Harmonic generation and energy transport in dielectric and semiconductors at visible and UV wavelengths: the case of GaP

V. Roppo,1,2 N. Akozbek,3 D. de Ceglia,3 M. A. Vincenti,3 and M. Scalora2,*

1Universitat Politècnica de Catalunya, Departament de Física i Enginyeria Nuclear, Rambla Sant Nebridi, 08222 Terrassa, Spain
2Charles M. Bowden Research Center, RDECOM, Redstone Arsenal, Alabama 35898, USA
3Aegis Technologies Group, 410 Jan Davis Dr., Huntsville, Alabama 35806, USA
*Corresponding author: michael.scalora@us.army.mil

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We study the propagation and momentum transport of the inhomogeneous component of second and third harmonic pulses in dielectrics and semiconductors, at visible and UV wavelengths, focusing on materials like GaP. In these spectral regions GaP is characterized by large absorption, metallic behavior or a combination of both. We show that phase locking causes the generated inhomogeneous signals to propagate through a bulk metallic medium without being absorbed. This means that it occurs even in centrosymmetric materials thanks to the magnetic Lorentz force. We show that the transport of energy and momentum is quite peculiar and it can appear as anomalous, and that the direction of the Poynting vector of some of the harmonic pulses does not follow Snell’s law after crossing the interface. These results make it clear that there are new opportunities in ultrafast nonlinear optics and nanoplasmonics in new wavelength ranges. © 2011 Optical Society of America

1. INTRODUCTION

Harmonic generation is a nonlinear process that has also been used to generate new coherent light sources toward shorter wavelengths (for a recent review see [1]). Compact, coherent sources, particularly in the extreme ultraviolet (EUV) spectral region, have important applications ranging from nanotechnology and high resolution microscopy to spectroscopy [1]. Most nonlinear optical materials absorb strongly in the UV region (λ ≤ 400 nm). For this reason harmonic generation has been limited to visible wavelengths to avoid absorption losses. Absorption is generally considered detrimental because it can limit the conversion efficiency over the coherence length. In fact, one reasonably expects that any generated harmonic will be fully reabsorbed inside the medium if the sample is much longer than the characteristic absorption and coherence lengths. In contrast, it has been shown recently that even in the absorption or metallic regions of semiconductors it is possible to generate and propagate harmonic signals for distances that are much larger than the absorption length or skin depth, respectively, thanks to the onset of phase locking [2]. In this work we go a step further, and show that this phenomenon can take place not only in the presence of bulk nonlinearities (i.e., χ(2)) but also in centrosymmetric media thanks to the nonlinear action of the magnetic Lorentz force. In addition we will show that the direction of the individual momentum vectors of the generated harmonics can point in directions that do not necessarily obey Snell’s law. Before we delve into the details in what follows we provide a brief description of the previous results.

In transparent materials the general solution for second and third harmonic generation (SHG and THG) from a boundary layer consists of a reflected signal and two forward-propagating components. The latter displays k vectors that are solutions of the homogenous and inhomogeneous wave equations [2,4]. For example, Maker fringes result from the interference between the two forward-propagating components that at phase matching merge into a single solution [5]. Recently, complete phase and group velocity locking were demonstrated also for noncollinear propagation in a transparent LiNbO3 wafer, for different combinations of incident polarizations [6].

