Wave propagation in birefringent materials with off axis absorption or gain

Mahmood Sabooni\textsuperscript{a,b} and Adam N. Nilsson\textsuperscript{b}, Gerhard Kristensson\textsuperscript{c}, Lars Rippe\textsuperscript{b}

\textsuperscript{a} Department of Physics, University of Tehran, 14399-55961, Tehran, Iran
\textsuperscript{b} Department of Physics, Lund University, P.O. Box 118, SE-22100 Lund, Sweden
\textsuperscript{c} Department of Electrical and Information Technology, Lund University, P.O. Box 118, SE-22100 Lund, Sweden

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The polarization direction of an electromagnetic field changes and eventually reaches a steady state when propagating through a birefringent material with off axis absorption or gain. The steady state orientation direction depends on the magnitude of the absorption (gain) and the phase retardation rate. The change in the polarization direction is experimentally demonstrated in weakly doped (0.05\%) Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} crystals, where the light polarization, if initially aligned along the most strongly absorbing principal axis, gradually switch to a much less absorbing polarization state during the propagation. This means that the absorption coefficient, \(\alpha\), in birefringent materials generally varies with length. This is important for, e.g., laser crystal gain media, highly absorbing and narrow band spectral filters and quantum memories.

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INTRODUCTION

For electromagnetic plane wave propagation in birefringent materials the instantaneous polarization direction of the electric field vector, \(\mathbf{E}\), normally changes during the propagation. For example, the polarization of a wave propagating along one of the principal axes in a (non-absorbing) birefringent material with (initially) linear polarization at an angle \(\gamma\) relative one of the other principal axes, changes from linear to elliptical (with major axes \(|\mathbf{E}| \cos \gamma\) or \(|\mathbf{E}| \sin \gamma\)) and then to linear polarization at angle \(-\gamma\) and then back to elliptical etcetera as it propagates through the material. For a case where the wave is not propagating along one of the principal axes the wave vector and the Poynting vector are in general not parallel and there are walk off effects (e.g. chapter 6 in Ref. \textsuperscript{[1]}).

In this work we consider an initially linearly polarized plane wave, propagating along a principal axis in a nonmagnetic off axis absorbing birefringent material. In this case polarization rotation can occur even if the initial linear polarization is aligned with one of the principal axes. In materials with absorption or gain the development of the polarization can be complicated. Not only could there be oscillations between linear and elliptical polarization and walk off effects, there can also be conversions from light of one polarization to another polarization due to the absorption (or gain).

Such effects should for example be present in laser crystal gain media when the gain is anisotropic and the gain tensor is not aligned with the principal axes \textsuperscript{[2]}. Recently there has also been an interest in highly absorbing and very narrow bandwidth filters for very specific applications such as quantum memories \textsuperscript{[3]}, or highly absorbing narrowband filters with exceptionally large extendue for high performance ultrasound optical tomography \textsuperscript{[4,5]}

and also dynamically tunable high performance filters \textsuperscript{[6]}. All these recent papers use filters based on rare-earth-ion-doped inorganic crystals where the absorption tensor is not aligned with the principal axes. As a consequence the light polarization gradually switches during propagation to a much less absorbing polarization state even if the input polarization is perfectly aligned with the most strongly absorbing principal axis. This significantly decreases the achievable filter attenuation from what might be anticipated based on just the material absorption coefficient.

In this work we theoretically analyze the polarization rotation in absorbing birefringent materials with a model adapted from \textsuperscript{[7]}. We provide some simple relations of when the effects need to be taken into account and give possible suggestions on how to maintain the original high absorption. The theoretical analysis is supported by an experimental demonstration of the polarization rotation effect.

The paper is organized as follows. First an intuitive theoretical background is given. This is followed by simulation results and discussions of polarization steady states and the effects of absorption on light propagating through a crystal. Experimental results are then presented where the incoming and outgoing polarization directions are studied for crystals with different absorption and length. Lastly a remark about the maximum absorption axis is made before the paper is concluded with a summary. A rigorous mathematical treatment of the problem can be found in the Supplementary Information section.

THEORETICAL BACKGROUND

A detailed description of the theoretical framework is given in the Supplementary Information, but a simple
and intuitive understanding can be achieved by considering the effects of birefringence and absorption separately. In Fig. 1a we see the principal axes of a birefringent crystal as well as the direction of the transition dipole moment, $\mu$. This direction does not coincide with any of the principal axes of the crystal. An incoming field, $E_{in}$, which is on resonance with the absorbing transition and with its polarization oriented along one of the principal axes, $D_2$, generates a polarization, $P$, along $\mu$ with a 90° phase shift. This polarization generates a field, $E_P$, along $\mu$ with a relative phase relation to $E_{in}$ as shown in Fig. 1b. The total resulting electromagnetic field, $E_{tot} = E_{in} + E_P$, now also have a component along the $D_1$ axis as shown in Fig. 1c. Since the crystal is birefringent the polarization state oscillates between linear (Fig. 1d) and elliptical (Fig. 1f) as the field propagates through the crystal. Of course in reality the effects of absorption and birefringence are intertwined and occur simultaneously. The theoretical examination in the Supplementary Information makes it possible to simulate the propagation of any incoming electric field polarization through a birefringent crystal with an absorption axis with an angle to the principal axes. The method works for incoming plane waves and assumes that the lateral (transverse) dimensions of the crystal are infinite as well. Discussions around the results from these simulations can be found in the next section.

