the exponential increase of photon number along the path of each undesired spontaneously emitted photon can be approximated by a linear increase, then the number per volume element of photons produced in Δt by undesirable induced emission is $(\Delta t/\tau_K)\rho_K^2 \sigma_s d$ (where d is the average distance in the target traveled by the undesirable photons). We wish to insure that this quantity is much smaller than ρ_K. For a long thin target, with d of the order of d_y, we require $(\Delta t/\tau_K)\rho_K \sigma_s d_y \ll 1$. The probability of a randomly emitted photon traveling the whole length of the target is $\sim(d_y/d_z)(d_x/d_z)(4\pi)^{-1}$, hence we also require $(4\pi)^{-1}\omega_K(\rho_K d_x d_y d_z)(d_y d_x/d_z^2) \ll 1$. We also want to keep the number of spontaneous deexcitations during Δt small, i.e.,

$$\Delta t/\tau_K \ll 1 . \quad (5a)$$

It follows that we need

$$\epsilon \equiv \rho_K \sigma_s d_y < 1 . \quad (5b)$$

If Δt is so large that inequalities (5a) and (5b) do not hold, then a non-negligible fraction of all K vacancies is filled by undesirable processes.

(d) The pumping x-ray photons have momentum $\vec{k}_0$ parallel to the (y, z) plane, and the angle between $\vec{k}_0$ and the z axis is θ (Fig. 1). The path length of unabsorbed pumping photons in the target is $\sim d_y/\sin\theta$. We neglect spatial variations of ρ_i, ρ_a, and ρ_K within the target and assume that only a small fraction of all pumping photons is absorbed. Then during one passage through the target, a pumping photon will augment ρ_K by creating $\rho_a \sigma_p d_y/\sin\theta$ K vacancies, where σ_p is the appropriate photoionization cross section. Let τ_b be the duration of a pumping photon beam pulse, for which we assume the beam intensity to be a step function in time. The number of pumping photons per unit volume and unit time (during τ_b) is $N_\gamma(d_x d_y d_z \tau_b)^{-1}$, so that we have

$$\frac{-d\rho_a}{dt} = \frac{d\rho_K}{dt} = \frac{N_\gamma}{\tau_b}(d_x d_y d_z)^{-1}\bar{\sigma}_p \rho_a d_y (\sin\theta)^{-1} , \quad (6)$$

where $\bar{\sigma}_p$ is averaged over the energy of the pumping photons. We have neglected unwanted spontaneous decay, in accordance with condition (5). Let pumping start at $t=0$, then we have the boundary condition $\rho_K(0)=0$, with which the solution of Eq. (6) is

$$\rho_K(t) = \rho_a(0)[1 - \exp(-N_\gamma \bar{\sigma}_p t/\tau_b d_x d_z \sin\theta)] . \quad (7)$$

The condition $\rho_K(0)=0$ implies

$$\rho_a(t) + \rho_K(t) = \rho_a(0) . \quad (8)$$

If at some time Δt a K vacancy exists in almost all active atoms, then $\rho_a(\Delta t)/\rho_K(\Delta t) \ll 1$. Using Eqs. (7) and (8), this can be rewritten

$$\delta \equiv \exp(-N_\gamma \sigma_p \Delta t/\tau_b d_x d_z \sin\theta) \ll 1, \quad \text{if } d_y/\sin\theta \lesssim d_z . \quad (9)$$

(e) After one photon of the right energy E_0 enters the target with momentum $\vec{k}_0$ parallel to the z axis, the number of photons with the same energy and momentum will increase exponentially due to induced emission. At $z = d_z$ we have

$$N(E_0, \vec{k}_0, z) = \exp\{[-\rho_i \sigma_{ti}(E_0) - \rho_a \sigma_{ta}(E_0) - \rho_K \sigma_{tK}(E_0) + \rho_K \sigma_s] d_z\} . \quad (10)$$

If ρ_a can be neglected, then

$$N(E_0, \vec{k}_0, z) = \exp\{[\rho_K(\sigma_s - \sigma_{tK}(E_0)) - \rho_i \sigma_{ti}(E_0)] d_z\} . \quad (11)$$

The device will produce significant amplification if

$$f \equiv \ln N(E_0, \vec{k}_0, z) = [\rho_K(\sigma_s - \sigma_{tK}(E_0)) - \rho_i \sigma_{ti}(E_0)] d_z \gg 1 . \quad (12)$$

Clearly, this implies the condition expressed by Eq. (4).