The generation and transmission of SH and TH wavelengths were recently demonstrated both theoretically and experimentally in bulk GaAs in a case where the harmonics were tuned well above the absorption band edge [7]. The phenomenon was attributed to phase locking between the fundamental and its generated inhomogeneous harmonics that occurs under phase mismatched conditions, irrespective of material parameters [8], as long as the material is somewhat transparent to the pump. The pump then impresses its dispersive properties to its harmonics, which in turn experience no absorption or other dephasing effects such as group velocity walk-off. The harmonics copropagate phase- and velocity-locked to the pump, as energy exchange ceases away from interfaces [8]. In cavity situations phase locking persists in a GaAs etalon [9] and in a GaAs defect layer surrounded by Bragg mirrors [10]. Even though the generated signals were tuned far below the absorption edge of GaAs (612 nm and 408 nm), conversion efficiencies were shown to improve by nearly four orders of magnitude compared to bulk GaAs. The generated inhomogeneous signals thus compete with the usual homogeneous signals that are much more abundant at the phase matching condition.
Perhaps more relevant for our purposes are the theoretical prediction and experimental verification that induced transparency occurs also in generally forbidden ranges, i.e., where the dielectric constant of materials displays metallic behavior (like GaP at 223 nm) [2]. In fact, the objective of this work is to perform a more detailed theoretical study of energy and momentum transport in GaP such that the harmonics are tuned in ranges that are either strongly absorptive or where the dielectric constant becomes negative, yielding metallic behavior. The interaction is generally phase mismatched and the recorded efficiencies were of the order of $10^{-9}$ and $10^{-12}$ for the SH and TH fields, respectively [2]. In both cases the homogeneous SH and TH signals vanish, leaving the inhomogeneous components unscathed. This concept is crucial to appreciate the dynamics that underlie the interaction. In [11] it was shown that the one dimensional field localization patterns change sensibly if the absorption of the material is turned on or off, because the generated signal is generally a superposition of homogeneous and inhomogeneous signals. In a two-dimensional environment the absence of absorption causes the momentum vector of a given harmonic to oscillate in direction and amplitude as energy flows back and forth between the two generated components until walk-off separates them. However, in absorptive or metallic environments, the momentum vector will be given only by the inhomogeneous component.

Absorption and momentum direction are two physical characteristics of matter-wave interaction that seldom come under scrutiny, especially in metals and semiconductors at visible and UV wavelengths. Bulk metals have their plasma frequencies in the UV-visible region due to their relatively large free carrier densities, and exhibit a negative permittivity from the visible all the way to microwave frequencies. Unlike bulk metals, however, semiconductors like GaAs, GaP, Si, and Ge exhibit a region of negative dielectric permittivity at deep UV wavelengths and are transparent in the infrared region. As we will show below, the fact that the momentum vector of the inhomogeneous components does not point in the expected direction suggests that it is possible to use momentum and energy flow as yet another degree of freedom exploitable in the design of nonlinear devices.

To date we have learned that anomalous (negative) refraction is generally associated with negative index materials ($\varepsilon < 0$ and $\mu < 0$) and occurs also for a TM-polarized field incident on a negative permittivity medium [12]. This has led to the development of a number of devices, such as tunable, high-transmission superlenses in the visible range using resonant metal/dielectric multilayer structures [13,14]. The same arrangement may be exploited in resonant multilayer stacks such as GaAs/KCl and GaAs/MgO operating in the deep UV spectral region [15]. These structures work with a single fundamental beam. However in this scenario it is possible to include the action of the anomalous momentum of the inhomogeneous component to shift the dynamics from the fundamental field to the harmonic frequencies.

This idea has already been pursued in a GaAs structure, where a GaAs grating is able to induce enhanced transmission of second harmonic generated light also at UV wavelengths [16]. These structures provide an alternative in the experimental realization of nonlinear optical and plasmonic phenomena in the UV-visible range since the fabrication of bulk, effective negative index materials has proven to be challenging due to absorption losses. Indeed absorption is still the major limiting factor for any practical applications, and various gain schemes have been considered. Semiconductors are used extensively in photonics and electronics industry and are easier to integrate with current electro-optical technologies. They can also be doped to control free carrier density and plasma frequency. In fact, doped semiconductors that exhibit a negative permittivity in the far IR region were shown to support negative refraction [17], plasmonic response [18], and superlensing in SiC [19]. In addition, semiconductors exhibit large nonlinear coefficients, an important factor for nonlinear frequency conversion and other types of parametric processes. It then becomes clear that knowledge of the behavior connected to the phase locking dynamics at UV and visible frequencies can lead to a number of new and unexpected results. The case of GaP is a ideal example because at the same time it allows us to investigate both high absorption and metallic regimes with a single propagation.