**Simulation Results**

In this section results from simulations of light propagation inside a birefringent crystal with a tilted transition dipole moment direction is analyzed. As a specific example a 0.05% Pr$^{3+}$:Y$_2$SiO$_5$ crystal is chosen where the transition dipole moment is tilted by 74° from the $D_1$ crystal axis as seen in Fig. 2 and calculated in the Supplementary Information. 

**Phase retardation in a birefringent crystal**

The propagation phase, $\phi$, can be written as $\phi = 2\pi \frac{n_z}{\lambda_0}$ radians, i.e. light with the wavelength $\lambda_0$ in vacuum experiences a phase shift of $\phi$ radians when traveling an optical length $nz$ through a material, where $n$ is the index of refraction and $z$ is the propagation depth. The difference between the accumulated phases, $\phi_2$ and $\phi_1$, for light propagating along two different crystal axes, $D_2$ and $D_1$ respectively, is termed the phase retardation:

$$\Delta \phi = \phi_2 - \phi_1 = 2\pi \frac{n_z}{\lambda_0}$$  \hspace{1cm} (1)

where $\Delta n = n_{D_2} - n_{D_1}$. The crystal length needed to obtain a quarter-wave plate, $\Delta \phi = \pi/2$, for a Y$_2$SiO$_5$ crystal with the different refractive indices; $n_{D_1} = 1.7881$, $n_{D_2} = 1.809$ and $n_b = 1.7851$ along the principal axes as calculated using Eq. (1) to be about 7.25 $\mu$m for light propagating along the $b$ axis.

**Polarization steady states**

From the effects explained in Fig. 1 it is clear that it is not possible to maintain a pure linear polarization when propagating through a crystal where the transition dipole moment axis does not coincide with any of the optical axes of the crystal. However, in general two steady state polarization solutions exist for a forward propagating wave. They are elliptically polarized and only differ by a 90° rotation of their major axes in the $D_1$-$D_2$ plane. The polarization of the steady state does not change when propagating through the crystal. The ellipticity and the direction of these polarizations depend on the following ratio:
FIG. 2. (Color online) (a) Transition dipole moment, \( \mu \), versus principal axes for Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5}(D\textsubscript{1} \perp D\textsubscript{2} \perp b). (b) Notation of linear incoming and elliptical outgoing polarizations are seen where \( \gamma \) is the angle between the \( D\textsubscript{1} \) axis and the incoming polarization and \( \psi \) is the angle between the \( D\textsubscript{1} \) axis and the major axis, \( a \), of the outgoing elliptical polarization.

\[
R = \frac{n_{D2}^2 - n_{D1}^2}{\chi_{abs}} \tag{2}
\]

where \( \chi_{abs} \) is the electric susceptibility connected to the absorption (for Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} see Eq. (20) in the Supplementary Information). In general only the steady state solution with the lowest absorption or highest gain is stable.

In Fig. 3a–c the steady state polarization solutions when the transition dipole moment makes an angle of 74.6° relative to the \( D\textsubscript{1} \) axis are shown for three different cases where \( R \gg 1 \), \( R \approx 1 \) and \( R \ll 1 \), respectively. For absorption the green curves are stable solutions and the red curves are unstable. The reverse is true in the case of gain. The first case, seen in Fig. 3a, is the same as for a 0.05% Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} crystal which has a transition dipole moment direction that is tilted 74.6° from \( D\textsubscript{1} \) and absorption coefficients along \( D\textsubscript{1} \) and \( D\textsubscript{2} \) of \( \alpha_{D1} = 3.6\pm0.5 \text{ cm}^{-1} \) and \( \alpha_{D2} = 47\pm5 \text{ cm}^{-1} \) which corresponds to an electric susceptibility for the absorption that is \( \chi_{abs} = (8.82\pm0.8)\times10^{-4} \). The difference \( n_{D2}^2 - n_{D1}^2 \) in Y\textsubscript{2}SiO\textsubscript{5} is 0.075 which gives a ratio \( R \approx 85 \gg 1 \). Here one can see that the steady state solutions lie respectively along the \( D\textsubscript{1} \) and the \( D\textsubscript{2} \) crystal axes, which is always the case for \( R \gg 1 \).

The next two cases seen in Fig. 3b and Fig. 3c use the same values mentioned above except that the absorption susceptibility, \( \chi_{abs} \), is increased by a factor 100 and 10,000 respectively. In the extreme where \( R \ll 1 \), seen in Fig. 3b, the solutions lie almost completely along the transition dipole moment (unstable for absorption) and perpendicular to it (stable for absorption). In between these extreme cases when \( R \approx 1 \), seen in Fig. 3c, the solutions lie between the crystal axes and the transition dipole moments and have a higher ellipticity than in the extremes.

