Resonant x-ray second-harmonic generation in atomic gases

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We explore the x-ray second-harmonic generation process induced by resonant two-photon absorption in systems with inversion symmetry. We show that this process becomes allowed in the x-ray region due to nondipole contributions. It is found that, although a plane-wave pump field generates only a longitudinal second-harmonic field, a Gaussian pump beam creates also a radially polarized transverse second-harmonic field which is stronger than the longitudinal one. Contrary to the longitudinal component, the transverse second-harmonic field with zero intensity on the axis of the pump beam can run in free space. Our theory is applied to Ar and Ne atomic vapors and predicts an energy conversion efficiency of x-ray second-harmonic generation of 3.2×10^{-11} and 1.3×10^{-12}, respectively.

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I. INTRODUCTION

Modern x-ray free-electron laser (XFEL) facilities can deliver high intensities as high as 10^{15}–10^{19} W/cm^2 making it possible to efficiently populate core-excited states and even create population inversion and x-ray lasing [1–7]. At these intensities, x-ray matter interaction becomes nonlinear creating room for studies of nonlinear effects, such as stimulated x-ray Raman scattering [3–5,8–10], pulse compression [3–5], x-ray four-wave mixing [3–5,11,12], and nonlinear wave mixing of x-ray and near-infrared beams [13]. Special attention was paid to the competition between stimulated x-ray emission and Auger decay [14–16]. Second-harmonic generation (SHG) is a nonlinear optical process of sum frequency generation which produces new photons with twice the frequency. SHG has traditionally been studied as an even-order nonlinear optical effect allowed in media without inversion symmetry [17] and is one of the best-understood nonlinear effects in optics [18]. In light of the XFEL development, its study in the Angstrom regime, e.g., on the natural scale of atomic and molecular structure of matter, has become of great interest both from a fundamental and a practical viewpoint. A pioneering study by Shwartz *et al.* [19] and Yudovich and Shwartz [20] gave recently experimental evidence for "off-resonant" SHG in diamond in the hard x-ray region with a pump frequency of 7.3 keV.

In the present paper, we show that, due to the large momentum of the photon **k**, the nonlinearity in the x-ray region is different from conventional nonlinearities in the visible regime and that SHG is generally possible to observe for centrosymmetric systems even when phase-matching conditions do not prevail. We present a theoretical study of x-ray SHG in atomic gases induced by resonant two-photon absorption (TPA). We show that the plane-wave pump field can create only longitudinally polarized x-ray SH fields which cannot propagate in free space (see, however, Ref. [21]), but also that a Gaussian pump pulse induces in addition transverse SH fields which contrary to the longitudinal component can run in free space. Our idea is, in a certain sense, inverse to the use of the longitudinal component of focused light beams in laser-driven particle accelerators [22]. Another important feature of the SHG problem studied here is that the transverse field, being strictly equal to zero on the beam axis, has an unusual radial polarization.

Our paper is organized as follows. We outline, in Sec. II A, the basic theory of SHG using plane-wave pump radiation which generates only the longitudinal field. Then, in the following Sec. II B, we show that a Gaussian pump field creates also the transversely polarized SH field. Section II C is devoted to the analysis of the longitudinally and radially polarized SH fields. We shed light on the role of the absorption of an SH field in Sec. II D. Some theoretical details can be found in Appendices A–C. We discuss our results further in Sec. III where we numerically analyze the efficiency of SHG

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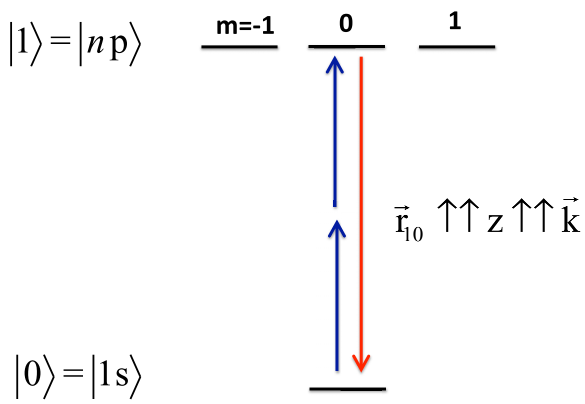

FIG. 1. The dipole moment $\mathbf{r}_{10}$ of the $1s \to np$ transition in the atom is parallel to $\mathbf{k}$. The axis of quantization z is along the photon momentum $\mathbf{k}$.

in Ne and Ar atomic vapors. Finally, in Sec. IV, we come to the conclusions.

II. THEORY

Quantum mechanically, the second-order nonlinearity in the optical susceptibility originates from a perturbational solution of the Schrödinger's equation. To get insight into the physics of the SHG process in the x-ray region, we consider the propagation of a x-ray pump field $\mathbf{E}_p$ in an atomic gas far away from the absorption edge. To induce the SHG we choose twice the frequency of the pump field to be resonant with the frequency of a two-photon transition $2\omega \approx \omega_{10}$. The scheme of SHG is shown in Fig. 1 where the pump field resonantly promotes the $1s$ electron of an atom to the np unoccupied level. The resonant population of state $|1\rangle$ in the course of TPA is followed by the emission of the SH field $\mathbf{E}$. Let us start from the atom-field interaction, which reads as [we use Système International (SI) units],

$$V = V^{(1)} + V^{(2)} = -\frac{e}{2mc}(\mathbf{p}\cdot\boldsymbol{\mathcal{A}}_p + \boldsymbol{\mathcal{A}}_p\cdot\mathbf{p}) + \frac{e^2}{2mc^2}\boldsymbol{\mathcal{A}}_p^2, \tag{1}$$

where m and e are the mass and charge of the electron, respectively. c is the speed of light, and $\mathbf{p}$ is the operator of electronic momentum. Below, we will use more frequently the electric field instead of the vector potential $\boldsymbol{\mathcal{E}} = -\partial\boldsymbol{\mathcal{A}}/\partial t$. The square of the vector potential of the pump field $\boldsymbol{\mathcal{A}}_p^2$ describes the TPA process in the first order of perturbation theory whereas the scalar product $\mathbf{p}\cdot\boldsymbol{\mathcal{A}}_p$ contributes to the TPA in second order of perturbation theory.

A. Plane-wave pump field

It is instructive to consider first the interaction with the simplest and most fundamental electromagnetic wave, the transverse plane-wave $\boldsymbol{\mathcal{A}}_p = (\mathbf{A}_p/2)\exp[-\imath(\omega t - \mathbf{k}\cdot\mathbf{r} - \mathbf{k}\cdot\mathbf{r}^{(e)})] + \text{c.c.}$,

$$\begin{aligned}\boldsymbol{\mathcal{E}}_p &= \frac{1}{2}\mathbf{E}_p e^{-\imath(\omega t - \mathbf{k}\cdot\mathbf{r} - \mathbf{k}\cdot\mathbf{r}^{(e)})} + \text{c.c.},\\ \mathbf{E}_p &= \mathbf{e}E_p^{(0)} = -\frac{\partial\mathbf{A}_p}{\partial t} = \imath\omega\mathbf{A}_p,\end{aligned} \tag{2}$$

with the polarization $\mathbf{e}$ being orthogonal to the photon momentum $\mathbf{k}$. Here, $\mathbf{r}^{(e)}$ is the coordinate of the electron with respect to the atom, and $\mathbf{r}$ is the radius vector of the atom in the laboratory frame. To avoid unnecessary complexity (see also below) we will focus only on the $\mathbf{A}_p^2$ term assuming that the wavelength of the photon is longer than the size of the core orbital $ka < 1$,

$$\begin{aligned}V^{(2)} &= \frac{e^2}{8mc^2}A^2 e^{-\imath 2(\omega t - \mathbf{k}\cdot\mathbf{r}^{(e)} - \mathbf{k}\cdot\mathbf{r})} + \text{c.c.} + \text{const}\\ &\approx \frac{e^2}{8mc^2}A_p^{(0)2} e^{-\imath 2(\omega t - \mathbf{k}\cdot\mathbf{r})}(1 + \imath 2\mathbf{k}\cdot\mathbf{r}^{(e)}) + \text{c.c.} + \text{const}.\end{aligned} \tag{3}$$