In the above discussion, it was implicitly assumed that the photon under consideration passes through the target with certainty: $a_x \leqslant d_x$, $a_y \leqslant d_y$. When this is not so, then ρ must be replaced in all of the above equations by ρ_{eff}, the "effective density" (seen by the photon). For example, if

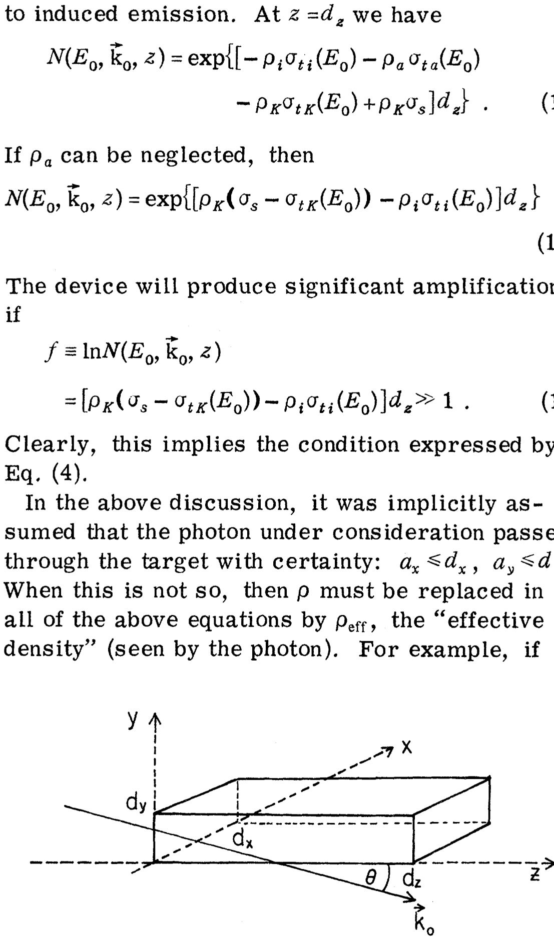

FIG. 1. Orientation of the target and momentum $\vec{k}_0$ of incident pumping photons, in the x, y, z coordinate frame.

$a_x \leq d_x$ but $a_y > d_y$, we have $\rho_{a\,\mathrm{eff}} = d_y a_y^{-1} \rho_a$. Similarly, ρ_K and ρ_i must be multiplied by $d_y a_y^{-1}$ to obtain their effective values.

The cross section for stimulated emission is

$$\sigma_s \cong \frac{\lambda_0^2}{4\pi^2} \frac{\tau_{\mathrm{rad}}^{-1}}{\Delta\nu_{\mathrm{tot}}} . \qquad (13)$$

If the total linewidth $\Delta\nu_{\mathrm{tot}}$ is determined by the Auger and radiative widths of the K-vacancy state alone, then σ_s can be expressed in terms of the K-shell fluorescence yield ω_K as

$$\sigma_s \cong (\lambda_0^2/2\pi)\omega_K . \qquad (14)$$

III. WORKING SUBSTANCE

To satisfy conditions (1), (4), (5), (9), and (12), we look for an atom with a relatively long K-vacancy lifetime τ_K. For atoms with more than three electrons, we have $\tau_K \lesssim 10^{-14}$ sec.[6] However, an isolated Li atom with a K-shell vacancy has a mean life of 500 μsec for the two-photon 60.7-eV $E1$ decay from the $^1S_0$ state[7,8] and a mean life of 50 sec for the $M1$ decay from the $^3S_1$ state[9]; Auger transitions are impossible, whence $\omega_K = 1$. We find $\sigma_p \approx 5\times10^{-18}$ cm^2 near 60 eV.[10] When the atom is not isolated, the effect of neighboring atoms must be taken into account. We consider three special cases.