The steady state solutions can be explained by the same phenomena that was discussed in Fig. 1 i.e. that an electric field in \( D\textsubscript{2} \) creates an electric field in \( D\textsubscript{1} \), and vice versa, due to the tilted absorption (gain) axis. The steady state solutions are polarization states where the sum of the components lost and absorbed in \( D\textsubscript{1} \) decays at the same rate as the sum of the components created and absorbed along \( D\textsubscript{2} \), which in other words means that the polarization stays the same.

The dependence on \( R \) can be intuitively explained by rewriting the numerator in Eq. (2) as:

\[
R = \frac{n_{D2}^2 - n_{D1}^2}{\chi_{abs}} = (n_{D2} + n_{D1})(n_{D2} - n_{D1}) \tag{3}
\]

If we now assume that \( n_{D1} \) and \( n_{D2} \) are roughly constant then the ratio \( R \) measures if \( n_{D2} \) is small/large compared to the absorption and \( \Delta n \) sets the rate at which phase retardation is accumulated (see Eq. (1)). In other words if \( R \gg 1 \) then the phase retardation dominates over the absorption. This makes all polarizations that are symmetric along the crystal axes equivalent (due to the effects explained in Fig. 2a–c) and therefore the only steady state solutions that can exist are along \( D\textsubscript{1} \) and \( D\textsubscript{2} \), see Fig. 3b. In the other extreme where \( R \ll 1 \) the absorption dominates and any phase retardation between the two electric field components can be ignored which leads to one solution orthogonal and one solution parallel to the transition dipole moment axis, see Fig. 3c.

Electric field absorption when \( R \gg 1 \)

From the simulations one can obtain the (expected) exponential decay in the electric field components along \( D\textsubscript{1} \) and \( D\textsubscript{2} \) as a function of propagation. For the example of Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} the decay coefficients can be seen in Fig. 4a for a linear input polarization along \( D\textsubscript{2} \) (\( \gamma = 90° \)). Even though the input polarization only has a \( D\textsubscript{2} \) component of the electric field, a \( D\textsubscript{1} \) component is almost immediately created in the crystal due to the effects explained in Fig. 1a,b. The relative size of the created component is the same as the ratio of the \( D\textsubscript{2} \) and \( D\textsubscript{1} \) component of the steady state, since at the steady state (when the polarization does not any longer change with propagation) the \( D\textsubscript{2} \) component is almost completely created by the tilted absorption of the \( D\textsubscript{1} \) component. Therefore it is a measure of how strong the connection between the two components is. For the case of Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} the initial electric field component along \( D\textsubscript{1} \) is approximately a factor of 330 less than the \( D\textsubscript{2} \) component (see Fig. 4a and Eq. (35) in the Supplementary Information).

The exponential decay along \( D\textsubscript{2} \) is much larger than along \( D\textsubscript{1} \) in the beginning of the propagation due to the fact that the transition dipole moment lies much closer
to $D_2$. However towards the end of the propagation the stable steady state solution is reached and both components decay at the same rate. Continuing in Fig. 4, the input polarization is changed to $\gamma = 45^\circ$ and the polarization steady state is reached sooner since the input polarization is more similar to the stable steady state. For a linear input polarization the highest absorption rate is achieved along $D_2$ but slows down after propagating around 5.4 mm, see Fig. 4a. This leaves us with the ultimate limit, which we call the “steady state limit”, of the maximum electric field absorption with a high absorption rate of the incoming light for this material to be $10^{-6} - 10^{-5}$ and this is reached for a 5.4 mm long crystal. To clarify, the light continues to be absorbed after this limit but at a much slower rate. In light of this limit it is clear that scaling a crystal longer than $\approx 5.4$ mm to gain absorption serves limited purpose and one can get exponentially higher absorption by avoiding the steady state limit. Note also that much earlier, at around 2.7 mm, the electric field component along $D_1$, which decays much slower, is of the same size as the component along $D_2$ and the $D_1$ component dominates after 2.7 mm, even though the $D_1$ component was zero before entering the crystal, this component can however be blocked with a polarizer after the crystal.

The (approximate) propagation distance needed to reach the steady state limit as a function of the input polarization $\gamma$ is shown in blue in Fig. 4. The electric field component along $D_1$ and $D_2$ when the steady state limit is reached is also shown in green and red respectively. Note that the $D_1$ electric field component is always a factor of $\approx 330$ larger than the $D_2$ component when the steady state solution is reached. Close to $\gamma = 90^\circ$ the propagation distance decreases quite rapidly as $\gamma$ departs from $90^\circ$ and a good control over the purity of the electric field polarization is required to have a high absorption and a steady state limit at 5.4 mm. The purity needed is in the order of the ratio of the $D_1$ and $D_2$ electric field components for the stable steady state, which for Pr$^{3+}$:Y$_2$SiO$_5$, seen in Fig. 3a, is a factor $\approx 330$ for the electric field, or equivalently a factor $10^5$ for the intensity components (see Eq. (35) in the Supplementary Information). This means that a polarizer with better suppression than $10^5$ and an angle precision of around $0.17^\circ$ is required to achieve the longest propagation distance with maximum absorption.