The term $A_p^{(0)2}$ being independent of the electron radius vector $\mathbf{r}^{(e)}$ cannot induce transitions between electronic states. Thus, the transition between the ground (s) and the core-excited (p) states is induced solely by the matrix element,

$$V_{10}^{(2)} \approx -\imath\frac{eE_p^{(0)2}}{4mc^2\omega^2}(\mathbf{k}\cdot\mathbf{d}_{10})e^{-\imath[(2\omega-\omega_{10})t - 2\mathbf{k}\cdot\mathbf{r}]}, \tag{4}$$

of the second term $\mathbf{k}\cdot\mathbf{r}^{(e)}$ on the right-hand side of Eq. (3). The rotating-wave approximation is used here by keeping only the near-resonant term. This pure nondipole process opens the $s \to p$ TPA channel with the transition dipole moment $\mathbf{d}_{01} = e\mathbf{r}_{01} = e\langle 0|\mathbf{r}^{(e)}|1\rangle$ (Fig. 1). We chose the axis z of quantization to lie along the photon momentum $\mathbf{k}$. In this frame, the pump field populates only the np_z level (see Fig. 1), and the problem is reduced to the interaction with a two-level atom with the transition dipole moment parallel to the photon momentum,

$$\mathbf{d}_{01} \parallel \mathbf{k}. \tag{5}$$

The resonant TPA population of the core-excited state of p symmetry is followed by the dipole allowed one-photon transition $p \to s$ which creates the SHG field with the double frequency 2ω. This explains why the SHG is possible in systems with inversion symmetry in the x-ray region.

To quantify the studied process, one should compute the polarization $\boldsymbol{\mathcal{P}}$. The induced macroscopic polarization of the medium being the expectation value of the dipole moment $\mathbf{d}$ is specified in terms of the density-matrix $\rho(t)$,

$$\boldsymbol{\mathcal{P}} = N\,\mathrm{Tr}(\mathbf{d}\rho) = N[\mathbf{d}_{01}(t)\rho_{10}(t) + \mathbf{d}_{10}(t)\rho_{01}(t)], \tag{6}$$

where N is the concentration of atoms and $\mathbf{d}_{01}(t) = \mathbf{d}_{01}\exp(\imath\omega_{10}t)$ is the dipole moment in the interaction representation [22,23]. The off-diagonal element of the density-matrix $\rho_{10}(t) = \varrho_{10}\exp[-\imath(\nu t - 2\mathbf{k}\cdot\mathbf{r})]$ satisfies the following kinetic equation in the interaction picture [23],

$$\left(\frac{\partial}{\partial t} + \Gamma - \imath\nu\right)\varrho_{10} = \frac{1}{\hbar m}\left(\frac{eE_p^{(0)}}{2\omega}\right)^2(\mathbf{k}\cdot\mathbf{r}_{10})(\rho_{11} - \rho_{00}), \tag{7}$$

where $\nu = 2\omega - \omega_{10}$ is the detuning from the two-photon resonance and Γ is the lifetime broadening of core-excited state $|1\rangle$. We neglect the very weak depopulation of the ground state in the course of the two-photon absorption ($\rho_{00} \approx 1,\ \rho_{11} \ll 1$) and assume that the duration τ of the pump pulse is longer than the lifetime of the core-excited state $1/\Gamma$. In this case, one can use the stationary solution of Eq. (7),

$$\varrho_{10} = \varrho_{01}^{*} = -\frac{1}{\hbar m}\left(\frac{eE_p^{(0)}}{2\omega}\right)^2 \frac{(\mathbf{k}\cdot\mathbf{r}_{10})}{\Gamma - \imath\nu} \tag{8}$$

to find the induced macroscopic polarization taking into account Eq. (5),

$$\boldsymbol{\mathcal{P}} = \mathbf{P}e^{-\imath 2(\omega t - \mathbf{k}\cdot\mathbf{r})} + \text{c.c.},$$
$$\mathbf{P} = \hat{\mathbf{k}}p, \quad p = -\left(\frac{eE_p^{(0)}}{2\omega}\right)^2 \frac{Nker_{01}^2}{m\hbar(\Gamma - \imath\nu)}. \tag{9}$$

Here, $\hat{\mathbf{k}} = \mathbf{k}/k$ is the unit vector along $\mathbf{k}$. One can see that the pump radiation creates a macroscopic polarization $\boldsymbol{\mathcal{P}}$ oriented along the direction of propagation of the pump field $\mathbf{k}$, and, hence, the SH field $\boldsymbol{\mathcal{E}}$, which is created in the course of the spontaneous transition $|1\rangle \to |0\rangle$, is also parallel to $\mathbf{k}$. This longitudinal field exists everywhere where there is pump field and the medium, and this field copies exactly the polarization according to Maxwell's equation for the induction $\nabla\cdot\boldsymbol{\mathcal{D}} = \partial(\epsilon_0\mathcal{E} + \mathcal{P})/\partial z = 0$:

$$\mathbf{E} = -\frac{2}{\epsilon_0}\mathbf{P} \neq 0, \quad \boldsymbol{\mathcal{D}} = 0, \quad \boldsymbol{\mathcal{H}} = 0. \tag{10}$$

This does not contradict the well-known fact that the plane-wave longitudinal field does not exist in free space [21]. This statement means that the longitudinal field cannot propagate in free space. The longitudinal field $\boldsymbol{\mathcal{E}}$ exists only in the region where the pump field creates longitudinal polarization $\boldsymbol{\mathcal{P}} \propto \mathbf{k}$. This longitudinal field oscillating in time and space is a pure electric-field $\boldsymbol{\mathcal{H}} = 0$.

As we have already noted above the TPA process is a first-order process with respect to $\boldsymbol{\mathcal{A}}_p^2$ and a second-order process with respect to $\mathbf{p}\cdot\boldsymbol{\mathcal{A}}_p$. Here, we study the two-photon transition $s \to p$, which is a pure nondipole effect. Since both $\boldsymbol{\mathcal{A}}_p^2$- and $\mathbf{p}\cdot\boldsymbol{\mathcal{A}}_p$-induced TPA result in the same orientation of the TPA-induced polarization we consider here only the $\boldsymbol{\mathcal{A}}_p^2$ contribution. The taking into account of the $\mathbf{p}\cdot\boldsymbol{\mathcal{A}}_p$ TPA process will only result in a rescaling of the SHG efficiency.

