if the limit $\delta/T \to 0$ is taken.

Now if $\epsilon = 8T + \delta$ is to be the energy of the bound state with δ vanishingly small, we need [cf. Eq. (3.38)]

$$\lim_{\delta/T\to 0} f(8T+\delta) = 4/U_0 \quad , \tag{C8}$$

i.e.,

$$\frac{2}{T} - \lim_{\delta/T\to 0}\frac{1}{[4T^2 G_{\vec{0},\vec{0}}(8T+\delta)]} = \frac{4}{U_0} \quad .$$

But

$$\frac{1}{G_{\vec{0},\vec{0}}(8T+\delta)} = \frac{\lambda_1^{1/2}}{2K(m)/\pi} \xrightarrow[(\delta/T)\to 0]{} \frac{4T^{\frac{1}{2}}\pi}{[-\frac{1}{4}\ln(4\delta/T)]}$$

$$= \frac{-8\pi T}{\ln(4\delta/T)} \quad . \tag{C9}$$

Therefore the equation for δ is

$$\lim_{\delta/T\to 0} f(8T+\delta) = \frac{2}{T} + \frac{2\pi}{T\ln(4\delta/T)} = \frac{4}{U_0} \, , \tag{C10}$$

and we readily obtain the expression connecting δ with U_0 in the neighborhood of $U_0 = 2T$ as

$$\delta/T = \tfrac{1}{4}\exp[-\pi/(1 - 2T/U_0)] \quad . \tag{C11}$$

This gives the desired threshold behavior of the binding energy, $\epsilon/T = 8 + \delta/T$.

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Mixing of X-Ray and Optical Photons

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Nonlinear effects involving x-ray and optical photons are described with particular emphasis on the generation of sum and difference frequencies. Efficiencies for sum and difference frequency generation are calculated and found to be large enough to be observable. The expected advent of x-ray lasers should enhance the usefulness of such mixing techniques in the measurement of excited-state wave functions. Under favorable circumstances, the mixing technique may provide a means of efficiently tuning x-ray laser outputs.

INTRODUCTION

Although x-ray lasers are not now available, there exists the possibility of observing and investigating nonlinear x-ray effects. We have recently observed spontaneous parametric x-ray conversion.[1] Freund and Levine[2] pointed out that this appeared to be a feasible endeavor, and also considered other nonlinear x-ray effects such as x-ray harmonic generation.

Here nonlinear effects involving optical and x-ray photons are described with particular emphasis on generation of sum and difference frequencies. If an x-ray laser is developed, then such processes could be used to shift x-ray laser outputs by a small but precise and significant amount. The sum and difference frequency generation may be described as Bragg scattering from an optically induced microscopic charge distribution. Measurements of the mixing conversion efficiency can yield

detailed information about the excited states responsible for the microscopic charge distribution in much the same way that previous Bragg-scattering measurements have yielded information about ground-state charge density. The ability to probe microscopic light-induced charge densities is unique to radiation of short wavelength such as x rays. This contrasts with optical measurements, which can directly measure only macroscopic quantities.

The sum and difference frequency production may be described as follows: The incident light beam in a crystal interacts with the atoms resulting in a microscopic charge density which differs from the ground-state charge density. The induced charge density has a periodicity determined by the light wave vector and the periodicity of the crystal. The incident x-ray beam scatters according to Bragg's law from the moving induced charge density, the output x-ray frequency being up- or down-shifted, as required by the Doppler process or energy-momentum conservation.

One could proceed formally in the manner of Armstrong *et al.*[3] An examination of their work reveals that for light–x-ray mixing one type of matrix element dominates their expression for the nonlinear polarization. A calculation using those matrix elements, with proper attention given to microscopic features of the nonlinear polarization, would lead to results equivalent to those in this work.

Only sum and difference generation will be considered in detail, because other processes can be treated in a manner similar to that which follows. The work of Armstrong *et al.*[3] or Franken and Ward[4] reveals that the electric dipole approximation is usually appropriate for the treatment of strong optical effects, whereas the statement[5] that x rays scatter from the electronic charge density is accurate.

Bragg's law is rederived in a form especially adapted for application in this work. In Sec. II the microscopic light-induced charge distribution, responsible for the Bragg scattering of interest in this paper, is calculated. After a description of phase-matching conditions, in Sec. IV a simple recipe is presented for evaluating the size of the x-ray optical nonlinear interaction. The effects of beam intensities, crystal imperfection, collimation, and absorption are included with estimates of the efficiency of the process. The mixing of optical and x-ray photons can be observed with techniques available at present.