To avoid the steady state limit one has to suppress the $D_1$ component of the light. This can be done with e.g. a linear polarizer. To achieve high absorption it is therefore better to use several smaller crystals (lengths of $\leq 5.4$ mm) with linear polarizers blocking any $D_1$ component in between each set of crystals instead of using one long crystal.

**EXPERIMENTAL RESULTS**

To examine the polarization change during propagation several experiments were performed on crystals with different lengths. We used three 0.05% Pr$^{3+}$:Y$_2$SiO$_5$ crystals with different lengths (1 mm, 6 mm and 12 mm) and measured the outgoing polarization direction $\psi$ as a function of the incoming polarization angle $\gamma$, see Fig. 2b. The crystals were kept at cryogenic temperature (2 K). The setup is illustrated in Fig. 5. The absorption was probed with attenuated pulses to minimize saturation effects on the absorption and between each experiment a set of hole eraser pulses were used.
over a wide spectral range to make sure that no permanent hole burning occurred.

According to the simulations for a Pr\(^{3+}\):Y\(_2\)SiO\(_5\) crystal there is a minimum (sufficient) value of the absorption where the steady state solution is reached which means that the outgoing polarization is almost entirely along the \(D_1\) axis. A 12 mm Pr\(^{3+}\):Y\(_2\)SiO\(_5\) crystal is employed to illustrate this effect as shown in [Fig. 5], where three measurements at different frequencies within the inhomogeneous profile were performed. The inhomogeneous line-width, \(\Gamma\), of this crystal is measured to be about 9 GHz. The three sets of measurements at frequencies 0 GHz, 4 GHz and 13 GHz relative to the line center are shown in [Fig. 5] as yellow triangles, cyan squares and blue dots respectively together with simulations for each case displayed as a red dotted, dashed and solid curve. For high and intermediate absorption the outgoing polarization is along the \(D_1\) axis which agrees with the fact that the \(D_2\) component is absorbed much quicker than the \(D_1\) component.

As shown in [Fig. 1] the outgoing polarization shifts significantly during propagation. It is therefore impossible to determine if the outgoing polarization is in the range \(\psi = 0 - 90^\circ\) or \(\psi = 90 - 180^\circ\) for a given input polarization without a very precise knowledge of the crystal length. A quarter-wave plate distance was calculated previously to be \(L \approx 7.25\ \mu\text{m}\) which means that a full evolution of the phase is done in \(\approx 29\ \mu\text{m}\) in Y\(_2\)SiO\(_5\). If the crystal length is not known within this precision it is indeed impossible to determine the outgoing polarization direction. The crystal length used in the simulations is therefore changed from exactly 12 mm by a few tens of \(\mu\text{m}\) to the value that best matches the experimental results.

The absorption coefficients in [8] are measured in a crystal with a line-width of \(\Gamma = 6.2\ GHz\) and which nominally has the same Pr concentration, 0.05\% as in the present crystal. This would correspond to a peak absorption susceptibility of \(\chi^{\text{abs}}_{\text{peak}} = (8.82 \pm 0.8) \times 10^{-4}\). The inhomogeneous profile can be approximated as a Lorentzian profile \([8]\) that in our experiments have a line-width of \(\Gamma = 9\ GHz\). Assuming that the integral of the absorption should be the same for our crystal and the crystal used in [8] the peak absorption susceptibility of our crystal should be reduced by a factor 9/6.2 which gives \(\chi^{\text{abs}}_{\text{peak}} = (6.08 \pm 0.55) \times 10^{-4}\). Since the three measurements seen in [Fig. 5] are performed at different positions on the inhomogeneous profile the absorption susceptibility used in the simulations must be adjusted accordingly:

\[
\begin{align*}
\chi^{\text{abs}}_{\text{peak}} &= \chi^{\text{abs}}_{\text{peak}} \left(\frac{\Gamma/2}{f_0/2 + (\Gamma/2)}\right)^2 \\
\chi^{\text{abs}}_{\text{peak},1} &= [f - f_0 = 0\ GHz] \approx (6.08 \pm 0.55) \times 10^{-4} \\
\chi^{\text{abs}}_{\text{peak},2} &= [f - f_0 = 4\ GHz] \approx (3.39 \pm 0.31) \times 10^{-4} \\
\chi^{\text{abs}}_{\text{peak},3} &= [f - f_0 = 13\ GHz] \approx (0.65 \pm 0.06) \times 10^{-4}
\end{align*}
\]

(4)
low that the outgoing polarization varies from $D_1$. The results from the 1 and 6 mm crystals give similar results that agree with the simulations. Also here the absorption susceptibility must be reduced with respect to the peak value to account for the experiments taking place at the side of the inhomogeneous profile.