B. Gaussian pump beam and paraxial equation

In this section, we will show that a Gaussian pump beam,

$$\boldsymbol{\mathcal{E}}_p = \frac{1}{2}\mathbf{E}_p e^{-\imath(\omega t - kz)} + \text{c.c.},$$
$$\mathbf{E}_p = \hat{\mathbf{x}}E_p^{(0)} g\left(t - \frac{z}{c}\right)\frac{w_0}{w(z)}\exp\left(-\frac{\rho^2}{w^2(z)}\right)$$
$$\times \exp\left[\imath\left(k\frac{\rho^2}{2R(z)} - \psi(z)\right)\right] \tag{11}$$

makes it possible to transform the longitudinal SHG x-ray field into a transverse field which can propagate in free space in contrast to the pure longitudinal field. Equation (11) for a pulsed Gaussian beam is obtained in Appendix A by convoluting of the fundamental Gaussian mode with the Gaussian distribution of spectral components. As shown in Appendix A, $\mathbf{E}_p$ satisfies the paraxial equation. Equation (11) identifies $R(z) = z[1 + (z_R/z)^2]$ as the radius of curvature of the wave front of the beam at z, w_0 as the beam waist

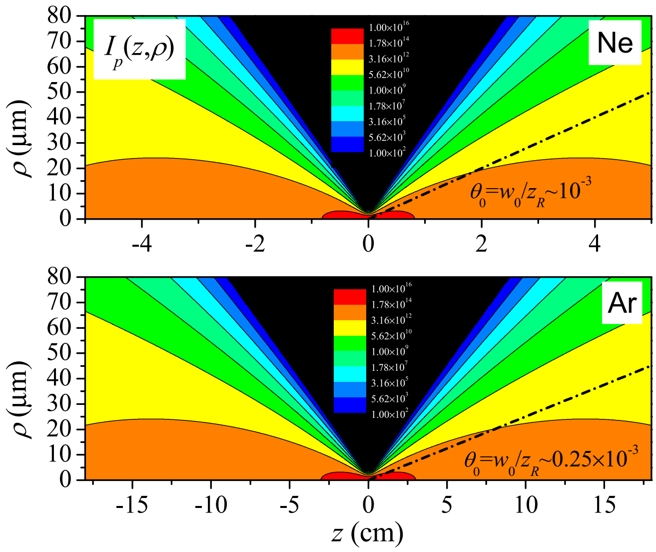

FIG. 2. Two-dimensional (2D) map of the pump intensity at $t = z/c$ for Ne and Ar atomic vapors. The legend shows the intensity in W/cm^2.

and $g(t) = \exp(-t^2/2\tau^2)$ as the temporal shape of the pulse with duration τ. Here, $w(z) = w_0\sqrt{1 + (z/z_R)^2}$, $\psi(z) = \arctan(z/z_R)$, $\rho = \sqrt{x^2 + y^2}$, $w_0/z_R \sim 1/kw_0 \ll 1$. The Gaussian beam remains well collimated up to the Rayleigh range $z_R = kw_0^2/2$ (Fig. 2).

Since the wave front is not orthogonal to z, as one can see from the phase $\phi = 2k(z + \rho^2/2R)$ of $\mathcal{E}_p^2 \propto \exp(\imath\phi)$, the polarization $\mathbf{P}$ is slightly tilted from the z axis. To find the matrix element $V_{01}^{(2)}$ of the interaction with the Gaussian pump beam (11), we need the value of this interaction at the point of the electron $\mathbf{r}^{(e)}$ with respect to the atom $\mathbf{r} = (\boldsymbol{\rho}, z)$, namely, at $\mathbf{r} + \mathbf{r}^{(e)}$,

$$\langle 0|e^{\imath(\phi+\delta\phi)}|1\rangle \approx e^{\imath\phi}\langle 0|1 + \imath\,\delta\phi|1\rangle = \boldsymbol{\kappa}\cdot\langle 0|\mathbf{r}^{(e)}|1\rangle e^{\imath\phi}\imath 2k, \tag{12}$$

where we used the Taylor expansion $\phi(\mathbf{r} + \mathbf{r}^{(\mathbf{e})}) = \phi(\mathbf{r}) + \delta\phi$ with $\delta\phi = \nabla\phi\cdot\mathbf{r}^{(e)}$. Similar to the derivation of Eq. (9), one obtains a polarization that is oriented along $\nabla\phi \equiv (\partial_z\phi, \partial_\rho\phi) = 2k\boldsymbol{\kappa}$,

$$\mathbf{P} = -\boldsymbol{\kappa}\left(\frac{eE_p}{2\omega}\right)^2 \frac{Nker_{01}^2}{m\hbar(\Gamma - \imath\nu)}, \quad \boldsymbol{\kappa} = \hat{\mathbf{z}} + \hat{\boldsymbol{\rho}}\frac{\rho}{R}, \tag{13}$$

instead of the beam axis $\hat{\mathbf{z}} \parallel \mathbf{k}$.

Let us write the optical wave equation for the SHG field $\boldsymbol{\mathcal{E}}$,

$$\nabla(\nabla\cdot\boldsymbol{\mathcal{E}}) - \nabla^2\boldsymbol{\mathcal{E}} + \frac{1}{c^2}\frac{\partial^2\boldsymbol{\mathcal{E}}}{\partial t^2} = -\mu_0\frac{\partial^2\boldsymbol{\mathcal{P}}}{\partial t^2}, \tag{14}$$

in the usual manner [18] starting from the couple of Maxwell's equations (in SI units) for nonmagnetic materials ($\mu = 1$),

$$\nabla\times\boldsymbol{\mathcal{E}} = -\mu_0\frac{\partial\boldsymbol{\mathcal{H}}}{\partial t}, \quad \nabla\times\boldsymbol{\mathcal{H}} = \frac{\partial\boldsymbol{\mathcal{D}}}{\partial t}. \tag{15}$$

Contrary to conventional theories [18] where $\nabla\cdot\boldsymbol{\mathcal{E}} \sim (\mathbf{k}\cdot\mathbf{e}) = \mathbf{0}$ for the transverse electro-magnetic-field ($\mathbf{k}\perp\mathbf{e}$), we cannot ignore $\nabla\cdot\boldsymbol{\mathcal{E}}$ here. This is because the polarization $\boldsymbol{\mathcal{P}}$

is essentially a longitudinal one [see Eq. (13)]: $\nabla \cdot \boldsymbol{\mathcal{P}} \neq 0$. To resolve this problem, we use Maxwell's equation for the induction $\boldsymbol{\mathcal{D}} = \varepsilon_0 \boldsymbol{\mathcal{E}} + \boldsymbol{\mathcal{P}}$,

$$\nabla \cdot \boldsymbol{\mathcal{D}} = 0, \quad \nabla \cdot \boldsymbol{\mathcal{E}} = -\frac{1}{\epsilon_0} \nabla \cdot \boldsymbol{\mathcal{P}}, \tag{16}$$

which makes it possible to rewrite the wave equation (14) as follows:

$$\begin{aligned} -\nabla^2 \boldsymbol{\mathcal{E}} + \frac{1}{c^2}\frac{\partial^2 \boldsymbol{\mathcal{E}}}{\partial t^2} &= \frac{1}{\epsilon_0}\left(-\frac{1}{c^2}\frac{\partial^2 \boldsymbol{\mathcal{P}}}{\partial t^2} + \nabla(\nabla \cdot \boldsymbol{\mathcal{P}})\right), \\ \boldsymbol{\mathcal{E}} &= \frac{1}{2}\mathbf{E} e^{-\imath 2(\omega t - kz)} + \text{c.c.}, \\ \boldsymbol{\mathcal{P}} &= \mathbf{P} e^{-\imath 2(\omega t - kz)} + \text{c.c.} \end{aligned} \tag{17}$$

This wave equation differs from the conventional one [18] by the extra term $\nabla(\nabla \cdot \boldsymbol{\mathcal{P}}) \neq 0$ which is not equal to zero because of the longitudinal contribution in $\boldsymbol{\mathcal{P}}$. We would like to point out that when the pump field is a plane wave, there is only a longitudinal SH field $\boldsymbol{\mathcal{E}} \parallel \hat{\mathbf{z}}$ (see Sec. II A). In this case, the wave equation (14) becomes $\partial^2(\epsilon_0 \boldsymbol{\mathcal{E}} + \boldsymbol{\mathcal{P}})/\partial t^2 = 0$, which is very different from Eq. (17) because $\nabla(\nabla \cdot \boldsymbol{\mathcal{E}}) - \nabla^2 \boldsymbol{\mathcal{E}} = (\partial^2/\partial z^2 - \partial^2/\partial z^2)\boldsymbol{\mathcal{E}} \equiv 0$.

