Photon acceleration and polariton wakefields in dielectric crystals

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Abstract. Nonlinear processes associated with short laser pulse propagation in dielectric crystals are considered. They include photon acceleration of probe high frequency photons interacting with long wavelength polariton fields, and polariton wakefield excitation by short laser pulses. This could be useful to the understanding of tera-Hertz radiation sources.

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1. Introduction

Polaritons are well-known photon states coupled with transverse phonons that can be excited in dielectric crystals, and their nonlinear properties have been studied for many years [1]. But quite recently, interest has been focused on the interaction of short and long wavelength polariton...
radiation, as an important process that could lead to the development of new sources of terahertz radiation [2]. Until now, the theoretical studies in this area have mainly been focused on nonlinear wave scattering and wave mixing processes. We propose here to study new processes that, unlike the previous ones, are specific to short laser pulses and can only occur for pulse durations below a certain threshold.

We will focus our attention on two complementary aspects of the nonlinear wave interactions. One is the photon frequency shift, or photon acceleration, suffered by the short wavelength photons in the presence of long wavelength perturbations; for instance, the perturbations associated with the nonlinear refractive index in the presence of a polariton field. Apart from its intrinsic value, as a phenomenon where photons can behave very much as particles and manifest their particle-like character, photon acceleration can also be useful for the development of new tunable optical sources [3]. The second process considered here is the possible excitation of polariton wakefields produced by driving short laser pulses with higher frequency, for instance in the near infrared domain. This process could be useful to the excitation of much longer wavelength radiation, such as terahertz radiation.

These two processes, photon acceleration and polariton wakefield, can be seen as two different, and in some sense complementary, ways of transferring electromagnetic energy between the long wavelength and the short wavelength domains. In contrast with the usual scattering and wave mixing processes, they do not satisfy the usual frequency wave mixing and phase matching conditions, and are particularly relevant to the physics of ultra-short laser pulses. In contrast, they satisfy other types of resonant conditions, related to the pulse durations and their group velocities, as shown below.

The paper is organized in the following way. In section 2, we state the basic equations and establish the nonlinear polariton dispersion relation. In section 3, we discuss acceleration of high frequency photons in a dielectric crystal, in the presence of a long wavelength polariton field, and estimate the expected frequency shifts. In section 4, we address the problem of polariton wakefield excitation by a short laser pulse propagating in a dielectric crystal, establish the expressions for the wakefield amplitude and discuss the conditions for optimum polariton excitation. Finally, in section 5, we state our conclusions.

2. Dispersion relations

We assume that the photon–phonon electric field is polarized along the axis of symmetry of the crystal, and that the lattice also vibrates in the same direction. The transverse polariton wave is assumed to propagate along a given direction z. The electric field $E(z, t)$ and the lattice displacement $Q(z, t)$ are then described by the following coupled equations [2, 4]

$$\left(\frac{\partial^2}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2}\right) E = \mu_0 \frac{\partial^2}{\partial t^2} (P_L + P_{NL}) \tag{1}$$

and

$$\left(\frac{\partial^2}{\partial t^2} + \Gamma \frac{\partial}{\partial t} + \omega_0^2\right) Q = \frac{eE}{M} + \frac{\chi Q}{2M} E^2, \tag{2}$$
where the linear and nonlinear polarization vectors are determined by

\[
P_L(t) = \epsilon_0 \int_0^\infty \chi(t) E(t - \tau) \, d\tau + eNQ \tag{3}
\]

and

\[
P_{NL}(t) = \epsilon_0 \left( \frac{1}{2} \chi E^2 + N_i \chi Q E \right). \tag{4}
\]

Here, \(M, e, N_i\) are the reduced ion mass, effective charge and number of ion pairs per unit volume, \(\omega_0\) is the resonant frequency of the lattice, and \(\Gamma\) the damping rate of the transverse phonons. We have also used the linear response function \(\chi(t)\), and the nonlinear susceptibilities \(\chi_E\) and \(\chi_Q\). Equation (2) describes the coupled electron–ion oscillations in the crystal \([1]\), and the damping rate \(\Gamma\) is mainly due to scattering off impurities and imperfections, and to coupling with thermal vibrations of the crystalline structure. Typical values of the resonance frequency \(\omega_0\) are in the range between hundreds of giga-hertz and a few tera-Hertz. As an example, for a gallium phosphide crystal (GaP), we have \(\omega_0 = 11.0\) THz and \(\Gamma \approx 0.12\) THz \([2]\). On the other hand, the high frequency photons will be typically in the visible or near infrared.