\[ \chi_{abs} = (0.65 \pm 0.06) \times 10^{-6} \]

\[ \chi_{abs} = (3.39 \pm 0.31) \times 10^{-6} \]

\[ \chi_{abs} = (6.08 \pm 0.55) \times 10^{-6} \]

MAXIMUM ABSORPTION AXIS

It has previously been shown, see Ref. [10], that slow light effects split an incoming linear polarization into two parts, where one part has a significantly larger slow light effect than the other. The transition dipole moment direction was then considered as the maximum absorption direction but it turns out that the maximum (minimum) absorption and consequently time delay happens when we set the input polarization along the $D_2$ ($D_1$) axis. In general this depends on the ratio $R$ in Eq. (2) since if $R \ll 1$ the maximum absorption direction is almost exactly along the transition dipole moment axis, but as stated this is not the case for Pr$^{3+}$:Y$_2$SiO$_5$ where $R \approx 85 \gg 1$.

CONCLUSION

In conclusion, we have modeled and analyzed the polarization of light propagating through an absorbing (amplifying) birefringent crystal. Steady state solutions for the polarization of a propagating wave were found and discussed. This led to the conclusion that only increasing the length of a birefringent medium (e.g. a rare-earth-ion-doped crystal) is not the most efficient method to reach higher absorption. This can be an important issue in applications like quantum memories and spectral filtering employing birefringent crystals [3-6]. Since the model works for any birefringent material with either absorption or amplification it can be useful in other applications such as laser crystal gain media or with other materials that are not specifically discussed here.

It also became clear that the polarization direction with maximum absorption (gain) depends upon the ratio between the phase retardation and the magnitude of the absorption (gain). For an absorbing Pr$^{3+}$:Y$_2$SiO$_5$ crystal the maximum absorption axis is along $D_2$ while the stable polarization steady state solution is almost completely along $D_1$.

This work will hopefully open up new opportunities to investigate more interesting physics and applications regarding the propagation effects in rare-earth-ion-doped crystals in the future.

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**SUPPLEMENTARY INFORMATION**

Light propagation in a birefringent crystal

The propagation of an electromagnetic field in a general material containing absorption can be a quite complex problem to solve due to the interaction of light and matter, but one could reduce this complexity without loosing the generality of the solution by considering a simplification based on the properties of the medium. We start the simplification by considering a plane-stratified medium, which is a valid assumption for the materials investigated in our case. The assumption of plane-stratified media is a proper approximation for materials that show variation in the propagation direction (z-axis) but no variation in the lateral (x-y plane) direction. In other words, we have homogeneity (same absorption or gain) in the x-y plane and possible inhomogeneity (varying absorption or gain) along the z-axis. The main theoretical concept of the calculations in this paper is discussed with more details in Ref. [2].

For macroscopic media, Maxwell’s equations describe the dynamics of the fields as follows (the harmonic time convention $e^{-iωt}$ is used):

\[
\begin{align*}
\nabla \times \mathbf{E}(\mathbf{r}, ω) &= ik_0(ε_0\mathbf{B}(\mathbf{r}, ω)) \\
\nabla \times η_0\mathbf{H}(\mathbf{r}, ω) &= -ik_0(ε_0η_0\mathbf{D}(\mathbf{r}, ω))
\end{align*}
\]

where $η_0 = \sqrt{\frac{μ_0}{ε_0}}$ is the intrinsic impedance of vacuum, $c_0 = 1/ε_0μ_0$ and $k_0$ are the speed and wave number of light in vacuum, respectively. In addition, $\mathbf{E}$, $\mathbf{B}$, $\mathbf{H}$, and $\mathbf{D}$ are the electric field, magnetic flux density, magnetic field and the electric flux density, respectively.

In order to apply Maxwell’s macroscopic equations, it is necessary to specify the relations between $\mathbf{E}$, $\mathbf{B}$, $\mathbf{H}$, and $\mathbf{D}$. These equations are called the constitutive relations.

\[
\begin{align*}
\mathbf{D}(\mathbf{r}, ω) &= ε_0[ε(\mathbf{z}, ω) \cdot \mathbf{E}(\mathbf{r}, ω) + η_0ξ(\mathbf{z}, ω) \cdot \mathbf{H}(\mathbf{r}, ω)] \\
\mathbf{B}(\mathbf{r}, ω) &= \frac{1}{μ_0}[ξ(\mathbf{z}, ω) \cdot \mathbf{E}(\mathbf{r}, ω) + η_0μ(\mathbf{z}, ω) \cdot \mathbf{H}(\mathbf{r}, ω)]
\end{align*}
\]

where $\mathbf{r} = x\hat{x} + y\hat{y} + z\hat{z}$ and $ω$ is the angular frequency. $ε$ and $μ$ are the permittivity dyadic and permeability dyadic of the medium, respectively, while $ξ$ and $ζ$ are called crossed magneto-electric dyadics. All four dyadics and all four fields can depend on the angular frequency $ω$, but this variable is suppressed below to simplify the notation.