Now, we are at the stage to simplify the wave equation (17). In our case, the wave propagates primarily along the z axis with a small divergence angle (Fig. 2),

$$\theta_0 \approx \frac{1}{2kw_0} = \frac{w_0}{z_R} \sim \frac{\lambda}{w_0} \ll 1. \tag{18}$$

Here, λ is the wavelength of the pump field. We assume also that the pulse duration τ is much longer than the period of the field oscillations $2\pi/\omega$. This makes it possible to neglect $\partial^2\mathbf{E}/\partial z^2$ and $\partial^2\mathbf{E}/\partial t^2$ in Eq. (17) (see Ref. [24]) and to get the following paraxial equation for the SHG field,

$$\left(\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t} - \frac{\imath}{4k}\Delta_\perp\right)\mathbf{E} = \frac{\imath}{2k}\mathbf{f}, \quad kw_0 \gg 1, \quad \tau\omega \gg 1, \tag{19}$$

where $\Delta_\perp = \nabla_\rho^2 = \partial^2/\partial x^2 + \partial^2/\partial y^2$ is the Laplacian operator over the transverse Cartesian coordinates. The source term on the right-hand side of the paraxial equation has now both longitudinal (f_z) and transverse components (f_ρ),

$$\begin{aligned} \tilde{\mathbf{f}} &= \hat{\mathbf{z}}\tilde{f}_z + \hat{\boldsymbol{\rho}}\tilde{f}_\rho, \\ \tilde{\mathbf{f}} &= \frac{1}{\epsilon_0}\left(-\frac{1}{c^2}\frac{\partial^2 \tilde{\mathbf{P}}}{\partial t^2} + \nabla(\nabla \cdot \tilde{\mathbf{P}})\right) \approx \frac{1}{\epsilon_0}[(2k)^2\tilde{\mathbf{P}} + \nabla(\nabla \cdot \tilde{\mathbf{P}})], \\ \tilde{\mathbf{P}} &= \mathbf{P}e^{-\imath 2(\omega t - kz)}, \quad \tilde{\mathbf{f}} = \mathbf{f}e^{-\imath 2(\omega t - kz)} \end{aligned} \tag{20}$$

Taking into account Eqs. (11), (13), and (18), one can get the following expression for the transverse and longitudinal components of $\mathbf{f}$:

$$\begin{aligned} f_\rho &= -\frac{\imath 8k\rho}{\epsilon_0 w^2}P, \\ f_z &= \frac{\imath 2kP}{\epsilon_0(z^2 + z_R^2)}\left[\frac{2k\rho^2 z_R(\imath z_R + z)}{z^2 + z_R^2} - \imath 4z_R - z\right]. \end{aligned} \tag{21}$$

One should point out that the origin of f_ρ is the term $\nabla(\nabla \cdot \tilde{\mathbf{P}}) = \hat{\boldsymbol{\rho}}\,\partial_\rho(\partial_z \tilde{P}) + \cdots$. A simple estimation shows that the transverse contribution dominates: $|f_\rho/f_z| \sim kw_0 \gg 1$. As one can see from the paraxial equation (19) the transverse and longitudinal components of $\mathbf{f}$ generate, respectively, the transverse and longitudinal components of the SH field $\mathbf{E}$.

C. Spatial distribution of the transverse and longitudinal SH fields. Radial polarization

It is convenient to write the solution of the paraxial equation (19) in terms of the retarded Green's function (see Appendix B),

$$\begin{aligned} &\mathbf{E}(z, \boldsymbol{\rho}, t) = \frac{1}{2\pi}\int G(z - z', \boldsymbol{\rho} - \boldsymbol{\rho}', t - t')\mathbf{f}(z', \boldsymbol{\rho}', t')dz'd\boldsymbol{\rho}'dt', \\ &\left(\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t} - \frac{\imath}{4k}\Delta_\perp\right)G(z - z', \boldsymbol{\rho} - \boldsymbol{\rho}', t - t') \\ &\qquad = \delta(z - z')\delta(\boldsymbol{\rho} - \boldsymbol{\rho}')\delta(t - t')\Theta(t - t'), \\ &G(z - z', \boldsymbol{\rho} - \boldsymbol{\rho}', t - t') \\ &\qquad = -\imath\delta\left(t' - t - \frac{z' - z}{c}\right)\Theta(t - t')\frac{k}{\pi(z - z')} \\ &\qquad\quad \times \exp\left(\imath\frac{k|\boldsymbol{\rho} - \boldsymbol{\rho}'|^2}{z - z'}\right), \end{aligned} \tag{22}$$

which guarantees that no contribution at remotely early times t before the source $\tilde{\mathbf{f}}(z', \boldsymbol{\rho}', t') = \mathbf{f}(z', \boldsymbol{\rho}', t')\exp[-\imath 2(\omega t' - kz')]$ has been activated. Taking into account that $\hat{\mathbf{z}}' = \hat{\mathbf{z}}$, $\hat{\boldsymbol{\rho}}' = \hat{\boldsymbol{\rho}}\cos\varphi + \hat{\mathbf{y}}\sin\varphi$, $\hat{\mathbf{y}} \perp \hat{\boldsymbol{\rho}}$, one can perform an integration over directions of $\boldsymbol{\rho}'$ on the plane $(\hat{\boldsymbol{\rho}}, \hat{\mathbf{y}})$ orthogonal to the z axis using Eq. (B5),

$$\begin{aligned} &\int_0^{2\pi} d\varphi[\hat{\boldsymbol{\rho}}' f_\rho(z', \rho', t') + \hat{\mathbf{z}}f_z(z', \rho', t')]\exp\left(\imath\frac{k|\boldsymbol{\rho} - \boldsymbol{\rho}'|^2}{z - z'}\right) \\ &\quad = 2\pi\exp\left(\imath\frac{k(\rho^2 + \rho'^2)}{z - z'}\right) \\ &\qquad \times\left[-\hat{\boldsymbol{\rho}}\imath f_\rho(z', \rho', t')J_1\left(\frac{2k\rho\rho'}{z - z'}\right)\right. \\ &\qquad \left. + \hat{\mathbf{z}}f_z(z', \rho', t')J_0\left(\frac{2k\rho\rho'}{z - z'}\right)\right], \end{aligned} \tag{23}$$

where $J_n(x)$ is a Bessel function. One can obtain the remaining integral over ρ' using Eq. (B5) and get the following expressions for transverse and longitudinal contributions:

$$\begin{aligned} &\mathbf{E}(z, \boldsymbol{\rho}, t) = \hat{\boldsymbol{\rho}}E_\rho(z, \boldsymbol{\rho}, t) + \hat{\mathbf{z}}E_z(z, \boldsymbol{\rho}, t), \\ &E_i(z, \rho, t) = E_p^{(0)}g^2\left(t - \frac{z}{c}\right)J_i(z, \rho), \quad i = (\rho, z), \end{aligned} \tag{24}$$

where

$$\begin{aligned} J_\rho(z, \rho) &= -2\rho s_0\int_{-\infty}^{z} dz'\frac{e^\Phi}{w^4(z')\alpha^2(z')}, \\ J_z(z, \rho) &= \frac{\imath 4\pi w_0 s_0}{(kw_0)^3}\int_{-\infty}^{z} dz'\frac{e^\Phi}{w^4(z')\alpha(z')} \\ &\quad \times\left[\frac{2(\imath z_R + z')}{w^2(z')\alpha(z')}\left(z - z' - \frac{2k^2\rho^2}{\alpha(z')}\right) - (\imath 4z_R + z')\right], \\ \Phi &= \frac{\imath k\rho^2}{z - z'} - \imath 2\psi(z') - \frac{k^2\rho^2}{(z - z')\alpha(z')}, \end{aligned}$$