In short, the questions we address may be formulated as follows: What are the nonlinear sources that allow the phase locking mechanism? How does energy and momentum transport occur for harmonics tuned in high absorption and metallic (where $\varepsilon < 0, \mu = 1$) ranges?

## 2. THEORETICAL MODEL

In what follows we present the model we use to describe ultrashort pulse propagation phenomena that include SHG and THG in GaP (data from [20]). The study is completely generic and the same strategy can be applied to other dielectrics and/or semiconductors. We assume the medium is composed of a collection of doubly resonant Lorentz oscillators (the double resonance system is clearly visible in the linear dielectric functions shown in Fig. 1.)

With reference to Fig. 2, we assume that both TE- and TM-polarized fields may be present and are decomposed as a superposition of harmonics as follows:

$$E = \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} = \begin{pmatrix} i(E_{0x} e^{-i\omega_0 t} + (E_{TM}^0) e^{i\omega_0 t}) e^{-2i\omega t} + (E_{TM}^0) e^{-2i\omega t} + (E_{TM}^0) e^{-3i\omega t} + (E_{TM}^0) e^{-3i\omega t} \\ j(E_{TM}^0) e^{-i\omega_0 t} + (E_{TM}^0) e^{i\omega_0 t}) e^{-2i\omega t} + (E_{TM}^0) e^{-2i\omega t} + (E_{TM}^0) e^{-3i\omega t} + (E_{TM}^0) e^{-3i\omega t} \\ k(E_{TM}^0) e^{-i\omega_0 t} + (E_{TM}^0) e^{i\omega_0 t}) e^{-2i\omega t} + (E_{TM}^0) e^{-2i\omega t} + (E_{TM}^0) e^{-3i\omega t} + (E_{TM}^0) e^{-3i\omega t} \end{pmatrix}$$

$$H = \begin{pmatrix} H_x \\ H_y \\ H_z \end{pmatrix} = \begin{pmatrix} i(H_{TM}^0 e^{-i\omega_0 t} + (H_{TM}^0) e^{i\omega_0 t} + H_{TM}^0 e^{-2i\omega t} + (H_{TM}^0) e^{-2i\omega t} + H_{TM}^0 e^{-3i\omega t} + (H_{TM}^0) e^{-3i\omega t} \\ j(H_{TM}^0 e^{-i\omega_0 t} + (H_{TM}^0) e^{i\omega_0 t} + H_{TM}^0 e^{-2i\omega t} + (H_{TM}^0) e^{-2i\omega t} + H_{TM}^0 e^{-3i\omega t} + (H_{TM}^0) e^{-3i\omega t} \\ k(H_{TM}^0 e^{-i\omega_0 t} + (H_{TM}^0) e^{i\omega_0 t}) e^{-2i\omega t} + (H_{TM}^0) e^{-2i\omega t} + H_{TM}^0 e^{-3i\omega t} + (H_{TM}^0) e^{-3i\omega t} \end{pmatrix}.$$
Then, a TM-polarized field has a magnetic field component along \(x\) and electric fields that point along \(y\) and \(z\). A TE-polarized field has magnetic field components along the \(y\)- and \(z\)-directions, and a single electric field component that points in the \(x\)-direction. The second order polarization vector of GaP (or GaAs) may be written as follows [21]:

\[
\begin{align*}
\begin{pmatrix}
P_{NL,x}^{(2)} \\
P_{NL,y}^{(2)} \\
P_{NL,z}^{(2)}
\end{pmatrix}
&= 2d_{14} \begin{pmatrix}
E_y E_z \\
E_z E_x \\
E_x E_y
\end{pmatrix},
\end{align*}
\] (3)