In a single crystal, the physical and mechanical properties can often be orientation dependent. When the properties of a material vary with orientation, the material is said to be anisotropic. Alternatively, when the properties of a material are the same in all directions, the material is said to be isotropic. Bi-anisotropic is a general class of linear media which exhibit so-called magneto-electric coupling between the electric and magnetic fields.

Based on the plane-stratified assumption, it is natural to decompose the electromagnetic field into tangential (x-y plane) and normal components. Considering the lateral homogeneity and substituting the constitutive relations (Eq. (6)) into Maxwell’s equations (Eq. (5)) gives a system of ordinary differential equations (ODEs) with the variable $z$.

\[
\frac{d}{dz} \left( \eta_0 J \cdot H_{xy}(z) \right) = ik_0 M(z) \cdot \left( \eta_0 J \cdot H_{xy}(z) \right) \tag{7}
\]

where $J$ is a two-dimensional rotation dyadic (rotation of 90° in the x-y plane) as follows:

\[
J = \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix}
\]

and

\[
J \cdot J = -I_2 = \begin{pmatrix} -1 & 0 \\ 0 & -1 \end{pmatrix}
\]

For a more detailed discussion of how to obtain Eq. (7) which is beyond the scope of this paper, the reader is referred to Ref. [2].

According to the main assumption, the plane-stratified media assumption, the four dyadics $ε(z), ξ(z), ζ(z)$, and $μ(z)$ in Eq. (6) depend only on one spatial variable, $z$, which describes the propagation distance. The Pro$_3+\cdot$Y$_2$SiO$_5$ crystal, which is our medium, is appropriately modeled as a non-magnetic anisotropic medium with zero crossed magneto-electric dyadics. Therefore, as a second assumption, we assume non-magnetic properties ($μ = 1$) and also no crossed magneto-electric dyadics ($ξ = ζ = 0$) in Eq. (6). There exists a general set of equations for the connection between the fundamental matrix $M(z)$ and the four main dyadics in Ref. [2], but based on the three assumptions described above and a normal incident we can simplify $M(z) = M$ for our case as:

\[
M = \begin{pmatrix} \bar{0} & -\bar{1} \\ -\bar{e}_{⊥⊥} & \bar{ε}_{⊥} \bar{ε}_{z} \bar{I}_2 \end{pmatrix}
\]

where

\[
\begin{align*}
\bar{e}_{⊥⊥} &= (ε_{xx} \quad ε_{xy}) \\
\bar{e}_{⊥} &= (ε_{xz}) \\
\bar{ε}_{z} &= (ε_{zx} \quad ε_{zy}) \\
\bar{I}_2 &= \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}
\end{align*}
\]

M tensor eigenvalues and eigenvectors

Following Eq. (7), we need to investigate the eigenvalues and eigenvectors of the $M$ tensor in order
to calculate the tangential components of the electric and magnetic field. The eigenvectors \( u_i \) and \( v_i \) correspond to the eigenvalues \( +\lambda_i \) and \( -\lambda_i \) (\( i = 1, 2 \)) for the forward and backward waves, respectively:

\[
\begin{align*}
  u_i &= \{ E_{ix}, E_{iy}, -\eta_0 H_{iy}, \eta_0 H_{ix} \} \\
  v_i &= \{ -E_{ix}, -E_{iy}, -\eta_0 H_{iy}, \eta_0 H_{ix} \}
\end{align*}
\quad \text{for } i = 1, 2
\]

(12)

For a full analytical derivation of the eigenvalues and eigenvectors see section “Theoretical analysis of the propagation matrix \( M \”).

### E-field evolution in the propagation direction

To understand the E-field and H-field properties while the wave is propagating in the \( z \) direction, we can employ the fundamental equation for one-dimensional wave propagation [Eq. (7)]. By considering the eigenvectors and eigenvalues of the \( M \) matrix [Eq. (12)], the wave properties along the propagation direction \( (z) \) is calculated for the forward wave:

\[
\frac{d}{dz} u_i = ik_0 \lambda_i u_i = u_i(z) = u_i(0)e^{ik_0 \lambda_i z}
\]

(13)

It is possible to calculate the backward wave, \( v_i(z) \), in the same way.

By convention, the polarization of light is described by specifying the orientation of the wave’s electric field at a point in space over one period of the oscillation. Therefore, to understand the polarization direction inside the medium it is enough to only derive the E-field. The initial light polarization before interaction with the medium could be described by the E-field:

\[
E_0 = E_{0x}\hat{x} + E_{0y}\hat{y}
\]

(14)

In addition, the transverse electric field \((x\cdot y)\) components inside the medium could be written as a sum of the E-field of the two eigenvectors:

\[
E_{\text{inside}} = A E_1(z) + B E_2(z) = A(E_{1x}\hat{x} + E_{1y}\hat{y}) + B(E_{2x}\hat{x} + E_{2y}\hat{y})
\]

(15)

Based on the boundary condition for the tangential E-fields at the interface:

\[
\begin{align*}
  \hat{x} : E_{0x} &= AE_{1x} + BE_{2x} \\
  \hat{y} : E_{0y} &= AE_{1y} + BE_{2y}
\end{align*}
\]