I. BRAGG SCATTERING FROM INDUCED CHARGES

An incident plane-wave monochromatic x-ray beam with a vector potential

$$\vec{A}_I(\vec{r},t)=\vec{\alpha}_I\, e^{-i(\omega_I t-\vec{k}_I\cdot\vec{r})}+\text{c.c.}\,, \tag{1}$$

where c.c. denotes the complex-conjugate term, induces a current in a medium. The expectation value of the current is $\vec{J}(\vec{r},t)=e\langle\vec{v}\rangle$, where $\vec{v}$ is the velocity operator[6] given by

$$\vec{v}(\vec{r},t)=(1/m)[\vec{P}-(e/c)\vec{A}_I(\vec{r},t)]\,, \tag{2}$$

where m is the electron mass, e is the electron charge, c is the vacuum value of the light speed, and $\vec{P}=-i\hbar^{-1}\sum_j\nabla_j$ is the total electron momentum operator, and the sum is over all electrons. The effects of nuclei are neglected because of their larger mass. Here $\langle\vec{v}\rangle$ denotes the expectation value of $\vec{v}$ at a point $(\vec{r},t)$ in space and time.

To calculate $\vec{J}$, it is necessary in the optical region to find the perturbed wave functions $\psi(\vec{r},t)$. In the x-ray region it is usually the case that the $\vec{A}_I$ term in Eq. (2) dominates the $\vec{P}$ term,[5] and the unperturbed wave functions may be used to calculate an accurate result to first order in $\vec{A}_I$. In this work the scattering is envisioned as arising from the temporarily and spatially varying charge distribution induced by light. The light predominately interacts with the weakly bound outer electrons. The resultant scattered x-ray intensity due to the $\vec{A}_I$ term is consequently larger than that due to the $\vec{P}$ term approximately by a factor ω_I/ω_B, where ω_B is an optical binding frequency.[5] Consequently, to obtain a first-order result, it is accurate to use only wave functions unperturbed by the x rays, so that

$$J(\vec{r},t)=-(e^2/mc)\langle\vec{A}_I\rangle=-r_0c\vec{A}_I(\vec{r},t)\rho(\vec{r},t)\,, \tag{3}$$

where $r_0=e^2/mc^2$ is the classical electron radius and $\rho(\vec{r},t)$ is the electron density at point $(\vec{r},t)$ in the absence of x rays.

The outgoing scattered radiation $\vec{A}_0$ in a direction $\vec{k}_0$ is determined by Maxwell's equation

$$\frac{\partial^2\vec{A}_0}{\partial\xi^2}-\frac{1}{c^2}\,\frac{\partial^2\vec{A}_0}{\partial t^2}=-\frac{4\pi}{c}\,\vec{J}(\vec{r},t)\,, \tag{4}$$

where ξ is a coordinate measured in the direction $\vec{k}_0$, and $\vec{A}_0$ is transverse to $\vec{k}_0$. If the envelope $\vec{\alpha}_0$ does not change appreciably in the distance $2\pi/k_0$, then it is appropriate to remove the rapidly varying components

$$\vec{A}=\vec{\alpha}_0\, e^{-i(\omega_0 t-\vec{k}_0\cdot\vec{r})}+\text{c.c.}\,, \tag{5}$$

where $\omega_0=k_0c$, so that

$$\frac{\partial\vec{\alpha}_0}{\partial\xi}+\frac{1}{c}\,\frac{\partial\vec{\alpha}_0}{\partial t}=\frac{2\pi i}{k_0c}\,\hat{\epsilon}\cdot\vec{J}(\vec{r},t)e^{+i(\omega_0 t-\vec{k}_0\cdot\vec{r})}\,. \tag{6}$$

It is seen that only components of $\vec{J}$ behaving as $e^{-\omega_0 t+i\vec{k}_0\cdot\vec{r}}$ contribute significantly to the radiation $\vec{A}_0$. Here $\hat{\epsilon}$ is a unit vector parallel to $\vec{\alpha}_0$.

A particular Fourier component ρ_L of the charge density is associated with a charge wave:

$$\rho_Q(\vec{r}, t) = \rho_L(\vec{Q})\, e^{-i(\omega_L t - \vec{Q}\cdot\vec{r})} + \text{c. c.} \tag{7}$$

The solution to Eq. (6), using Eq. (3) with $\rho(\vec{r}, t) = \rho_Q(\vec{r}, t)$, and taking $\mathfrak{A}_0(0) = 0$, is given by

$$\mathfrak{A}_0(\xi) = \frac{-2\pi r_0(\hat{\epsilon}^* \cdot \mathfrak{A}_I)}{k_0}\, \rho_L(\vec{Q})\, \frac{e^{-i\Delta k\xi} - 1}{\Delta k}\ , \tag{8}$$

where $\Delta k = |\vec{k}_0| - |\vec{k}_I \pm Q|$, the momentum mismatch, is assumed to be small compared to k_0. The process is restricted to small angular directions, because k_0^{-1} is a microscopic quantity. It is assumed that $\vec{k}_0$ is parallel to $\vec{k}_I \pm \vec{Q}$, $|\vec{k}_0| = \omega_0 c$, and $\omega_0 = \omega_I \pm \omega_L$. The peaking of the scattering amplitude described by Eq. (8) near $\Delta k = 0$ represents the well-known phase-matching condition for conservation of reduced momentum. The upper and lower signs refer to sum and difference frequency generation, respectively.