Let us first consider the linear approximation, by assuming that \(\chi_E = \chi_Q = 0\). For monochromatic wave perturbations with frequency \(\omega\) and wavenumber \(k\), of the form \(E, Q \sim \exp(ikz - i\omega t)\), we obtain from equations (1) and (2)

\[
(k^2 c^2 - \omega^2)E = \omega^2 \left[ \chi(\omega) E + eN_i Q / \epsilon_0 \right], \quad (\omega^2 + i\Gamma \omega - \omega_0^2)Q = -(e/M)E. \tag{5}
\]

Defining the plasma frequency of the medium \(\omega_p\), and the high frequency limit of the permittivity \(\epsilon_\infty(\omega)\), such that

\[
\omega_p^2 = \frac{e^2 N_i}{\epsilon_0 M}, \quad \epsilon_\infty(\omega) = 1 + \chi(\omega), \tag{6}
\]

we obtain from equation (5) the linear polariton dispersion relation

\[
\frac{k^2 c^2}{\omega^2} = \epsilon_\infty(\omega) - \frac{\omega_p^2}{(\omega^2 + i\Gamma \omega - \omega_0^2)}. \tag{7}
\]

In the high frequency limit, such that \(\omega^2 \gg \omega_p^2, \omega_0^2\) this reduces to \(k^2 c^2 / \omega^2 = \epsilon_\infty(\omega)\).

Let us now assume the nonlinear case, where \(\chi_E \neq 0, \chi_Q \neq 0\). We can now study the propagation of high frequency photons, with frequency \(\omega'\) and wavenumber \(k'\), in the presence of a background polariton field \(E(z, t)\) and \(Q(z, t)\). For these high frequency photons, the associated electric field \(E' \sim \exp(ik'z - i\omega' t)\) can be described by equation (1), where we can now neglect the lattice displacement \(Q'\), and the nonlinear polarization can be written as

\[
P_{NL}' = \epsilon_0 E' \int \chi_{NL}(\omega, \omega') Q_\omega \exp(ikz - i\omega t) \frac{d\omega}{2\pi}. \tag{8}
\]
Here, $k$ is assumed as a function of $\omega$, as determined by the polariton dispersion relation (7), and $Q_\omega$ is the spectral polariton amplitude. Using the above equations, the nonlinear susceptibility can be written as

$$\chi_{\text{NL}}(\omega, \omega') = \chi_Q N_i - \frac{M}{2e} (\omega^2 + i\Gamma \omega - \omega_0^2).$$

For the simple case of a monochromatic low frequency polariton background, such that $Q(z, t) = Q_0 \cos(kz - \omega t)$, equation (8) will simply reduce to

$$P'_{\text{NL}} = \epsilon_0 \chi_{\text{NL}}(\omega, \omega') Q(z, t) E'. \tag{10}$$

For very high frequency photons propagating in the crystal in the presence of a polariton excitation, such that $\omega' \gg \omega$, and the time derivatives of $Q(z, t)$ can be neglected in the propagation equation (1), we can derive the following nonlinear dispersion relation

$$D(\omega', k'; z, t) \equiv k'^2 c^2 - n^2(\omega', k'; z, t) = 0, \tag{11}$$

where

$$n(\omega', k'; z, t) = [\epsilon_\infty(\omega') + \chi_{\text{NL}}(\omega, \omega') Q(z, t)]^{1/2} \tag{12}$$
is the photon refractive index inside the crystal.