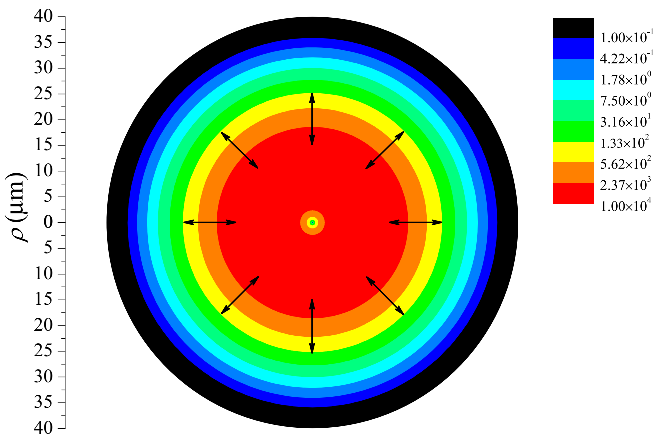

FIG. 3. Radial distribution of I_ρ for Ne atomic vapor at $z = 0.02$ m. Black arrows show schematically the radially polarized SHG field.

$$\alpha(z') = \frac{2(z-z')}{w^2(z')} - \imath k\left(\frac{z-z'}{R(z')}+1\right),$$

$$s_0 = 8\pi\frac{G}{\Gamma - \imath\nu} N z_R r_{01} r_e. \tag{25}$$

Here, $r_e = e^2/(4\pi\epsilon_0 mc^2) = 2.82\times 10^{-13}$ cm is the classical electron radius and $G = E_p^{(0)} d_{01}/\hbar$ is the Rabi frequency. It is important to note that there is no transverse field on the beam axis,

$$E_\rho(z, \rho = 0, t) = 0. \tag{26}$$

Equation (24) indicates that the transverse SH field $\hat{\boldsymbol{\rho}}E_\rho$ is oriented along the radius $\boldsymbol{\rho}$ perpendicular to the beam axis (Fig. 3). This means that the transverse field has radial polarization (see also Sec. III).

D. Role of photoabsorption

In the equations above, the photoabsorpion of x rays is ignored. This approximation is valid for the pump beam whose frequency is far from any resonance. In contrast, the SHG field is in strict resonance with the dipole allowed transition $|0\rangle \to |1\rangle$ ($1s-3p$ for Ne and $1s-4p$ for Ar). Therefore, this absorption channel should be taken into account. With the solution (24) at hand, we are almost prepared to include the photoabsorption in the SH field. As shown in Appendix C, the photoabsorption of the SHG field modifies only the integrands at the right-hand side of Eqs. (25) for $J_\rho(z,\rho)$ and $J_z(z,\rho)$. Namely, these integrands should be multiplied by the factor,

$$\exp\left(\frac{z'-z}{2\ell}\right), \tag{27}$$

where $\ell = 1/N\sigma_{\rm abs}$ is the photoabsorption length whereas $\sigma_{\rm abs}$ is the resonant photoabsorption cross section. According to simulations the photoabsorption length should be larger or comparable with the Rayleigh range,

$$\ell \sim z_R \tag{28}$$

to make it possible for the SHG field to reach the optimal value.

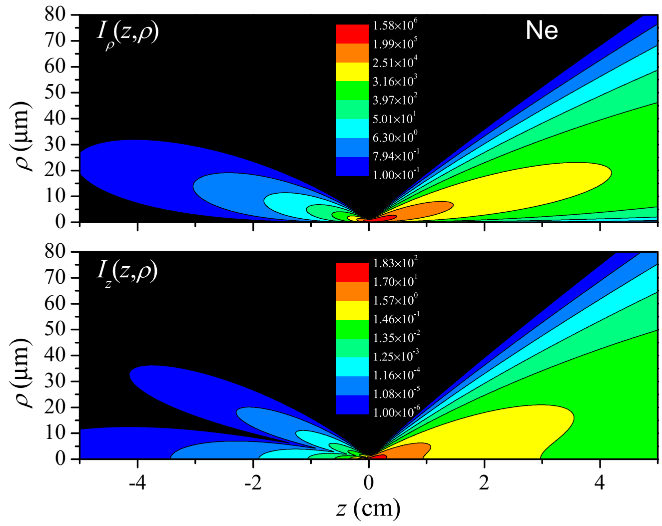

FIG. 4. Two-dimensional map of the SHG intensity at $t = z/c$ of Ne atomic vapor. The photoabsorption is negleted. The legend shows the intensity in W/cm^2.

III. RESULTS OF SIMULATIONS AND DISCUSSION

We applied the theory outlined above to two atomic systems Ne and Ar under the strict two-photon resonance ($2\omega = \omega_{10}$) with $1s \to 3p$ transitions for Ne and $1s \to 4p$ transitions for Ar. In the simulations, the peak pump intensity used was $I_p^{(0)} = c\epsilon_0|E_p^{(0)}|^2/2 = 10^{16}$ W/cm^2 (Fig. 2), and the following parameters were adopted for Ne: $\hbar\omega_{1s-3p} = 867.4$ eV, $\sigma_{\rm abs}(1s-3p) = 1.5\times 10^{-18}$ cm^2 [25], $2\hbar\Gamma = 0.27$ eV [26], and for Ar: $\hbar\omega_{1s-4p} = 3203.42$ eV, $\sigma_{\rm abs}(1s-4p) = 0.12\times 10^{-18}$ cm^2 [27], and $2\hbar\Gamma = 0.66$ eV [28]. The concentration of the atoms and the beam waist were equal to $N = 10^{19}$ cm^{-3} and $w_0 = 1.0$ μm, respectively. The Rayleigh range was $z_R \approx 10^3$ and $z_R \approx 4\times 10^3$ μm for Ne and Ar, respectively. The corresponding values of the photoabsorption lengths $\ell \approx 0.67\times 10^3$ and 8×10^3 μm satisfy the condition (28).

We solved the paraxial equation with homogeneous distribution of the concentration. The SHG radiation is characterized by the intensity distributions of the transverse $[I_\rho(z,\rho,t)]$ and longitudinal $[I_z(z,\rho,t)]$ components of the SH field (24),

$$I_i(z,\rho,t) = \tfrac{1}{2}c\epsilon_0|E_i(z,\rho,t)|^2, \quad i = (\rho, z), \tag{29}$$

and by the energy conversion efficiency,

$$\beta_i = \frac{W_i(z)}{W_p}, \quad W_i(z) = 2\pi\int_0^\infty dt \int_0^\infty d\rho\, I_i(z,\rho,t). \tag{30}$$

First we studied the SH field neglecting the photoabsorption. In Figs. 4 and 5, we display the spatial distribution of the SH intensities $I_\rho(z,\rho,t)$ and $I_z(z,\rho,t)$ for Ne and Ar, respectively. One can see that the transverse and longitudinal SH fields show very different radial structures with $I_\rho = 0$ on the axis of the beam $\rho = 0$, and the transverse field $\hat{\boldsymbol{\rho}}E_\rho$ (24) has an unusual radial polarization as shown in Fig. 3. The transverse SH field I_ρ is much stronger than the longitudinal one I_z, and the energy conversion efficiency of the transverse