In the case that $\omega_L = 0$, the above result represents ordinary Bragg scattering satisfying the condition $\vec{k}_0 = \vec{k}_I \pm \vec{G}$, where $\vec{G}$ is a crystalline reciprocal lattice vector and $\rho_L(\vec{G})$ is the time-independent Fourier component of the ground-state electronic charge distribution of the lattice.

Of interest here is the case where $\rho_Q(\vec{r}, t)$ is time-dependent because it is light induced. In a crystal, the light has a macroscopic wave vector $\vec{q}_L$, but the associated charge density $\rho(\vec{r}, t)$ has Fourier components at wave vectors $\vec{Q} = \pm\vec{q}_L + \vec{G}$, where $\vec{G}$ is any of the crystalline reciprocal lattice vectors. For this case, $\rho_L(\vec{Q})$ is the Fourier component of the light-induced charge density. Thus a single light beam may cause scattering in a number of directions.

The effects of absorption of light or x rays has not been taken into account. Let $\vec{k}_I$ and $\vec{q}_L$ represent waves entering a crystal face, and $\vec{k}_0$ a wave leaving the crystal face. Let α_I, α_L, and α_0 represent, respectively, the associated absorption constants. If the z axis is the outward normal to the crystal face, then the expression for ρ should be multiplied by $\exp(\frac{1}{2}\alpha_L z/\cos\beta_L)$, and the expression for A_I sould be multiplied by $\exp(\frac{1}{2}\alpha_I z/\cos\beta_I)$. Here β_L and β_I are the angles that the light and incident x-ray beam make with face normal inside the crystal. With β_0 defined to be corresponding quantity for A_0, Eq. (6) must be modified to include on the left-hand side a term $-\frac{1}{2}\alpha_0 A_0$, and on the right-hand side, $\vec{J}$ should be multiplied by

$$e^{\alpha_T t/2} = \exp\left[\tfrac{1}{2}\cos\beta_0\left(\frac{\alpha_L}{\cos\beta_L} + \frac{\alpha_I}{\cos\beta}\right)_I \xi\right] \tag{9}$$

to take into account the attentuation of the input beams. Equation (6), using Eq. (3), becomes

$$\frac{\partial\mathfrak{A}_0}{\partial\xi} + \frac{1}{c}\frac{\partial\mathfrak{A}_0}{\partial t} + \tfrac{1}{2}\alpha_0\mathfrak{A}_0 = -i\lambda_0 r_0[\hat{\epsilon}^* \cdot \vec{\mathfrak{A}}_I(z=0)] \times e^{i\Delta k\xi_e \alpha_T/2}\rho_L(\pm\vec{q}_L + \vec{G})_{z=0}\ . \tag{10}$$

The solution to Eq. (10) is

$$\mathfrak{A}_0(z=0) = -i\lambda_0 r_0 \cos\varphi\, \mathfrak{A}_I\, \rho_L(\vec{q}_L + \vec{G}) \times [\Delta k - i\tfrac{1}{2}(\alpha_T + \alpha_0)]^{-1}\big|_{z=0}\ , \tag{11}$$

it being assumed that the crystal thickness is large compared to $(\alpha_T + \alpha_0)^{-1}$, and φ is the angle between the input and output x-ray polarization vectors. Effectively the range of momentum mismatch allowed for efficient conversion is determined by the condition $|\Delta k| < \alpha_T + \alpha_0$. The fractional scattered intensity is

$$S = \frac{|\mathfrak{A}_0|^2}{|\mathfrak{A}_I|^2}\,\frac{\cos\beta_I}{\cos\beta_0} = \frac{\lambda_0^2 r_0^2(\cos^2\varphi)|\rho_L(q_L + G)|^2}{\Delta k^2 + [\frac{1}{2}\alpha_T + \alpha_0]^2}\,\frac{\cos\beta_I}{\cos\beta_0}\ , \tag{12}$$

where the factor $\cos\beta_I/\cos\beta_0$ takes into account possibly different input and output beam diameters. If the beams are not arranged as assumed above, an analogous calculation can be easily performed. For an unpolarized x-ray input the averaging of $\cos^2\varphi$ results in a replacement of $\frac{1}{2}(1 + \cos^2 2\theta_B)$ for $\cos^2\varphi$ in Eq. (12), where $2\theta_B$ is the angle between input and output x-ray wave vectors.