### 3. Photon acceleration

The above dispersion relation is valid in the geometric optics approximation, for $\omega' \gg \omega$ and $k' \gg k$. It describes the photon behaviour locally, both in space and time, when the nonlinear refractive index of the high frequency photons is modulated by the low frequency polariton field, represented by $Q(z, t)$. As a result, photon acceleration (or frequency shift) of the high frequency photons will take place, as can be seen from the photon ray equations

$$\frac{dz}{dt} = v', \quad \frac{dk'}{dt} = F', \tag{13}$$

where the group velocity $v'$ and the force acting on the photons $F'$ are determined by

$$v' = -\frac{\partial D/\partial k'}{\partial D/\partial \omega'}, \quad F' = \frac{\partial D/\partial z}{\partial D/\partial \omega'} \tag{14}$$

Using equation (11), this can be explicitly written as

$$v' = \frac{c}{n + \omega' (\partial n/\partial \omega')}, \quad F' = \frac{\omega' v'}{2n c} \chi_{\text{NL}} \frac{\partial Q}{\partial z}. \tag{15}$$
We then get the following expressions for the photon ray equations

\[
\frac{dz}{dt} = \frac{c}{n} \frac{1}{1 + G(k', z, t)} \quad \frac{dk'}{dt} = \frac{k'}{2n^2} \frac{\partial Q}{\partial z},
\]

where we have used the auxiliary function

\[
G(k', z, t) = \frac{k'c}{2n^3} \frac{\partial}{\partial \omega'} \left[ \epsilon_\infty + \chi_{NL} Q(z, t) \right].
\]  

The photon acceleration process due to the presence of the polariton field \( Q(z, t) \) can be studied by integrating these equations of motions. But here, we are more interested in determining the order of magnitude of the expected photon acceleration effects, in generic conditions. For this purpose, it is then useful to introduce some reasonable simplifying assumptions. Firstly, we assume that we have small dispersion in the region of interest, which means that \( G(k', z, t) \ll 1 \), or equivalently, that \( \omega' (\partial \epsilon_\infty / \partial \omega') \ll 1 \) and \( \omega' (\partial \chi_{NL} / \partial \omega') \ll 1 \). Secondly, we assume that the nonlinear terms remain small, or that \( \chi_{NL} |Q(z, t)| \ll \epsilon_\infty \). The ray equations (16) can then be reduced to

\[
\frac{dz}{dt} = \frac{c}{\sqrt{\epsilon_\infty}} \left[ 1 - \frac{1}{2} \frac{\chi_{NL}}{\epsilon_\infty} Q(z, t) \right], \quad \frac{dk'}{dt} = \frac{k'c}{2n^2} \frac{\chi_{NL}}{n^2} \frac{\partial Q}{\partial z}.
\]  

If we neglect the nonlinear higher order corrections to the nonlinear terms, we can rewrite these equations of motion in the canonical form

\[
\frac{dz}{dt} = \frac{\partial H}{\partial k'}, \quad \frac{dk'}{dt} = - \frac{\partial H}{\partial z},
\]  

where the corresponding Hamiltonian function is given by

\[
H(k', z, t) = \frac{k'c}{\sqrt{\epsilon_\infty}} \left[ 1 - \frac{1}{2} \frac{\chi_{NL}}{\epsilon_\infty} Q(z, t) \right].
\]  

Notice that these equations of motion predict a frequency shift, due to the explicit time dependence of the Hamiltonian. If \( H(z', z, t) \) was not time dependent the equations (19) would only describe refraction. But in our case, the photon frequency shift is always present, as stated by

\[
\frac{d\omega'}{dt} = \frac{\partial \omega'}{\partial t} = - \chi_{NL} \frac{\omega^3}{2k^2c^2} \frac{\partial Q}{\partial t}.
\]

At this point, it should also be noticed that, for a polariton field of the form \( Q(z, t) = Q_0 \cos(kz - \omega t) \), this Hamiltonian function only depends on space and time through the combination \( (z - v_p t) \), where \( v_p = \omega / k \) is the polariton phase velocity. This suggests the use of a coordinate transformation \( (z, k') \rightarrow (\eta, p) \) determined by the generating function
\[ F(z, p, t) = (z - v_p t) p. \] Such a canonical transformation will then be given by

\[ k' = \frac{\partial F}{\partial z} = p, \quad \eta = \frac{\partial F}{\partial p} = (z - v_p t) \quad (22) \]

and the equations of motion will be transformed into

\[ \frac{d\eta}{dt} = \frac{\partial h}{\partial p}, \quad \frac{dp}{dt} = -\frac{\partial h}{\partial \eta}, \quad (23) \]

where the new Hamiltonian function is

\[ h(\eta, p) = H + \frac{\partial F}{\partial t} = H(\eta, p) - v_p p \quad (24) \]

or, in explicit form

\[ h(\eta, p) = \frac{pc}{\sqrt{\epsilon_\infty}} \left[ 1 - \frac{1}{2} \frac{\chi_{NL}}{\epsilon_\infty} Q_0 \cos(k\eta) \right] - v_p p. \quad (25) \]