where the coordinates correspond to the crystals’ principal axes. Substituting the \(E\)-field vector defined in Eq. (2) into Eq. (4) leads to the following equations:

\[
P_{NL,x}^{(2)} = 2d_{14} \begin{pmatrix}
(E_{TM,TE}^{(2)} + (E_{TM,TE}^{(2)})^*)E_{TM,TE}^{(2)} + E_{TM,TE}^{(2)}E_{TM,TE}^{(2)} & + (E_{TM,TE}^{(2)})^*E_{TM,TE}^{(2)}e^{-2i\omega t} + \\
(E_{TM,TE}^{(2)} + (E_{TM,TE}^{(2)})^*)E_{TM,TE}^{(2)} & + (E_{TM,TE}^{(2)})^*E_{TM,TE}^{(2)}e^{-2i\omega t} + \\
(E_{TM,TE}^{(2)} + (E_{TM,TE}^{(2)})^*)E_{TM,TE}^{(2)} & + (E_{TM,TE}^{(2)})^*E_{TM,TE}^{(2)}e^{-2i\omega t} + \\
\end{pmatrix}
\] (4)

\[
P_{NL,y}^{(2)} = 2d_{14} \begin{pmatrix}
(E_{TM,TE}^{(2)} + (E_{TM,TE}^{(2)})^*)E_{TM,TE}^{(2)} + E_{TM,TE}^{(2)}E_{TM,TE}^{(2)} & + (E_{TM,TE}^{(2)})^*E_{TM,TE}^{(2)}e^{-2i\omega t} + \\
(E_{TM,TE}^{(2)} + (E_{TM,TE}^{(2)})^*)E_{TM,TE}^{(2)} & + (E_{TM,TE}^{(2)})^*E_{TM,TE}^{(2)}e^{-2i\omega t} + \\
(E_{TM,TE}^{(2)} + (E_{TM,TE}^{(2)})^*)E_{TM,TE}^{(2)} & + (E_{TM,TE}^{(2)})^*E_{TM,TE}^{(2)}e^{-2i\omega t} + \\
\end{pmatrix}
\] (5)

\[
P_{NL,z}^{(2)} = 2d_{14} \begin{pmatrix}
(E_{TM,TE}^{(2)} + (E_{TM,TE}^{(2)})^*)E_{TM,TE}^{(2)} + E_{TM,TE}^{(2)}E_{TM,TE}^{(2)} & + (E_{TM,TE}^{(2)})^*E_{TM,TE}^{(2)}e^{-2i\omega t} + \\
(E_{TM,TE}^{(2)} + (E_{TM,TE}^{(2)})^*)E_{TM,TE}^{(2)} & + (E_{TM,TE}^{(2)})^*E_{TM,TE}^{(2)}e^{-2i\omega t} + \\
(E_{TM,TE}^{(2)} + (E_{TM,TE}^{(2)})^*)E_{TM,TE}^{(2)} & + (E_{TM,TE}^{(2)})^*E_{TM,TE}^{(2)}e^{-2i\omega t} + \\
\end{pmatrix}
\] (6)

From Eqs. (4)–(6) alone one may surmise that if a TM-polarized field were incident on GaP it would generate a TE-polarized SH signal via the term \(E_{TM,TE}^{(2)} \sim i(E_{TM,TE}^{(2)} - E_{TM,TE}^{(2)})e^{-2i\omega t}\) in Eq. (5). In turn, together with the TM-polarized pump, a non-zero \(E_{TM,TE}^{(2)}\) triggers TM-polarized, cascaded THG via the terms: \(E_{TM,TE}^{(2)} \sim j(E_{TM,TE}^{(2)} - E_{TM,TE}^{(2)})e^{-3i\omega t}\) and \(E_{TM,TE}^{(2)} \sim k(E_{TM,TE}^{(2)} - E_{TM,TE}^{(2)})e^{-3i\omega t}\) in Eqs. (5) and (6), respectively. Finally, some photons are returned to the original TM-polarized pump via a down-conversion process by the terms: \(E_{TM,TE}^{(2)} \sim j(E_{TM,TE}^{(2)} - E_{TM,TE}^{(2)})e^{-i\omega t}\) and \(E_{TM,TE}^{(2)} \sim k(E_{TM,TE}^{(2)} - E_{TM,TE}^{(2)})e^{-i\omega t}\) for the SH wave.