II. LIGHT-INDUCED CHARGE

Light primarily interacts with the outer electrons of the atoms comprising the medium. The linear interaction between an electromagnetic wave and an atom is described by a Hamiltonian operator

$$\mathcal{H}_0 + \mathcal{H}' = \mathcal{H}_0 - (e/2mc)(\vec{P}\cdot\vec{A} + \vec{A}\cdot\vec{P})\ , \tag{13}$$

where $\vec{P}$ is the momentum operator, and $\vec{A}$ is the vector potential. In the case of light, the electric dipole terms dominate[7] and we have approximately

$$\mathcal{H}' = -\vec{p}\cdot\vec{E}\ , \tag{14}$$

where $\vec{p}$ is the electric dipole moment operator.[6] We shall see shortly that the light-induced charge is in phase with the electric field and, therefore, out of phase with the light vector potential. This feature allows a simplification of the following formulas if the light field is described through its electric field $\vec{E}$.

In a material a light wave with electric field

$$\vec{E}(\vec{r}, t) = \vec{E}_L \cos(\omega_L t - \vec{q}_L\cdot\vec{r}) \tag{15}$$

causes the medium to become polarized. The wave function of an atom at position $\vec{r}_i$ is given by

$$\psi^{(i)}(\vec{r}, t) = \varphi_g^{(i)}(\vec{r}) + \tfrac{1}{2}\vec{E}_L\cdot\sum_j \frac{\vec{p}_{Ij}}{\hbar}\,\varphi_j^i(\vec{r}) \times\left(\frac{e^{i(\vec{q}_L\cdot\vec{r} - \omega_L t)}}{\omega_j - \omega_L} + \frac{e^{-i(\vec{q}_L\cdot\vec{r} - \omega_L t)}}{\omega_j + \omega_L}\right)\ , \tag{16}$$

if one accepts the tenets of first-order steady-state

time-dependent perturbation theory in the electric dipole approximation. The spatial functions φ_j are excited-state wave functions with eigenvalues $\hbar\omega_j$ of $\mathcal{H}_0$ for the atom at $\vec{r}_i$, and $\varphi_g^{(i)}$ is the ground-state wave function with $\mathcal{H}_0\varphi_g = 0$ for the atom at $\vec{r}_i$. The vector dipole moment matrix $\vec{p}_j$ is taken between states φ_g and φ_j. With no applied magnetic field, the functions φ_g and φ_j are chosen to be real. The resultant electron density is given by

$$\rho(\vec{r}, t) = \sum_i |\psi^{(i)}(\vec{r}, t)|^2 , \tag{17}$$

which may be written as

$$\rho(\vec{r}, t) = \rho_g(\vec{r}) + \Delta\rho(\vec{r}, t) , \tag{18}$$

where $\rho_g(\vec{r})$ is the electron density in the absence of light, and to first order in $\vec{E}$ we have

$$\Delta\rho(\vec{r}, t) = \vec{E}(\vec{r}, t) \cdot \vec{R}(\vec{r}) , \tag{19}$$

where

$$\vec{R}(\vec{r}) = \hbar^{-1} \sum_{ij} \frac{2\omega_j \vec{p}_j}{\omega_j^2 - \omega_L^2} \varphi_j^{(i)}(\vec{r}) \varphi_g^{(i)}(\vec{r}) . \tag{20}$$

We note that

$$\int_{uc} \vec{R}(\vec{r})\, d^3r = 0 , \tag{21}$$

where uc denotes integration over a unit cell, because φ_j and φ_g are orthogonal wave functions. Also we have

$$\int_{uc} \vec{r}\, \vec{R}(\vec{r})\, d^3r = (1/eN)\overleftrightarrow{\chi} , \tag{22}$$

where e is the electronic charge, N is the number of unit cells/cm^3, and $\overleftrightarrow{\chi}$ is the optical susceptibility tensor.