The most interesting aspect of these new photon equations is that their Hamiltonian function is not explicitly dependent on time, or in other words, it is a constant of motion. This allows us to establish the maximum photon frequency shift due to its nonlinear coupling with the polariton field, without solving the photon equations of motion. For this purpose, let us define two distinct positions in the new coordinates, \( \eta_1 \) and \( \eta_2 \), that correspond to a maximum and a minimum of the field \( Q(\eta) \). For instance, we can take \( \eta_1 = 0 \) and \( \eta_2 = \pi/k \). Calling \( p_1 \) and \( p_2 \) the values taken by the photon momentum (or wavenumber) at these two particular positions, and \( \omega'_1 \) and \( \omega'_2 \) the corresponding photon frequencies, we can establish that, for a given photon trajectory

\[ h(\eta, p) = \omega'_1 - v_p p_1 = \omega'_2 - v_p p_2. \quad (26) \]

Using the relation between frequencies and wavenumbers, this allows us to establish the following relation

\[ \omega'_2 = \omega'_1 \frac{1 - (v_p/c)n(\eta_1)}{1 - (v_p/c)n(\eta_2)} = \omega'_1 \frac{1 - (v_p/c)\sqrt{\epsilon_\infty} + \chi_{NL} Q_0}{1 - (v_p/c)\sqrt{\epsilon_\infty} - \chi_{NL} Q_0}. \quad (27) \]

We see from this result that the photon frequency shift can be arbitrarily large close to the resonant condition \( n(\eta_2) = c/v_p \), which corresponds to the equality between the final photon group velocity and the polariton phase velocity, \( v_p = v'(\eta_2) \). See figure 1 for an illustration. This is particularly interesting for the case of a single cycle polariton field, but can also be applied to describe what occurs for each cycle of a larger polariton wave, where oscillations of the photon frequency shift can take place, due to the conservative (or reversible) character of the above photon ray equations.

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Figure 1. Relative frequency shift $Y = \frac{\omega'_2}{\omega'_1}$, as a function of $X = (v_p/c)\sqrt{\epsilon_{\infty}}$, for two different values of the nonlinear parameter $\delta = (\chi_{NL}Q_0/\epsilon_{\infty})$: (1) $\delta = 0.02$ and (2) in bold, $\delta = 0.2$.

4. Polariton wakefield

The description outlined above can lead to the prediction of interesting new phenomena, such as photon acceleration or frequency shift in a dielectric crystal. But this is only relevant to the case where the polariton field is strong enough to disturb the trajectory of probe high frequency photons. We will now consider the opposite case of a strong laser pulse propagating in the crystal, when the polariton modes are initially not present.

In this new situation, a polariton wakefield can be excited by the laser pulse. We will show that the amplitude of the polariton wakefield is very sensitive to the duration of the short laser pulse. We start with the polariton equations for the electric field $E$ and for the transverse lattice displacement $Q$, written in the following way

\[
\left( \frac{\partial^2}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) E = \frac{\partial^2}{\partial t^2} \left[ P_L + \frac{1}{2}\epsilon_0 \chi_E I(z, t) \right]
\] (28)

and

\[
\left( \frac{\partial^2}{\partial z^2} + \Gamma \frac{\partial}{\partial t} + \omega_0^2 \right) Q = \frac{eE}{M} + \frac{\chi_0}{2M} I(z, t),
\] (29)

where $I(z, t) = |E'(z, t)|^2$ is the envelope of the driving laser pulse. Notice that, with respect to our initial equations (1) and (2), we have neglected here the high frequency nonlinear terms that are associated with the well-known wave mixing and scattering processes. Here, we will concentrate on the low frequency nonlinearities that are relevant to the physical processes associated with very short laser pulses. Notice that the linear polarization vector (3) can also be written as