Since a TM-polarized SH component is notably absent from the predictions made using the \(\chi^{(2)}\) of Eq. (3), in the absence of other sources this portion of the SH signal must originate in surface and volume nonlinear phenomena ordinarly associated with centrosymmetric materials [10,22]. Like all materials, GaP is also characterized by surface and volume nonlinear sources arising from symmetry breaking and from the magnetic portion of the Lorentz force, independently of its bulk \(\chi^{(2)}\) and \(\chi^{(3)}\) properties. These contributions are taken into account by deriving equations of motion for the components of the polarization of bound electrons beginning from a classical Lorentz oscillator model [15,16,22]:

\[
\begin{align*}
\dot{P}_{b,2\omega} + \gamma_{b,2\omega} P_{b,2\omega} + \tilde{a}_{0}^{2} \gamma_{b,2\omega} P_{b,2\omega} = & \frac{n_{0,2\omega} e^{2} k_{0}^{2}}{m_{b} c^{2}} E_{2\omega} \\
+ & \frac{e \lambda_{0}}{m_{b} c^{2}} \begin{pmatrix}
E_{w} \cdot P_{b,2\omega} + \\
-\frac{i}{3} E_{w} \cdot P_{b,2\omega} + \\
-3 E_{w} \cdot P_{b,2\omega}
\end{pmatrix} \\
+ & \frac{\lambda_{0}}{m_{b} c^{2}} \begin{pmatrix}
(P_{b,2\omega} - i a_{0} P_{b,2\omega} \times H_{w}) + \\
(P_{b,2\omega} + i a_{0} P_{b,2\omega} \times H_{w}) + \\
(P_{b,2\omega} - 3 a_{0} P_{b,2\omega} \times H_{w})
\end{pmatrix}.
\end{align*}
\] (8)

\[
\begin{align*}
\dot{P}_{b,3\omega} + \gamma_{b,3\omega} P_{b,3\omega} + \tilde{a}_{0}^{3} \gamma_{b,3\omega} P_{b,3\omega} = & \frac{n_{0,3\omega} e^{2} k_{0}^{2}}{m_{b} c^{2}} E_{3\omega} \\
+ & \frac{e \lambda_{0}}{m_{b} c^{2}} \begin{pmatrix}
\frac{1}{2} E_{w} \cdot P_{b,2\omega} + \\
E_{w} \cdot P_{b,2\omega} + \\
E_{w} \cdot P_{b,2\omega}
\end{pmatrix} \\
+ & \frac{\lambda_{0}}{m_{b} c^{2}} \begin{pmatrix}
(P_{b,2\omega} - 2 i a_{0} P_{b,2\omega} \times H_{w}) + \\
(P_{b,2\omega} - a_{0} P_{b,2\omega} \times H_{w}) + \\
(P_{b,2\omega} - i a_{0} P_{b,2\omega} \times H_{w})
\end{pmatrix}.
\end{align*}
\] (9)
Fig. 1. (Color online) Dielectric constant of GaP (left) and GaAs (right), taken from [22]. The regions of negative dielectric constant (~100 nm–250 nm) are denoted by the shaded areas.