A. Lithium gas

In a pure Li gas, we have $\rho_i = 0$, and the K-vacancy lifetime τ_K is governed by the collisional deexcitation rate, i.e., by the density. The average distance traveled by a Li atom between collisions will be

$$l_{\mathrm{coll},a} = [(\rho_a + \rho_K)\alpha\sigma_d]^{-1} , \qquad (15)$$

where $\alpha\sigma_d \cong 10^{-15}\ \alpha$ cm^2 is the maximum cross section (over the velocity range of interest) for filling a K vacancy by collisional deexcitation; α is of order one. Furthermore if deexcitation in collisions between ions and free electrons is taken into account, it is found that very low density $\rho_K < 4\times10^{11}(\alpha\beta)^{-1}$ cm^{-3} and an unreasonable length $d_z \sim 10^6$ cm would be required to satisfy conditions derived in Sec. II.

B. Metal

When the Li is located in a metal (either pure Li, or an alloy), a 10–50% p-state admixture in band electrons might lead to a radiative lifetime of the order of 1 nsec; τ_K would be governed by radiationless transitions; one could expect $\omega_K \approx 10^{-4}$, but $\Delta\nu_{\mathrm{tot}}$ would be large, and hence, σ_s small [Eq. (13)].

C. Lithium hydride

In most crystals, τ_K is short, as can be inferred from LiF Auger spectra.[11] The total lifetime will be approximately determined by the Auger rate. However, one would expect radiationless transition probabilities to be much smaller than in the metal, particularly for ionic bonding, because Li is an electron donor. In LiH, one may expect $\omega_K \approx 10^{-3}$ and $\tau_K \approx 10^{-13}$ sec, possibly longer. We consider gaseous LiH in some detail.

IV. PUMPING OF LiH WITH SYNCHROTRON RADIATION

Let the pumping x rays be synchrotron radiation from an electron beam of dimensions comparable to those in the interaction region of the storage ring SPEAR at the Stanford Linear Accelerator Center. The rms width of the electron beam there is 3×10^{-2} cm, while the vertical beam dimension is smaller and depends upon the coupling between modes of beam oscillation.[12] We assume that the angular divergence of the pumping radiation is $\Delta\theta = 1$ mrad, as for the radiation from SPEAR when the circulating electron energy is 1.5 GeV. We take the spectrum of the pumping radiation to be constant from 65 to 85 eV and zero elsewhere, which can approximately be achieved with appropriate mirrors and windows. (The energy is kept above E_0 so that the pumping radiation cannot induce stimulated emission.) Then we have $\bar{\sigma}_p = 5\times10^{-18}$ cm^2 and $\bar{\sigma}_{ti} = 1.5\times10^{-19}$ cm^2 for the inert H atoms.

For the dimensions of the LiH target (Fig. 1) we choose $d_x = 10 d_y = 3\times10^{-4}\alpha^{1/2}$ cm, $d_z = 8\times10^{-4}\alpha$ cm, where α, of order unity, is defined through Eq. (15). The extent of the wave front is $a_x = d_x$ and $a_y = d_y$.[13] The target contains LiH gas at a temperature of 1700 °K (at $t = 0$) and a density $\rho_a = 10^{20} \times (2\alpha)^{-1}$ cm^{-3}. The LiH is in chemical equilibrium with H_2 of density $\rho_{H_2} = 10^{21}\alpha^{-1}$ cm^{-3}. Calculations show that under these conditions there is negligible ionization of the LiH, and the Li gas density is only $\rho_{\mathrm{Li}} < 0.3\alpha^{1/2}\rho_{\mathrm{LiH}}$.

The collision mean free path is $l_c \approx 3.6\times10^{-4}\alpha$ cm. The LiH gas density and the target dimensions have been chosen so that the K-vacancy lifetime in LiH is not reduced by more than a factor of 2 through collisional deexcitation.

There are $3.4\times10^8\alpha$ LiH molecules in the target, and ~$1.7\times10^9\alpha$ x-ray photons are needed to create K vacancies in most of them.[14] The photons must be delivered in a time interval that is shorter than τ_K. A Gaussian x-ray pulse of duration τ_b must therefore contain ~1.3×10^{13} photons in the desired energy range if $\tau_K = 10^{-14}$ sec, or ~1.3×10^{11} photons if $\tau_K = 10^{-12}$ sec.[15] At SSRP, the number of photons per 50-mA pulse in the pertinent energy range is $\sim 5\times 10^8$, within 2 mrad for a circulating electron energy of 2.5 GeV. This intensity falls short, by 10^2–10^4, of the necessary value to create coherent soft x rays by the scheme we have described. The synchrotron pumping radiation intensity could be increased (i) by using only one electron beam, and (ii) by operating at lower circulating electron energies, if instabilities can be avoided, which results in smaller beam dimensions. It thus appears that this method of producing coherent soft x rays may become feasible in the near future, particularly if equipment specifically designed for this purpose[3-5] were available.