The vector $\vec{R}(\vec{r})$ may be expanded in a reciprocal crystalline lattice Fourier sum

$$\vec{R}(\vec{r}) = \sum_{\vec{G}} \vec{R}_{\vec{G}} e^{i\vec{G}\cdot\vec{r}} , \tag{23}$$

where the vectors $\vec{G}$ are all the reciprocal lattice vectors. The induced charge $\Delta\rho(\vec{r}, t)$ has Fourier components of the form

$$\rho_{\vec{G}} \simeq e^{\mp i\vec{q}_L\cdot\vec{r}} e^{\mp i\omega_L t} e^{i\vec{G}\cdot\vec{r}} \rho_L(\pm q_L + \vec{G}) + \text{c.c.} , \tag{24}$$

where $\vec{G}$ is one of the reciprocal lattice vectors, and

$$\rho_L(\pm\vec{q}_L + \vec{G}) = \vec{R}_{\vec{G}} \cdot \vec{E}_L . \tag{25}$$

The total induced charge $\Delta\rho(\vec{r}, t)$ is the sum of such expresssions with various $\vec{G}$.

In the nearly forward direction, with $\vec{G} = 0$, this equation specifies zero mixing efficiency, because $\vec{R}_{\vec{G}=0} = 0$ through the general relation

$$\vec{R}_{\vec{G}} = N \int_{uc} \vec{R}(\vec{r})\, e^{-i\vec{G}\cdot\vec{r}}\, d^3r \tag{26}$$

and Eq. (21). However, there is small mixing efficiency through the charge prescribed by the macroscopic Maxwell relation

$$e\rho = (1/4\pi)\nabla \cdot \vec{E} , \tag{27}$$

which for this case, involving light, reads

$$e\rho = (i/4\pi)\, \vec{q}_L \cdot \vec{E} . \tag{28}$$

Thus only birefringent media can contribute to Eq. (23) in the $\vec{G} = 0$ case, and effectively

$$\vec{R}_{\vec{G}=0} = (i/4\pi e)\vec{q}_L . \tag{29}$$

The reason for the previous result $\vec{R}_{\vec{G}=0} = 0$ is the use of the dipole approximation in deriving Eq. (16). In Eq. (16) the electric field is evaluated at position $\vec{r}$, which in the dipole approximation is not distinguished from $\vec{r}_i$, the atomic site. If one does distinguish between $\vec{r}$ and $\vec{r}_i$ in Eq. (16), then a nonzero result, namely Eq. (28), is obtained.

III. PHASE-MATCHING CONDITIONS

The output intensity of the sum or difference x rays is peaked when the energy and momentum conservation conditions are obeyed[8]:

$$\omega_0 = \omega_I \pm \omega_L , \tag{30}$$

$$\vec{k}_0 = \vec{k}_I + \vec{G} \pm \vec{q}_L , \tag{31}$$

as evidenced by Eq. (8). It is almost always possible for nonzero $\vec{G}$ to arrange for the above equations to be satisfied merely by changing the direction of $\vec{G}$. Because $\omega_L \ll \omega_I$, and $|\vec{q}_L| \ll |\vec{k}_I|$, the angles between vectors $\vec{k}_I$, $\vec{k}_0$, and $\vec{G}$ are nearly the same as the angles involved in the ordinary Bragg-scattering condition, where $\vec{q}_L = \omega_L = 0$ in the above equations.

For some applications, and certainly in measurements of light-induced charge distribution, it would be undesirable to have the output sum or difference frequency collinear with the scattered Bragg beam which is at the input x-ray frequency. Towards this consideration, assume that $\vec{q}_L$ is coplanar with $\vec{k}_I$ and $\vec{G}$, so that $\vec{k}_0$ is also in the same plane. Let the input light beam travel at an angle B from the input x-ray beam $\vec{k}_I$. We shall calculate to first order in ω_L/ω_0 the angle that $\vec{G}$ should be turned to phase match, and the resultant angle between $\vec{k}_0$ and a Bragg-scattered direction.

From the above momentum-energy conserving conditions, using $\omega_I = k_I c$, $\omega_0 = k_0 c$, and $\omega_L = q_L c/n$, we have

$$\vec{G} \cdot (\vec{G} + 2\vec{k}_I) = (q_L^2/n^2) \pm 2k_I(q_L/n) \mp 2\vec{q}_L \cdot (\vec{k}_I + \vec{G}) , \tag{32}$$

where n is the light refractive index. The ordinary Bragg condition is $\vec{G} \cdot (\vec{G} + 2\vec{k}_I) = 0$, which defines θ_B through $\vec{G} \cdot \vec{k}_I = -Gk_I \sin\theta_B$.