(16)

Therefore

\[
\begin{align*}
  A &= \frac{E_{0x} E_{2y} - E_{0y} E_{2x}}{E_{1x} E_{2y} - E_{1y} E_{2x}} \\
  B &= \frac{1}{E_{2x}}[E_{0x} - AE_{1x}]
\end{align*}
\]

(17)

Now we can extend our fields to a different \( z \) using Eq. (13) Eq. (15) and Eq. (17) as follows:

\[
u(z) = Au_1 e^{ik_0 \lambda_1 z} + B u_2 e^{ik_0 \lambda_2 z}
\]

(18)

Note that since the \( M \) matrix does not depend on \( z \) in this case the eigenvalues and eigenvectors \( +\lambda_i \), \( -\lambda_i \), \( u_i \) and \( v_i \) remains constant and it is therefore possible to propagate the fields (or equivalently \( u(z) \)) in Eq. (18) to any desired length in one step. In the more general case where the eigenvalues and eigenvectors changes with \( z \) one has to recalculate these in the simulations every distance \( \Delta z \) to be able to propagate the fields to \( z \), where \( \Delta z \) is chosen for numerical stability and resolution.

Note also that the two polarization steady states for a forward propagating wave discussed in the main text is simply \( u_1 \) and \( u_2 \) where the solution with the lowest imaginary part of its eigenvalue is the stable solution whilst the other is the unstable solution. This can be understood since a high absorption leads to a fast decay from that state, i.e. the highest absorption gives the unstable state. In the same way, a large negative imaginary part corresponding to a high gain will generate a large component and therefore become stable since the other component can be neglected.

### Light propagation in a \( \text{Pr}^{3+}:Y_2\text{SiO}_5 \) crystal

We start by calculating the permittivity tensor for \( \text{Pr}^{3+}:Y_2\text{SiO}_5 \) and then employ the theoretical approach discussed above to obtain the light polarization direction while propagating through this specific medium.

To define the permittivity tensor for \( \text{Pr}^{3+}:Y_2\text{SiO}_5 \) one could start by deriving the relation between permittivity and susceptibility for the host material \((Y_2\text{SiO}_5)\) and the absorber (Pr) separately as follows:

\[
\epsilon = (1 + \chi) = (1 + \chi_{\text{host}} + i\chi_{\text{abs}})
\]

(19)

The imaginary part of the susceptibility is proportional to the absorption, while the real part of the susceptibility is proportional to the real refractive index.

In section “Theoretical analysis of the propagation matrix \( M \)” a full analytical derivation of the electric susceptibility \( \chi_{\text{abs}} \) and the transition dipole moment angle \( \theta \) is given but the following equations [Eq. (20) and Eq. (21)] are approximately correct for the case of \( \text{Pr}^{3+}:Y_2\text{SiO}_5 \).

\[
\chi_{\text{abs}} = \frac{n_{bg} c_0}{\omega}(\alpha D_1 + \alpha D_2)
\]

(20)

where \( n_{bg} \approx 1.8 \) is a background refractive index and \( \omega \approx 2\pi \cdot 494 \cdot 10^{12} \text{ rad/s} \).
The anisotropic absorption coefficients for the two sites of a nominally 0.05% Pr\textsuperscript{3+}:Y\textsubscript{2}SiO\textsubscript{5} crystal at the $^3H_4 \rightarrow 1D_2$ transition is measured as shown in the Table 7.6 in Ref. [8]. Based on this measurement the absorption for site I (605.977 nm) is about 3.6 ± 0.5 along the $D_1$, 47 ± 5 along the $D_2$, and < 0.1 along the $b$ axis, all in the unit cm\textsuperscript{-1} (see Fig. 2a, where $D_1$, $D_2$ and $b$ are crystal principal axes). To estimate the susceptibility and the transition dipole moment angle $\theta$, Eq. (20) and the following equation are used (for more correct values see Eq. (44)).

\[
\begin{align*}
\tan(\theta)^2 &= \frac{\alpha_{D_1}}{\alpha_{D_2}} \\
\Rightarrow \theta &= 74.5 \pm 1.9^\circ \\
\Rightarrow \chi_{abs} &= (8.8 \pm 1.0) \cdot 10^{-4}
\end{align*}
\]

where $\chi_{abs}$ is given in the direction of the transition dipole moment shown in Fig. 2a. It can be rotated about the $b$ axis to the principal axis coordinate system to be able to perform the summation in Eq. (19).

\[
\chi_{abs}^{\text{crystal}} = R_z^{-1} \cdot \chi_{abs}^{\text{dipole}} \cdot R_z
\]

\[
= \chi_{abs} \begin{pmatrix}
\cos^2 \theta & \sin \theta \cos \theta & 0 \\
\sin \theta \cos \theta & \sin^2 \theta & 0 \\
0 & 0 & 0
\end{pmatrix}
\]

The next step is to calculate $\chi_{host}$ based on the Sellmeier dispersion and the measured coefficients from Table 7.1 and Eq. 7.2 in Ref. [8].