\[
P_L(t) = \epsilon_0 \tilde{\chi}(t) E(t) + eN_i Q(t),
\] (30)

where the averaged susceptibility $\tilde{\chi}$ is defined in general terms as

\[
\tilde{\chi}(t) = \frac{\int \chi(\omega) E(\omega) \exp(-i\omega t) d\omega}{\int E(\omega) \exp(-i\omega t) d\omega}.
\] (31)
where $\chi(\omega)$ is the usual spectral susceptibility. In the simple case of a nearly monochromatic polariton oscillation with frequency $\omega$, we can take the usual approximation $\bar{\chi} \simeq \chi(\omega)$. But, in our case, this approximation will have to be verified a posteriori, and we prefer to keep the value of $\bar{\chi}$ as unspecified.

For a short laser pulse propagating with group velocity $v'$, assumed constant, we can write $I(z, t) \equiv I(z - v't)$. This suggests the use new coordinates $\xi$ and $\tau$, such that:

$$\xi = z - v't, \quad \tau = t,$$

(32)

and

$$\frac{\partial}{\partial z} = \frac{\partial}{\partial \xi}, \quad \frac{\partial}{\partial t} = \frac{\partial}{\partial \tau} - v' \frac{\partial}{\partial \xi}.$$

(33)

In these new coordinates, the driving laser pulse will be at rest and we can assume that it leaves behind it a nearly static wakefield. This allows us to assume the quasi-static approximation such that $\partial / \partial \tau \simeq 0$. Replacing the new coordinates in the above equations (28) and (29), and use such an approximation, we are then led to the following equation

$$\left( v'^2 \frac{\partial^2}{\partial \xi^2} - \nu \frac{\partial}{\partial \xi} + \omega_0^2 \right) Q = \frac{eE}{M} + \frac{\chi_0}{2M} I(\xi).$$

(34)

The polariton electric field is now determined by

$$E = \frac{v'^2}{c^2} \gamma_p \left[ \frac{eN_i}{\epsilon_0} Q + \frac{1}{2} \chi_E I(\xi) \right],$$

(35)

and

$$\gamma_p = \left[ 1 - (1 + \bar{\chi}) \frac{v'^2}{c^2} \right]^{-1/2}$$

(36)

is the relativistic gamma factor of the laser pulse with respect to the polariton velocity $c/\sqrt{1 + \bar{\chi}} \simeq c/\sqrt{\epsilon_\infty}$. Replacing in equation (34), using the quantities $\nu = \Gamma / v'$ and $k_0 = \omega_0 / v'$, and introducing the new parameters $k$ and $B$, such that

$$k^2 = k_0^2 \left( 1 - \frac{\omega_p^2}{\omega_0^2} \frac{v'^2}{c^2} \gamma_p^2 \right), \quad B = \frac{1}{2Mc^2} \left( e\chi_E + \frac{c^2}{v'^2} \chi_0 \right),$$

(37)

we derive

$$\left( \frac{\partial^2}{\partial \xi^2} - v' \frac{\partial}{\partial \xi} + k^2 \right) Q = BI(\xi).$$

(38)

We can now use the quantity $g(\xi)$, given by

$$Q(\xi) = g(\xi) \exp(\nu \xi / 2),$$

(39)
Figure 2. Normalized polariton wakefield $Q(x)$ produced by a Gaussian laser pulse $I(x) = I_0 \exp(-x^2/x_0^2)$, for $B = 1$, $v = 1/10$ and $x_0 = 4$. The driving laser pulse is also shown.

which leads to the following simple equation, which coincides to that of a forced linear oscillator with unit frequency

$$\left(\frac{d^2}{dx^2} + 1\right) g = f(x),$$

(40)

where we have used the new variable

$$x = (k^2 - v^2/4)^{1/2} \xi,$$

(41)

and the new force term

$$f(x) = \frac{BI(x)}{(k^2 - v^2/4)} \exp\left(-\frac{v}{2} \frac{x}{\sqrt{k^2 - v^2/4}}\right).$$