In the light-mixing case, let $\delta\theta$ be defined by

$$\vec{G} \cdot \vec{k}_I = -Gk_I \sin(\theta_B + \delta\theta) , \tag{33}$$

where $\vec{G}$ is assumed set for the matching condition. Positive $\delta\theta$ corresponds to $\vec{G}$ being more antiparallel to $\vec{k}_I$, since $0 < \theta_B < \frac{1}{2}\pi$. To first order in ω_L/ω_I

and q_L/G we have

$$\delta\theta = \pm (q_L/G\cos\theta_B)[\cos(2\theta_B + B) - 1/n] , \quad (34)$$

where the sign of B is determined by the statement that some value $0 < B < \frac{1}{2}\pi$ allows $\vec{q}_L$ to be antiparallel to $\vec{G}$. The maximum $|\delta\theta|$ occurs when $B = -\pi + 2\theta_B$. For this value of B we have

$$\delta\theta = \mp (q_L/G\cos\theta_B)(1 + 1/n) . \quad (35)$$

If instead $\vec{k}_I$ and $\vec{q}_L$ are chosen to be collinear, then

$$\delta\theta = \pm (q_L/G\cos\theta_B)(\cos 2\theta_B - 1/n) , \quad (36)$$

if $\vec{k}_I$ and $\vec{q}$ travel in the same sense of direction, and

$$\delta\theta = \mp (q_L/G\cos\theta_B)(\cos 2\theta_B + 1/n) , \quad (37)$$

if the beams travel in opposite directions. The sign of $\cos 2\theta_B$ determines which case results in a larger $|\delta\theta|$.

The sum or difference frequency x rays travel in a direction $2\theta_B + \delta\theta'$ from the input $\vec{k}_I$, where

$$\delta\theta' = \mp (q_L/k_I\cos\theta_B)[\sin\theta_B/n + \sin(\theta_B + B)] . \quad (38)$$

The maximum $|\delta\theta'|$ occurs when $B = \frac{1}{2}\pi - \theta_B$, i.e., when the light travels nearly perpendicular to G. In general, the optimum angle B is dependent on particular experimental designs and intentions.

The range of angles in the input that can be mixed by a perfect crystal is determined by

$$\Delta k < \tfrac{1}{2}(\alpha_T + \alpha_0) , \quad (39)$$

because larger Δk results in small S given by Eq. (12). If the range of Δk in the input is primarily determined by the finite source collimation, then $\Delta k = k_I \Delta\Phi \sin 2\theta_B$. Equation (39) then determines a small angle $\Delta\Phi$ of order 10^{-9} rad, for values $\alpha_1 = \alpha_0 = \frac{1}{2}$, $\sin 2\theta_B = 0.5$, and $k_I = 5\times 10^8$. Thus, we feel justified in assuming that, with today's x-ray sources, the input x-ray beam will be collimated in the Bragg plane to an angle larger than $\Delta\Phi$. Under such conditions, the "integrated intensity,"[9] defined by $S_I = \int S(\Delta k)\, d\Phi$, where S is given by Eq. (12) is of interest. Using $\Delta k = k_I \sin 2\theta_B d\Phi$, it follows that

$$S_I = \frac{\lambda_0^3 r_0^2}{2(\alpha_T + \alpha_0)} \frac{1 + \cos^2\varphi}{\sin 2\theta_B} \frac{\cos\beta_I}{\cos\beta_0} (\vec{R}_{\vec{G}} \cdot \hat{\epsilon}_L)^2 . \quad (40)$$

If $\alpha_T + \alpha_0$ is dominated by x-ray absorption varying[5] as λ_0^3, then S_I decreases slowly as λ_0^{-1}. The number of output mixed x-ray photons N_0 is given by[9]

$$N_0 = S_I N_I , \quad (41)$$

where N_I is the number of input x-ray photons/rad. The collimation perpendicular to the Bragg plane is assumed to be consistent with the phase-matching condition. If the crystal is not perfect, Eq. (41) still applies,[9] under the condition that the input collimation is sufficiently broad to include the variation in direction of the reciprocal lattice vectors.

In contrast to nonzero $\vec{G}$ cases, measurement of the light-induced charge density when $\vec{G} = 0$, given by Eq. (28), will yield only information obtainable from measurements made purely in the optical regime. Mixing in the forward direction may be enhanced by processes other than considered here,[3] but the intensity of such scattering is small compared to that utilizing nonzero $\vec{G}$.

IV. MIXING EFFICIENCIES

In previous formulas, the quantity not known precisely is the induced charge density

$$\rho_L(\pm\vec{q} + \vec{G}) = \vec{R}_{\vec{G}} \cdot \vec{E}_L . \quad (42)$$