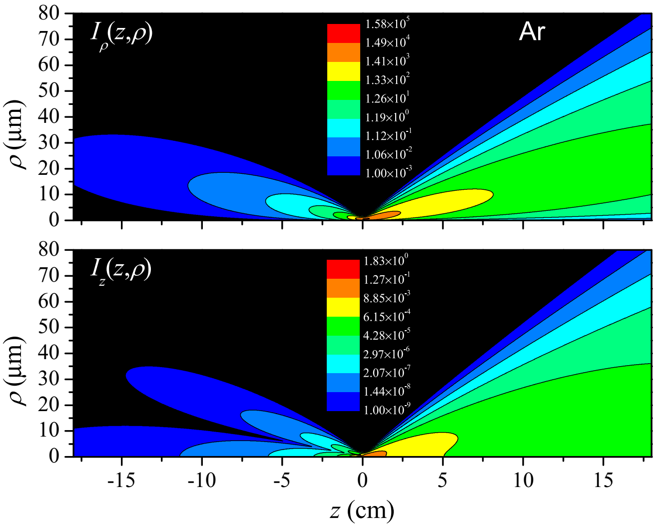

FIG. 5. Two-dimensional map of the SHG intensity at $t = z/c$ of Ar atomic vapor. The photoabsorption is neglected. The legend shows the intensity in W/cm^2.

SH field is about four to five orders of magnitude larger than that of the longitudinal SH field as shown in Fig. 6.

Because of the deeper ionization potential and smaller core-electron transition dipole moment, the conversion efficiencies of the x-ray SH fields from Ar atomic vapor are much smaller than those from Ne (Fig. 6). However, when the photoabsorption of the generated SH fields is considered, the final conversion efficiencies from Ar and Ne become comparable. Below, we investigated the conditions for the experimental observation of the SHG process with x rays in atomic Ne and Ar vapors by taking into account the resonant one-photon absorption of the SH field during propagation. The photoabsorption changes the spatial distribution of the SH field (Fig. 7) and reduces the energy conversion efficiency in one order of magnitude for Ne and in four times for Ar as one can see from Figs. 6 and 8. As is expected, the range of the SH field is limited by the photoabsorption length ℓ and is mainly confined in the focal region (see Fig. 7). Due to this circumstance, the energy conversion efficiency becomes maximal at $z = z_{\max} = 0.7$ mm for Ne and $z_{\max} = 0.5$ cm for Ar. This range defines the size of the gas cell which should be around $z_{\max}$.

IV. SUMMARY

In this paper, we investigated the second-harmonic generation in systems with inversion symmetry in the x-ray region.

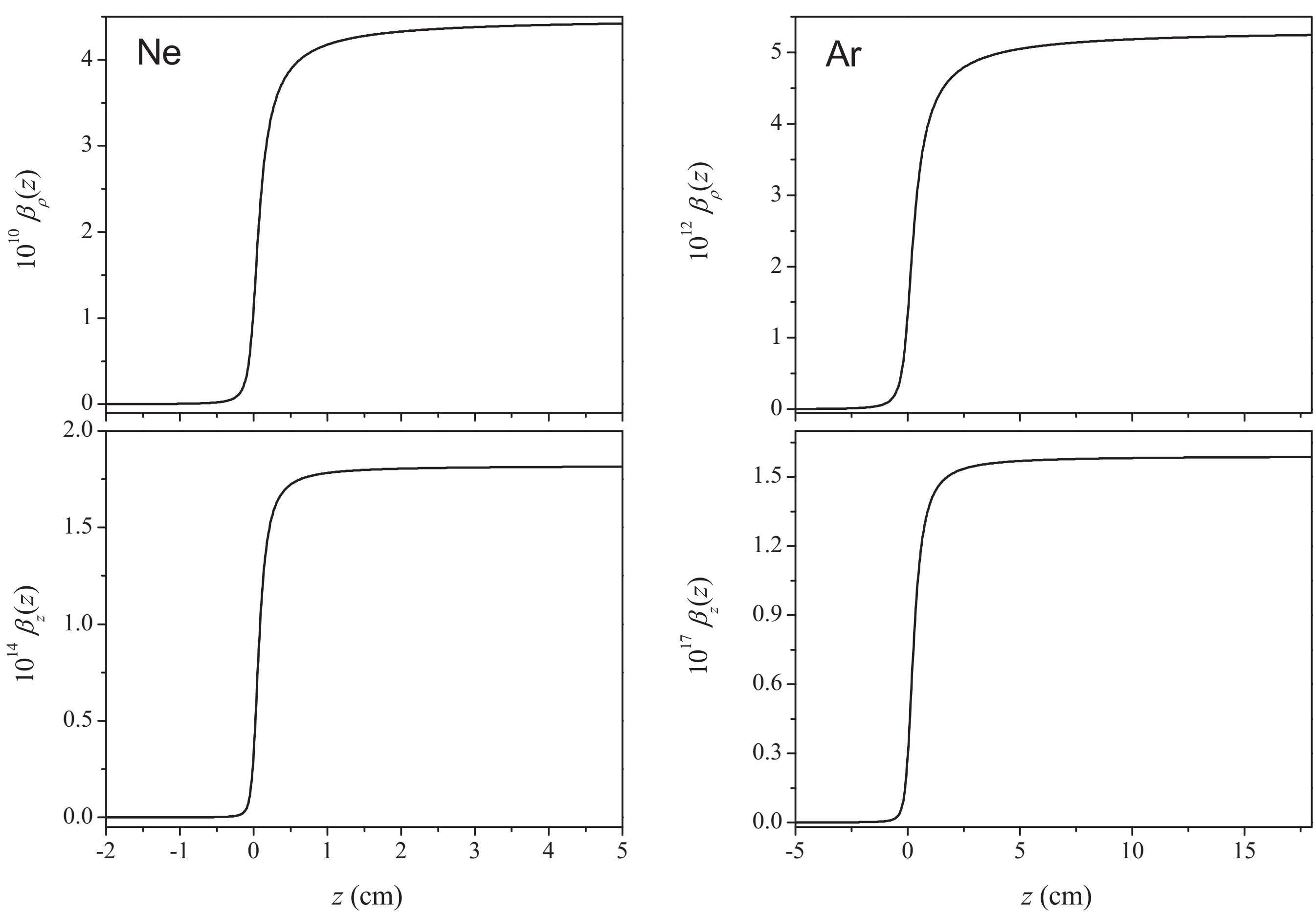

FIG. 6. Energy conversion efficiencies of the transverse SH fields $\beta_\rho(z)$ and the longitudinal SH fields $\beta_z(z)$ in Ne and Ar atomic vapors. The photoabsorption is neglected. The vertical axixes show the energy conversion efficiencies multiplied by the factor 10^n: For example, $10^{10}\beta_\rho$ and $10^{14}\beta_z$ for Ne.

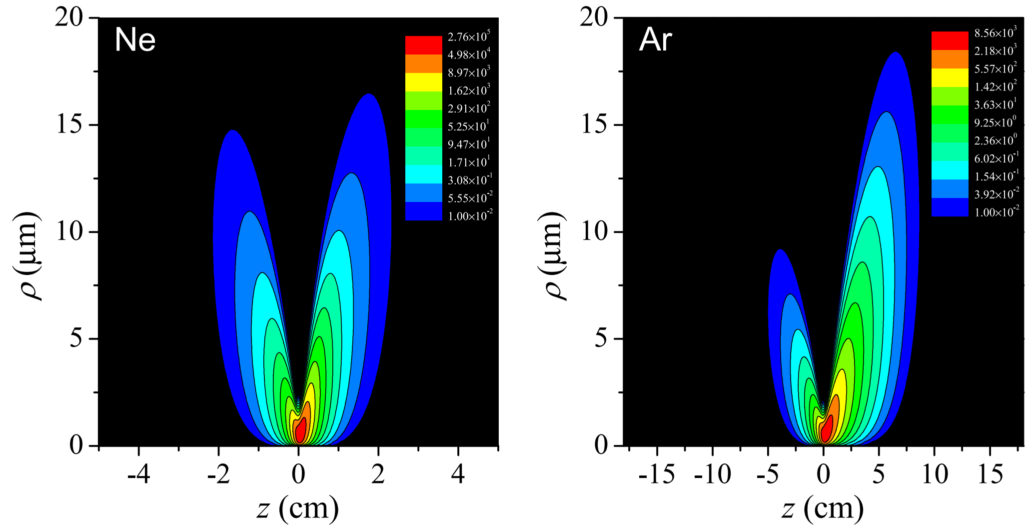

FIG. 7. Distribution of the transverse SH field I_ρ for Ne and Ar by taking into account the photoabsorption. The legend shows the intensity in $\mathrm{W/cm^2}$.