For materials with shorter λ_0, the same considerations hold, but because of the shorter τ_K, a shorter Δt is required, implying higher N_γ/τ_b.

This line of thinking should encourage detailed study of ω_K for elements such as Li, in various chemical surroundings.

ACKNOWLEDGMENTS

We wish to thank M. H. Chen, T. R. Dyke, G. E. Ice, J. C. Kemp, J. W. McClure, J. Rees, G. H. Wannier, and particularly H. Winick for discussions concerning this subject.

---

*Alfred P. Sloan Fellow.

†Work supported in part by the U. S. Atomic Energy Commission.

‡Work supported in part by the U. S. Army Research Office - Durham, and by the National Aeronautics and Space Administration.

[1] M. A. Duguay, in *Proceedings of the International Conference on Inner-Shell Ionization Phenomena and Future Applications, Atlanta, Georgia, 1972,* edited by R. W. Fink, S. T. Manson, J. M. Palms, and P. V. Rao, CONF-720404 (U. S. Atomic Energy Commission, Oak Ridge, Tenn., 1973), p. 2350.

[2] M. A. Duguay and P. M. Rentzepis, Appl. Phys. Lett. 10, 350 (1967); Yu. L. Stankevich, Dokl. Akad. Nauk SSSR 191 No. 4, 805 (1970) [Sov. Phys.—Doklady 15, 356 (1970)].

[3] P. L. Csonka, Phys. Lett. 24B, 625 (1967); CERN Report No. 67-15 (unpublished); P. L. Csonka and K. S. Kölbig, Phys. Rev. D 10, 251 (1974).

[4] H. Winick (private communication).

[5] P. L. Csonka, University of Oregon I.T.S. Report No. N.T. 052/74 (unpublished).

[6] V. O. Kostroun, M. H. Chen, and B. Crasemann, Phys. Rev. A 3, 533 (1971); W. Bambynek, B. Crasemann, R. W. Fink, H.-U. Freund, Hans Mark, C. D. Swift, R. E. Price, and P. Venugopala Rao, Rev. Mod. Phys. 44, 716 (1972).

[7] G. W. F. Drake, G. A. Victor, and A. Dalgarno, Phys. Rev. 180, 25 (1969).

[8] M. H. Prior and H. A. Shugart, Phys. Rev. Lett. 27, 902 (1971).

[9] G. W. F. Drake, Phys. Rev. A 3, 908 (1971).

[10] W. J. Veigele, E. Briggs, L. Bates, E. M. Henry, and B. Bracewell, Kaman Sciences Corporation Report No. KN-71-431(R), 1971 (unpublished).

[11] T. E. Gallon and J. A. D. Matthew, Phys. Status Solidi 41, 343 (1970).

[12] A. D. Baer, R. Gaxiola, A. Golde, F. Johnson, B. Salsburg, H. Winick, M. Baldwin, N. Dean, J. Harris, E. Hoyt, B. Humphrey, J. Gurow, R. Nellen, J. Niljan, and G. Warren, IEEE Trans. Nucl. Sci. NS-22, 1794 (1975).

[13] The pumping x rays can be focused on a spot of the chosen dimensions, provided that focusing mirrors are available for which the focal length is $\gtrsim 4\alpha^{1/2}$ times the mirror diameter, and provided the coupling constant between vertical and horizontal beam oscillations is ~ 0.1, a value already reached at SPEAR.

[14] The quantity $(4\pi)^{-1}\omega_K\rho_a d_x^2 d_y^2 d_z/l_c^2$ is large under the assumed parameters; hence spontaneous deexcitation events are likely. The quantity is reduced to ~ 1 if one chooses $d_x = d_y = 10^{-5}\alpha^{1/2}$ cm, while leaving a_x and a_y unchanged; then the probability of premature autodeexcitation is $\sim\frac{1}{2}$. The number of LiH molecules in the target, however, is then reduced by ~ 80, while the same number of pumping photons is required as before.

[15] The early part of the Gaussian pumping-radiation pulse could be kept from reaching the target by interposing an opaque foil that evaporates as the pulse approaches peak intensity.