One may calculate a microscopic charge ρ_m through the use of Maxwell's microscopic relation

$$\rho_m = [\nabla \cdot (\overleftrightarrow{\chi}_m \cdot \vec{E}_m)]/e , \quad (43)$$

where $\overleftrightarrow{\chi}_m$ is the microscopic material linear susceptibility, and $\vec{E}_m$ is the microscopic field. On a microscopic scale both $\overleftrightarrow{\chi}_m$ and $\vec{E}_m$ fluctuate over atomic diameters. Since we are interested in the Fourier component of ρ_m at wave vector $\vec{G}$, the gradient operator in effect multiplies $\chi_m E_m$ by G. Thus one may write

$$|\vec{R}_{\vec{G}}| = sG\chi/e , \quad (44)$$

where s, generally a fourth-rank tensor, is a dimensionless quantity, whose elements vary from very small values to roughly 10^{-1}. In fact, a comparison of the formula for $\vec{R}_{\vec{G}}$, and the usual formula for optical linear susceptibility reveals that

$$s = \left| \frac{\sum_j [\rho_j \omega_j/(\omega_j^2 - \omega_L^2)] \int_{uc} d^3r \varphi_g(\vec{r}) e^{-i\vec{G}\cdot\vec{r}} \varphi_j(\vec{r})}{\sum_j [\rho_j \omega_j/(\omega_j^2 - \omega_L^2)] \int_{uc} d^3r \varphi_g(\vec{r})(Gr_k)\varphi_j(\vec{r})} \right| , \quad (45)$$

where p_j in Eq. (45) refers to the component of $\vec{p}_j$ parallel to the light electric field, and r_k is the component of r parallel to the macroscopic induced polarization. In the limit of small $\vec{G}$, s formally reduces to the cosine of the angle between $\vec{G}$ and the induced macroscopic polarization. In practice, the available vectors $\vec{G}$ are, of course, determined by the crystalline structure, and the smallest vector $\vec{G}$ is often approximately equal to an inverse atomic radius. The numerator in Eq. (45) is, therefore, typically smaller than the denominator. This compares with the x-ray Bragg-scattering result that outer electrons do not contribute much to the Bragg-scattered intensity. Consequently, values of s should be expected to range from 10^{-1} to 10^{-2} to nearly 0. If the states involved in the optical transitions are states of definite parity, then,

if the electric field and the vector $\vec{G}$ are perpendicular, then $s=0$. Generally, s is smaller than usual under such conditions.

To estimate a value of s, consider solid hydrogen, taken to be in a simple cubic lattice structure. The dominant transition is taken to be the $1s-2p$ transition and the wave functions those of the hydrogen atom. The light is polarized along $\vec{G}$. The size of G is taken to correspond to the density of liquid hydrogen, 0.07 gm/cm^3, so that $G=2.2\times10^8$ cm^{-1}.

For arbitrary G, Eq. (45), upon the use of standard hydrogen wave functions, in this case reduces to

$$s=1/[1+(\tfrac{2}{3}Ga_0)^2]^3\,, \tag{46}$$

where $a_0=0.51\times10^{-8}$ cm is the Bohr radius. The above value of G, therefore, results in $s\cong0.27$. It is important to notice that if G or a_0 had been chosen to be twice the above value, then a value of $s\cong0.03$ would have been obtained. Since the mixing efficiency goes as s^2 and the time to do an experiment for a given signal-to-noise goes as s^4, the change of Ga_0 by a factor of 10 would have increased the time to do an experiment by 10^4. The critical dependence on Ga_0 of mixing efficiency will, however, allow fairly accurate efficiency measurements to yield considerably more accurate measurements of effective radii similar to a_0.

Equation (45) predicts zero mixing efficiency under certain arrangements of input angles and polarizations. For example, by varying the light polarization, one can go through a zero of mixing efficiency.

Choosing values of $\lambda_0=10^{-8}$ cm, a large value of $\alpha_T+\alpha_0=1$ cm^{-1}, $G=\frac{1}{2}\pi\times10^8$, $\chi=1/4\pi$, and a possibly pessimistic value $s=0.05$, then $S_I=N_0/N_I=2.5\times10^{-21}$, if a light input of 1 W/cm^2 is used to uniformly illuminate the x-ray spot region. A conventional x-ray tube dissipating 2 kW emits about 2×10^{14} characteristic photons/sec into a hemisphere, so $N_I\approx1.5\times10^{12}$/rad sec if the beam is collimated to 1° in the direction perpendicular to the scattering plane. For an input laser power of 100 W spread over a 0.01 cm^2 x-ray spot size, then the number of scattered photons N_0 should be approximately 4×10^{-5}/sec. This number can be increased, for example, through the use of a higher brightness x-ray source. However, the signal rate to scattered x-ray background rate is also determined by the degree of perfection of available crystals, and a greater scattering efficiency would be desirable.

By use of a high-power pulsed laser source, emitting light during a total time T each second, the quantity S_I and therefore the ratio of output mixed photons to background scattered photons may be increased during the time of the laser pulses. By not observing scattered background x-ray radiation when the laser pulse is off, one increases the signal count rate to background count rate. In this way, one may obtain greatly increased signal/noise ratios as long as the average light power is not severely reduced.