Our theory is applied to SHG in neon and argon pumped by a strong x-ray field tuned in resonance with the two-photon transition $1s \rightarrow 3p$ in Ne and $1s \rightarrow 4p$ in Ar. The nondipole population of these core-excited states is followed by the emission of the SH field. We describe the SHG in atoms in terms of a density matrix formalism and paraxial equation taking into account the resonant photoabsorption of the SH radiation. In contrast to the plane-wave pump field, the Gaussian pump beam generates transverse SH photons with radial polarization. By taking into account the x-ray photoabsorption effect, the energy conversion efficiencies to the transverse SH fields are expected to be orders of 10^{-11} and 10^{-12} in Ne (867.4 eV) and Ar (3203.4 eV) atomic vapors for the pump 10^{16} $\mathrm{W/cm^2}$, respectively.

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APPENDIX A: THE PULSED GAUSSIAN BEAM

To obtain the pulsed Gaussian beam (11) with the carrier frequency ω, we need to convolute the fundamental Gaussian

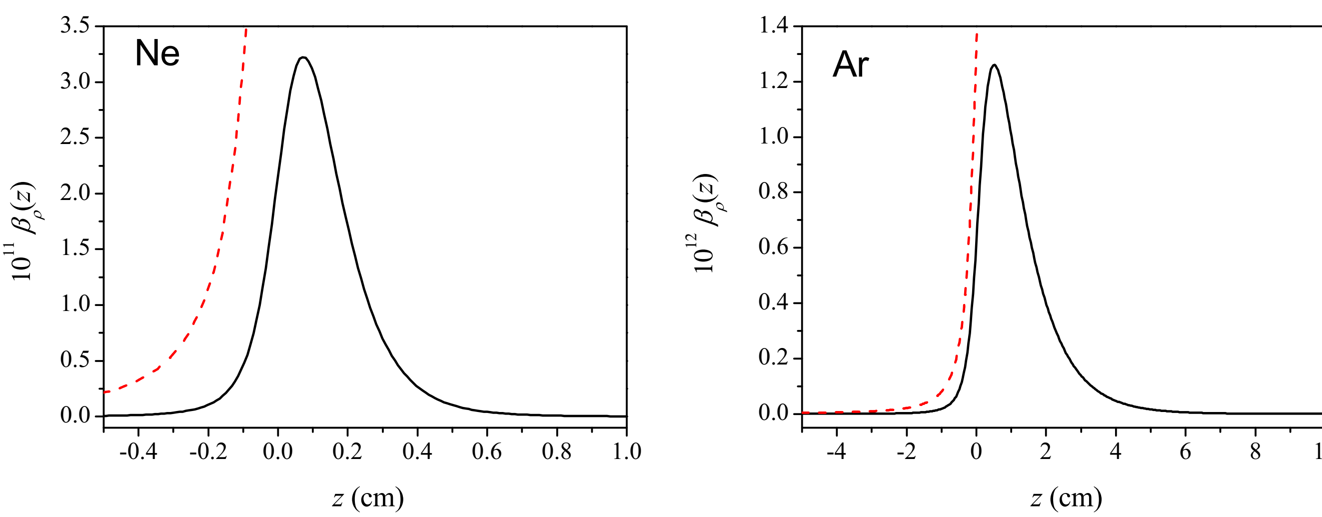

FIG. 8. Energy conversion efficiency $\beta_\rho(z)$ for Ne and Ar by taking into account the photoabsorption. The dashed lines display $\beta_\rho(z)$ calculated by neglecting the photoabsorption (see Fig. 6). Ne: $\beta_\rho^{\max} = 3.2 \times 10^{-11}$ at $z_{\max} = 0.7$ mm. Ar: $\beta_\rho^{\max} = 1.3 \times 10^{-12}$ at $z_{\max} = 0.5$ cm.

mode,

$$\boldsymbol{\mathcal{E}}(\omega,t)=\frac{1}{2}\mathbf{E}(\omega)e^{-\imath(\omega t-kz)}+\text{c.c.},\quad \mathbf{E}(\omega)=\hat{\mathbf{x}}E_p^{(0)}\frac{w_0}{w(z)}\exp\left(-\frac{\varrho^2}{w^2(z)}\right)\exp\left[\imath\left(k\frac{\varrho^2}{2R(z)}-\psi(z)\right)\right], \tag{A1}$$

with the spectral distribution $g(\omega'-\omega)=\exp[-(\omega'-\omega)^2\tau^2/2]/\tau\sqrt{2\pi}$ centered at the carrier frequency ω,

$$\boldsymbol{\mathcal{E}}_p(\omega,t)=\int_{-\infty}^{\infty}g(\omega'-\omega)\boldsymbol{\mathcal{E}}(\omega',t)d\omega'. \tag{A2}$$

The mode $\mathbf{E}(\omega)$ is the eigenfunction of the stationary paraxial or Helmholtz equation,

$$\left(\frac{\partial}{\partial z}-\frac{\imath}{2k}\Delta_\perp\right)\mathbf{E}(\omega)=0. \tag{A3}$$

The substitution of the fundamental mode (A1) in the convolution (A2) results in an expression,

$$\boldsymbol{\mathcal{E}}_p(\omega,t)=\frac{\hat{\mathbf{x}}}{2}E_p^{(0)}\int_{-\infty}^{\infty}d\omega' g(\omega'-\omega)\exp\left[-\imath\omega'\left(t-\frac{z}{c}-\frac{\rho^2}{2cR(z)}\right)\right]\frac{w_0}{w(z)}\exp\left(-\frac{\varrho^2}{w^2(z)}\right)e^{-\imath\psi(z)}+\text{c.c.} \tag{A4}$$

We neglect the ω' dependence of the Rayleigh range $z_R=k'w_0^2/2\approx kw_0^2/2$ because the variation of the frequency $\Delta\omega=|\omega'-\omega|\sim 1/\tau$ in the Fourier transform (A4) is negligibly small in comparison with the carrier frequency of the x-ray pulse: $\Delta\omega/\omega\sim 1/\tau\omega\ll 1$. Thus,

$$\boldsymbol{\mathcal{E}}_p(\omega,t)\approx\frac{\hat{\mathbf{x}}}{2}E_p^{(0)}\exp\left[-\imath\omega\left(t-\frac{z}{c}-\frac{\rho^2}{2cR(z)}\right)\right]\frac{w_0}{w(z)}\exp\left(-\frac{\rho^2}{w^2(z)}\right)e^{-\imath\psi(z)}g\left(t-\frac{z}{c}-\frac{\rho^2}{2cR(z)}\right)+\text{c.c.},\quad g(t)=\exp\left(-\frac{t^2}{2\tau^2}\right). \tag{A5}$$

Within the paraxial approximation [24] ($kw_0\gg 1$), we can neglect $\rho^2/2cR(z)$ in the Gaussian $g[t-z/c-\rho^2/2cR(z)]$ because

$$z\sim z_R\sim kw_0^2\gg\frac{\rho^2}{2R(z)}\sim\frac{w_0^2}{z_R}\sim\frac{1}{k},\quad \frac{\rho^2}{2cR(z)\tau}\sim\frac{1}{\tau\omega}\ll 1. \tag{A6}$$

However, we should keep $\rho^2/2cR(z)$ in the oscillatory term $\exp\{-\imath\omega[t-z/c-\rho^2/2cR(z)]\}$ because

$$\omega\frac{\rho^2}{2cR(z)}=\frac{k\rho^2}{2R(z)}\approx\frac{kw_0^2}{2z_R}\sim 1. \tag{A7}$$

Finally we get Eq. (11) for $\boldsymbol{\mathcal{E}}_p(\omega,t)$.