\[ \tilde{\epsilon}_{b,N\omega} = (\tilde{\epsilon}_b - N\omega), \quad \tilde{\alpha}_{b,N\omega}^2 = (\omega_{b}^2 - (N\omega)^2 + i\gamma_{b,N\omega}) \]

where \( N \) is an integer that denotes the given harmonic order; \( \lambda_b = 1 \mu m \) is the reference wavelength; \( \tilde{P}_{b,N\omega} \) is the polarization envelope of the \( N \)th harmonic; \( c \) is the speed of light in vacuum; the subscript \( b \) stands for bound; \( e \) is the electron charge; \( m_b^* \) is the effective mass of bound electrons. Nonlinear source terms in Eqs. (7)–(9) (bound charges in GaP via quadrupole-like contributions proportional to \( \nabla \cdot \tilde{P}_{b,N\omega} \) and magnetic terms like \( \tilde{P}_{b,N\omega} \cdot (\omega_{b}\tilde{B}_b) \times \nabla H_{b} \) give rise to TM-polarized SH and TH fields. With this in mind another look at Eqs. (4)–(6) reveals that the TM-polarized pump and its harmonics serve as nonlinear sources for all TE-polarized fields, including the pump [15,16,22]. The production of a TE-polarized pump field initiates with the introduction of either symmetry breaking (i.e., \( \nabla \cdot \tilde{P}_{b,N\omega} \), surface terms) or the Lorentz force. Once TE-polarized fields are generated, all interaction channels become active and the generation of all harmonic fields takes place. We should note that this is a peculiarity of the \( \chi^{(3)} \) tensor of GaP and other materials that have the same symmetry properties [21]. For further details about the model and the integration scheme employed we direct the reader to [15,16,22].

The inclusion of third order phenomena begins with the general expansion of the third order polarization as follows [21]:

\[ P_{NL,i}^{(3)} = \sum_{j,k,l=1,3} \sum_{\lambda=1,3} \sum_{x,y,z} \chi_{j,k,l}^{\lambda(i)} E_j E_k E_l \quad j,k,l = x,y,z. \] (10)

GaP has cubic symmetry of the type 3m, so that Eq. (5) reduces to:

\[ P_{NL,x}^{(3)} = \chi_{xxx}^{(3)} E_3 + 3\chi_{xyy}^{(3)} E_x E_y + 3\chi_{yzz}^{(3)} E_y E_z \]

\[ P_{NL,y}^{(3)} = \chi_{yy}^{(3)} E_3 + 3\chi_{xzy}^{(3)} E_x E_z + 3\chi_{yz}^{(3)} E_y E_z \]

\[ P_{NL,z}^{(3)} = \chi_{zz}^{(3)} E_3 + 3\chi_{xzx}^{(3)} E_x E_z + 3\chi_{yx}^{(3)} E_y E_x, \] (11)

Substituting Eqs. (1) and (2) into Eqs. (11) leads to \( \chi^{(3)} \) contributions to all harmonic components, with self- and cross-phase modulation along with terms that couple orthogonal polarization states. In particular, the presence of nonzero second harmonic fields provides a way for \( \chi^{(3)} \) to also contribute to the SHG process. Finally, the linear and nonlinear polarization components at each frequency are gathered and combined into Maxwell’s equations.

3. NUMERICAL RESULTS

We have already seen in the previous section that important results can be deduced by analyzing the equations of motion to deduce the possible energy fluxes that can be activated among the fundamental and the generated harmonics. Further details about the dynamics may be ascertained by numerically solving Maxwell’s system of equations coupled to the nonlinear polarizations exemplified by Eqs. (4)–(11). In our examples we use a pulse approximately 30 fs in duration tuned to 670 nm propagating across an air-GaP interface [10]. The SH and TH are thus tuned at 335 nm and 223 nm, respectively. The dispersion values of the permittivity at the fundamental, SH and TH are taken as \( \epsilon_{P} = 11.12, \quad \epsilon_{SI} = 11.96 + i24, \) and \( \epsilon_{TM} = 12.8 + i9.73, \) respectively.