The number of mixed x-ray photons, under such conditions, is proportional to the input x-ray flux during the time of the laser light pulse. The rest of the x rays are wasted. Hence, in addition to using a pulsed laser and gaining a factor $1/\sqrt{T}$ in signal/noise, one should use a pulsed x-ray source, and gain a similar factor $1/T$ in net output signal. For example, under the previously assumed conditions, with an average laser output of 1 W and an average x-ray tube electrical input of 20 W, with $T=10^{-8}$ sec, then $N_0=0.4$/sec.

According to Eqs. (12) and (45), a large mixing efficiency may occur in a perfect crystal at the phase-matched condition $\Delta k=0$ under ideal and special conditions. If x-ray lasers provide a sufficient flux/cm^2 rad of x rays, then light x-ray mixing can be used to efficiently tune the x-ray laser output.

CONCLUSIONS

Especially under short-pulsed conditions, optical x-ray photon mixing should be an observable effect, and potentially useful for investigating microscopic light-excited charge densities. X-ray lasers will extend the potential of such investigations, and the process itself may be used to tune x-ray laser outputs. The formalism exemplified here can be extended to other related effects, such as down-converting two x-ray beams to form optical photons, or mixing of two optical photons with one x-ray photon.

A comparison between the treatment given here for x-ray light scattering and the various terms in the nonlinear polarization of Armstrong *et al.*[3] reveals that there are other types of matrix elements not considered here which may become important in special circumstances. At low x-ray energies, the electron-photon cross section becomes modified because the electrons are bound. At high x-ray energies, relativistic effects should be considered.

This type of treatment may be extended to describe the interaction of other probes which interact with the outer electrons of optically driven material. For example, if instead of an x-ray cross section r_0^2, the electron-electron cross section was used, the calculations above would describe electron beam energy up- and down-shifting by electron-Bragg scattering from an optically-induced

charge wave.

It has come to our attention that Freund and Levine have performed a related calculation.[10]

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Electromagnetic Penetration and Confinement of a Hot Dense Plasma

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For a special model, it is possible to compute the self-consistent confinement and penetration of an energetic plasma by an rf field of large amplitude for arbitrary values of the quantity $(v/c)(\omega_p/\omega)$, where v is a characteristic electron thermal speed, ω_p is the plasma frequency deep in the plasma, and ω $(<\omega_p)$ is the rf frequency. Previous theories concerning nonlinear behavior of electromagnetic waves in plasmas have required this quantity to be small compared to unity.

I. INTRODUCTION

Previous theories of propagation of large-amplitude electromagnetic waves in plasmas[1–3] are based on assumptions that become invalid when the electromagnetic frequency is much less than the plasma frequency, $\omega \ll \omega_p$ (an overdense plasma). These assumptions are

$$\text{(i)}\ vB/c \ll E\ ,\ \text{(Refs. 1–3)} \qquad \text{(ii)}\ w \gg (1/T)\ ,\ \text{(Ref. 1)} \tag{1}$$

where v, B, and E are representative values of particle velocity, and magnetic and electric fields, and T is the characteristic time during which a representative particle samples the scale length of the fields. The second assumption is clearly necessary for the validity of the multiple time-scale method, commonly employed in theories of rf confinement.[3] Also, it is clearly necessary for replacing the Vlasov equation by local (moment) equations in describing the particle dynamics.[3]

In contrast to the present work in which large amplitude circularly polarized waves are confining a semi-infinite plasma, Gibbons and Hartle[4] have considered a case in which large amplitude linearly polarized waves are propagating in an infinite plasma. The $\vec{v}\times\vec{B}$ forces are correctly included there, as they are in the present paper.

Both (i) and (ii) above can be expressed approximately by the following inequality, valid when $\omega \ll \omega_p$:

$$(v/c)(\omega_p/\omega) \ll 1\ . \tag{2}$$

For plasmas of thermonuclear interest (kilovolt energies, densities greater than 10^{13} cm^{-3}), this condition becomes violated for electromagnetic frequencies less than 10^{10} sec^{-1}.

The purpose of the present paper is to solve the problem of self-consistent rf confinement and penetration of a Vlasov plasma under conditions in which inequality (2) is violated. We consider here a simple model problem subject to the following restrictions:

(a) The incident transverse electromagnetic field is circularly polarized.

(b) The electrons have no incident energy transverse to the rf wave vector.

(c) The ions are too heavy to respond to the rf field.

(d) The ions are cold, and therefore adjust themselves so as to exactly cancel out the (time-independent) electron space charge. [Because of the