Now, we are in the stage to show that $\mathbf{E_p}$ from Eq. (11) satisfies the paraxial equation,

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}-\frac{\imath}{2k}\Delta_\perp\right)\mathbf{E_p}=0. \tag{A8}$$

Let us apply the operator $\Box=-\nabla^2+\partial^2/c^2\partial^2 t$ to both sides of Eq. (A2),

$$\Box\boldsymbol{\mathcal{E}}_p(\omega,t)=\int_{-\infty}^{\infty}g(\omega'-\omega)\Box\boldsymbol{\mathcal{E}}(\omega',t)d\omega'. \tag{A9}$$

Using the paraxial approximation, $kw_0\gg 1,\ \tau\omega\gg 1$ and Eq. (A3), we get Eq. (A8),

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}-\frac{\imath}{2k}\Delta_\perp\right)\mathbf{E_p}=e^{\imath(\omega t-kz)}\int_{-\infty}^{\infty}d\omega' g(\omega'-\omega)\frac{k'}{k}e^{-\imath(\omega' t-k'z)}\left(\frac{\partial}{\partial z}-\frac{\imath}{2k'}\Delta_\perp\right)\mathbf{E}(\omega')=0, \tag{A10}$$

which shows that the pulsed Gaussian beam (11) is an eigenfunction of the paraxial operator.

The paraxial approximation is broken when $kw_0\lesssim 1,\ \tau\omega\lesssim 1$. In this case, one should restore in Eq. (19) the second derivatives over z and time t: $\partial/\partial z\to\partial/\partial z-(\imath/4k)\partial^2/\partial z^2,\ \Delta_\perp\to\Delta_\perp-\partial^2/c^2\partial t^2$. However, such conditions are difficult to reach in the x-ray region. For example, the condition $\tau\omega\sim 1$ corresponds to a few cycle x-ray pulse where the pulse duration is comparable with the period of the field oscillations.

APPENDIX B: GREEN'S FUNCTION FOR THE TIME-DEPENDENT PARAXIAL EQUATION

Let us find the Green's function of the nonstationary paraxial equation,

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}-\frac{\imath}{2K}\Delta_\perp\right)G(z,\boldsymbol{\rho},t)=\Theta(t)\delta(t)\delta(z)\delta(\boldsymbol{\rho}), \tag{B1}$$

where $\Delta_\perp = \partial^2/\partial x^2 + \partial^2/\partial y^2$, $\boldsymbol{\rho} = (x, y)$, $\delta(\boldsymbol{\rho}) = \delta(x)\delta(y)$, and $\Theta(t)$ is the step function which is equal to zero when $t < 0$. Taking the Fourier transform of the Green's function and of the Dirac δ functions, we get

$$G(z, \boldsymbol{\rho}, t) = \frac{\Theta(t)}{(2\pi)^4}\int_{-\infty}^{\infty} d\mu \int_{-\infty}^{\infty} d\nu \int_{-\infty}^{\infty} dp \times \int_{-\infty}^{\infty} dq\, G_{\mu,\nu,p,q} e^{\imath\mu t+\imath\nu z+\imath px+\imath qy},$$

$$G_{\mu,\nu,p,q} = -\frac{2\imath}{\frac{\mu}{c} + \nu + \frac{p^2+q^2}{2K}}. \tag{B2}$$

Keeping in mind that $t \geqslant 0$ and taking the integral along the half circle in the upper half plane around the pole $\mu = -c(\nu + \frac{p^2+q^2}{2K})$,

$$\int_{-\infty}^{\infty} \frac{e^{\imath\mu t}}{\frac{\mu}{c}+\nu+\frac{p^2+q^2}{2K}} d\mu = \imath c\pi \, \exp\left[-\imath ct\left(\nu + \frac{p^2+q^2}{2K}\right)\right], \tag{B3}$$

we obtain the following expression for the Green's function:

$$G(z, \boldsymbol{\rho}, t) = -\imath\delta\left(t - \frac{z}{c}\right)\Theta(t)\frac{K}{2\pi z}\exp\left(\imath\frac{K\rho^2}{2z}\right), \tag{B4}$$

which allows to find the SHG field (22) with help of the following integrals [29]:

$$J_0(a) = \frac{1}{2\pi}\int_0^{2\pi} e^{\imath a\cos\theta} d\theta,$$

$$\frac{1}{2\pi}\int_0^{2\pi} \cos\theta e^{-\imath a\cos\theta} d\theta = \imath\frac{d}{da}J_0(a) = -\imath J_1(a),$$

$$\int_0^{\infty} e^{-a^2\rho^2}\rho^{n+1}J_n(b\rho)d\rho = \frac{b^n}{(2a^2)^{n+1}}e^{-(b^2/4a^2)}, \quad \mathrm{Re}(a^2) > 0. \tag{B5}$$

APPENDIX C: PHOTOABSORPTION OF THE SHG FIELD

The strongest absorption channel is the absorption of SH field which is in resonance with $|0\rangle \to |1\rangle$ transition. To take into account this photoabsorption, we need to add $-\mathbf{E}/2\ell$ at the right-hand side of paraxial equation (19),

$$\left(\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t} - \frac{\imath}{4k}\Delta_\perp\right)\mathbf{E} = -\frac{1}{2\ell}\mathbf{E} + \frac{\imath}{2k}\mathbf{f}, \tag{C1}$$

where $\ell = 1/\sigma N$ is the length of resonant absorption of the SHG field with the photoabsorption cross-section σ. Using the substitution $\mathbf{E} = \tilde{\mathbf{E}}\exp(-z/2\ell)$, one can see that $\tilde{\mathbf{E}}$ satisfies paraxial equation (19),

$$\left(\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t} - \frac{\imath}{4k}\Delta_\perp\right)\tilde{\mathbf{E}} = \frac{\imath}{2k}\mathbf{f}e^{z/2\ell}, \tag{C2}$$

with a modified source term. This equation has the solution given by Eq. (22) with $\mathbf{f}$ replaced by $\mathbf{f}\exp(z'/2\ell)$. Taking this into account, we get immediately the solution of paraxial equation with photoabsorption (C1),

$$\mathbf{E}(z, \boldsymbol{\rho}, t) = \tilde{\mathbf{E}}e^{-z/2\ell} = \frac{e^{-z/2\ell}}{2\pi}\int G(z-z', \boldsymbol{\rho}-\boldsymbol{\rho}', t-t') \times \mathbf{f}(z', \boldsymbol{\rho}', t')e^{z'/2\ell}dz'd\boldsymbol{\rho}'dt'. \tag{C3}$$

This means that, to include the photoabsorption, we should multiply by $\exp[(z'-z)/2\ell]$ the integrand at the right-hand side of Eqs. (25) for $J_\rho(z, \rho)$ and $J_z(z, \rho)$.

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