In Fig. 3 we report a snapshot of the dynamics as the incident TM-polarized pulse generates TE- and TM-polarized harmonics that in turn generate down-converted TE-polarized pump photons in the manner previously described. The media file in Fig. 3 shows that all the generated pulses are locked to the pump during the entire process and propagate in the same direction, including the TM-polarized SH field. By observing the color scale of each harmonic and comparing it to the TM-polarized pump scale one can ascertain the approximate conversion efficiency of a given harmonic. The figure suggests that the generation of TE-polarized pump photons is similar to TM-polarized THG and several orders of magnitude larger than TE-polarized THG. The process is nevertheless more difficult to observe because one needs a large degree of discrimination between the intense TM-polarized pump and the much weaker TE-polarized signal. What makes this process unusual, unique, and perhaps exploitable for new types of applications is that phase locking occurs even in the absence of a nonlinear bulk coefficient, via the magnetic Lorentz force, as evidenced by the action of the TM-polarized SH signal. This is confirmed by performing the simulation with second and third order nonlinear coefficients set to zero. In that case, only TM-polarized fields are generated in a manner nearly identical to that of Fig. 3. Suffice it to say for the moment that SHG in centrosymmetric dielectric materials like silicon, for example, may yield unexpected benefits even in cavity environments because phase locking persists with the mere presence of the Lorentz magnetic force, as our calculations show.

In Fig. 4 we summarize the results by highlighting the directions of the individual light momenta for the pump and each of the generated fields. Figure 4(a) corresponds to the situation of Fig. 3. In Fig. 4(b) the simulation is repeated for an incident field tuned to 1200 nm, yielding comparable results. A comparison of
the propagation event in the media file in Fig. 3 and the scheme in Fig. 4(a) shows that there is an apparent disparity between the directions of propagation of the TM-polarized SH pulse and its associated momentum vector. For example, in Fig. 3 the TM-polarized harmonic is seen to clearly follow the path of the pump pulse, while its momentum points in a completely different direction [Fig. 4(a)]. The momentum refraction angles depicted in both figures are not to scale, and refer to the inhomogeneous or phase-locked components. The dynamics of both the SH and TH signals is anomalous to different degrees. The way they differ from the expected generation angles depends from the actual form of the nonlinear sources (namely the $\chi^{(2)}$ and $\chi^{(3)}$ tensors). For example, the TM- and TE-polarized SH signal are different in that the $\chi^{(2)}$ is zero for the former but not for the latter. As the pulse crosses the interface both homogeneous and inhomogeneous components begin to form, but the homogenous components are quickly reabsorbed within a very short distance from the surface. However, the momenta of the two components are sustained by very different dynamics. The TM-polarized SH beam develops as a result of nonlinear gain that comes as a combination of a surface ($\nabla \cdot P$) and a volume ($E \times B$) term, both vector quantities. In contrast, the gain for the TE-polarized component comes mostly from a term proportional to $E_{\text{TE}}^2 \sim \left|\epsilon\right|P_{\text{TM}}(z)E_{\text{TM}}^2 \exp(-2\omega z)$, as described above. Then one can see how the nature of the nonlinear gain, which is very dynamic near the surface, where both components exists and interact, can be very different for the two components.

To reconcile these apparent differences we calculate the electromagnetic momentum of a wave packet located inside a medium of thickness $L$ as a function of time using the usual expression:

$$P_{\xi, \tilde{y}}(\tau) = \frac{1}{c^2} \int_{\xi=0}^{\xi=L} \int_{\tilde{y}=\infty}^{\infty} S_\xi(\tilde{y}, \xi, \tau) d\tilde{y} d\xi,$$  \hspace{1cm} (12)

where

is the Abraham momentum density. After substituting Eqs. (1) and (2) into Eq. (12) one may then use the resulting

$$S_{\xi, \tilde{y}}(\tilde{y}, \xi, \tau) = \frac{c}{4\pi} E \times H$$  \hspace{1cm} (13)