(42)

The forced solution equation (40), compatible with the present physical problem, such that the signal $g(x)$ can only occur after the passage of the driver $I(x)$, is determined by

$$g(x) = \int_{-\infty}^{x} f(x') \sin(x - x') \, dx'.$$

(43)

From this equation, we can then retrieve the lattice perturbations $Q(\xi)$. These perturbations depend critically on the duration of the driving laser pulse. There is an optimum laser pulse width, of the order of $\Delta v \simeq 1$, as shown by the solution (43), above which the sine function under the integral will produce interfering oscillations that reduce the maximum value of $g(x)$, for a given driving field $I(x)$. This is illustrated with the numerical integration shown in the figures 2 and 3, for a laser pulse slightly larger and shorter than the optimal width, respectively, for moderate values of the dissipation parameter $\nu$. The normalized Gaussian profile of the laser pulse envelope is also shown for comparison.

We see that, for the case of shorter laser pulses, a polariton wavefield is clearly produced and propagates behind the laser pulse, with a phase velocity equal to the group velocity of the polaritons.
pulse. In contrast, for the larger pulse case, the main disturbance is directly associated with the laser pulse itself, and only a low amplitude residual wakefield remains. Therefore, the laser pulse duration, and in a lesser degree, the dissipation parameter $\nu$, are determinant factors for the production of a well-behaved polariton wakefield. In any case, the period of the wakefield oscillation is always the same, and corresponds to a frequency $\omega$, as observed in the laboratory frame, given by

$$\omega = kv' \left(1 - \frac{\nu^2}{4k^2}\right)^{1/2} = \omega_0 \left(1 - \frac{\omega_p^2}{\omega_0^2} \frac{\nu^2}{c^2 \gamma_p^2} \right)^{1/2} \left(1 - \frac{\nu^2}{4k^2}\right)^{1/2}. \quad (44)$$

Notice that these wakefield oscillations contain both lattice vibrations $Q(\xi)$ and electromagnetic field oscillations $E(\xi)$, at the same frequency $\omega$, which is always smaller than the lattice resonant frequency $\omega_0$. This means that an electromagnetic field disturbance at this frequency will be excited by a short laser pulse with a much larger frequency $\omega'$, moving with velocity $v' = \partial \omega' / \partial k'$, and will eventually radiate out of the crystal. In this way, excitation of low frequency (for instance tera-Hertz pulses), by high frequency (for instance, near infrared) laser pulses will be expected by the above mechanism. This can be seen as a result of the lattice perturbation associated with the ponderomotive force of the driver laser pulse. Such an excitation of low frequency radiation in the polar crystal is favoured for laser pulses with a duration of the order of the characteristic period of the transverse lattice vibrations $1/\omega_0$.

5. Conclusions

We have shown in this study that nearly resonant interactions between long wavelength polaritons and short wavelength laser pulses can take place. We have focused on two different aspects relevant to the case of short laser pulses. We have studied the possible frequency shift (or photon acceleration) of probe high frequency photons in the field of a polariton. We have determined the conditions for photon trapping and derived an expression for the maximum frequency shift.

We have also studied the complementary process of polariton wakefield excitation by a short laser pulse. We have shown that stronger wakefields are expected for laser pulses shorter that the
characteristic period of the transverse lattice vibrations. This process is due to the ponderomotive force of the laser acting on the lattice and can be seen as the optical analogue to the well-known electrostatic wakefields produced in a plasma by strong laser pulses [5] and currently used for particle acceleration purposes [6]. The present model could then be useful to develop tera-Hertz radiation sources driven by lasers [7]. It was recently shown that femto-second laser sources at around 800 nm could generate a few cycles of THz radiation [8].

In a real physical situation, these two processes can take place simultaneously. A short laser pulse propagates in the crystal and excites a polariton wakefield, that can accelerate the photons belonging to the laser pulse itself, thus leading to an increase of the laser spectral width. Ideally, these processes conserve the spectral phase and could eventually be used to compress the initial short laser pulses to even shorter durations. We therefore think that the present theoretical model could be useful to the understanding of nonlinear interaction of short pulses in a polar crystal and could stimulate new theoretical and experimental studies.

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