Fig. 3. (Color online) (Media 1) Snapshot of a TM-polarized pump pulse propagation $\sim$30 fs in duration tuned to 670 nm after it crosses the interface into GaP and generates SH and TH fields. The material is denoted by white boundaries and it is surrounded by air on both sides. (Top row) TM-polarized fields. (Bottom row) TE-polarized fields. Part of the signals are specularly reflected and part is transmitted with similar results for all generated fields. Only the pump refracts into the medium proceeds as predicted by Snell’s law. The SH and TH fields are locked to the pump."

Fig. 4. (Color online) Pump (red), SH (blue), and TH (magenta) momentum refraction angles. The pump’s momentum refraction angles coincide well with predictions made using Snell’s law. Pump tuning at 670 nm (a) and 1200 nm (b). The angles shown for the momenta of SH and TH beams refer only to the inhomogeneous portions of the signals, especially in (b) where both homogeneous and inhomogeneous SH signals are allowed in the absence of absorption.
components to extract the momentum refraction angles as a function of time for each frequency as the pulse enters and settles inside the medium, for example:

\[ \theta_{\alpha,\omega}(\tau) = \tan^{-1}\left[\frac{P_{g,\alpha}(\tau)}{P_{2,\alpha}(\tau)}\right]. \]  \hspace{1cm} (14)

This approach typically yields results nearly identical to those predicted by Snell’s law, even for pulses only a few optical cycles in duration [23,24].

The apparent inconsistency between the visual clues found in the media file of Fig. 3 and the results of Eq. (12) can be easily resolved because Eq. (12) in fact gives information about the total momentum of the bound pump-harmonics system, while Eq. (14) represents an arbitrary separation of the individual momenta. The situation is summarized in Fig. 5. Under the conditions we have explored, a phase mismatch ensures that the pump remains undepleted for all reasonable nonlinear coefficients and incident peak powers, so that each of the momenta of the harmonic signals is always several orders of magnitude smaller compared to the pump momentum. In this view, the harmonics remain trapped by the pump and move in one direction while their momenta may actually point in another [23]. Then, the total momentum and total energy for the bound state always nearly coincide with the pump’s momentum and energy. In other words, under ordinary conditions the generated harmonic pulses do not gain enough momentum/energy to significantly affect the direction of motion of the system, at least in the regime under consideration. However, one can easily create an artifact capable of depleting the pump and thus cause the momentum-energy refraction angles to depart from the prediction of Snell’s law due to the intervening nonlinear interaction, making possible the observation of anomalous refraction of the generated harmonic beams.

4. CONCLUSIONS

In summary, one of the unique properties of bulk semiconductors is that they exhibit a spectral range where the dielectric permittivity is negative, while providing a transparency region at near IR and longer pump wavelengths. For example, GaP is a potential candidate transparent above 500 nm. While the second harmonic pulse might be tuned around the 330 nm resonance of GaP, the third harmonic pulse is tuned in a region of negative permittivity [10]. These features cause the fields to behave in peculiar fashion, as phase locking forces energies flow and momenta to display apparent anomalies. In this work we have studied this dynamic theoretically and numerically. Another feature of semiconductors is that they can be doped to tune the dielectric properties [25]. It has been shown recently that nanorings could be used to realize metamaterials in the optical regime [26]. Another approach that has been proposed involves the use of a four-level atomic medium having electric and magnetic transitions in hydrogen and neon atoms [27]. Since semiconductors already exhibit an intrinsic negative permittivity, the introduction of suitable dopant atoms could make it possible to use magnetic transitions to create an effective negative magnetic permeability. The present effort represents another tassel in a more comprehensive effort that serves to bridge the gap between a number of physical effects, such as harmonic generation, subdiffraction limited imaging, negative refraction, and surface plasmons using nonlinear semiconductor materials.

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