\[
\chi_{host} = \begin{pmatrix}
2.1973 & 0 & 0 \\
0 & 2.2726 & 0 \\
0 & 0 & 2.1867
\end{pmatrix}
\]

Adding up all calculations, the final results for the permittivity tensor in the principal axis coordinate will be:

\[
\epsilon = \begin{pmatrix}
\epsilon_{xx} & \epsilon_{xy} & \epsilon_{xz} \\
\epsilon_{yx} & \epsilon_{yy} & \epsilon_{yz} \\
\epsilon_{zx} & \epsilon_{zy} & \epsilon_{zz}
\end{pmatrix}
\]

Solving for the eigenvalues of $M$ gives four eigenvalues:

\[
\lambda_i = \pm \left( n_{D_1}^2 + n_{D_2}^2 + i\chi_{abs} \right) \pm \chi_{abs} \sqrt{ \left( n_{D_2}^2 - n_{D_1}^2 \right)^2 - 2i(1 - 2\sin^2 \theta) \left( n_{D_2}^2 - n_{D_1}^2 \right) \frac{n_{D_2}^2 - n_{D_1}^2}{\chi_{abs}} - 1 }^{1/2}
\]

Since we are only interested in the forward propagating waves only the eigenvalues whose real part is positive are of interest. These eigenvalues are denoted $\lambda_1$ and $\lambda_2$.

Before we solve for the eigenvectors in the general case we have to account for some special cases where $\theta = 0^\circ$ or $\theta = 90^\circ$. In these cases the transition dipole moment axis is parallel to one of the crystal axes $D_1$ or $D_2$ which of course results in two steady states, one along $D_1$ and another along $D_2$. Another special case is when the absorption is zero, which again is a trivial case where the
polarizations of the steady states lie along $D_1$ and $D_2$.

Now that these special cases have been solved we will solve for the eigenvectors in the general case where we exclude the special cases mentioned above. The forward eigenvectors will be called $\mathbf{u}_i$:

The eigenvector equations we want to solve are:

\[
\begin{pmatrix}
0 & 0 \\
-\eta_0^2 D_1 - i\chi_{\text{abs}} \cos^2 \theta & -\eta_0^2 D_2 - i\chi_{\text{abs}} \sin^2 \theta \\
-\eta_0 D_1 \cos \theta \sin \theta & -\eta_0 D_2 \cos \theta \sin \theta
\end{pmatrix}
\begin{pmatrix}
E_{ix} \\
E_{iy}
\end{pmatrix}
= \lambda_i
\begin{pmatrix}
E_{ix} \\
E_{iy}
\end{pmatrix}
\]

The first two equations provides a relationship between the electric and magnetic fields and are omitted here since we are only interested in the ratio between the $D_1$ and $D_2$ components of the electric field to get the steady state solution, i.e. we only have to solve for the first two components of the eigenvector. This leaves us with two equations:

\[
\begin{align*}
(n_1^2 + i\chi_{\text{abs}} \cos^2 \theta)E_{ix} + (i\chi_{\text{abs}} \cos \theta \sin \theta)E_{iy} &= \lambda_i^2 E_{ix} \\
(i\chi_{\text{abs}} \cos \theta \sin \theta)E_{ix} + (n_2^2 + i\chi_{\text{abs}} \sin^2 \theta)E_{iy} &= \lambda_i^2 E_{iy}
\end{align*}
\]

Since we have the option to normalize the eigenvector

\[
\frac{E_{ix}}{E_{iy}} = \frac{2 \cos \theta \sin \theta}{1 - \cos^2 \theta - iR \mp \sqrt{1 - (1 - 2 \sin^2 \theta)^2 + (R + 1 - 2 \sin^2 \theta)^2}}
\]

It can be shown that $\frac{E_{ix}}{E_{iy}} = -\frac{E_{ix}}{E_{iy}}$, which means that the two steady state solutions only differ by a $90^\circ$ rotation in the $D_1-D_2$ plane since the two dimensional rotation matrix for an angle of $90^\circ$ is

\[
\mathbf{J} = \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix}
\]

Analysis when $R \to 0$

We can now solve for the steady state solutions (i.e. electric field components of the eigenvectors $\mathbf{u}_i$) in the limit $R \to 0$, which in other words means that the absorption is high compared to the phase retardation (see Eq. (3)). The eigenvalues becomes:

\[
\begin{align*}
\lambda_1 &\approx \sqrt{\frac{n_1^2 + n_2^2}{2}} \\
\lambda_2 &\approx \sqrt{\frac{n_1^2 + n_2^2}{2}} + i\chi_{\text{abs}}
\end{align*}
\]

i.e. one solution with no absorption (or gain) and a refractive index close to the refractive index of the material and one solution with high absorption (or gain) and a high refractive index.

The solution for the ratio of the electric field components becomes:

\[
\begin{align*}
\mathbf{u}_1(z) &= \mathbf{u}_1(0)e^{i\kappa_0 \lambda_1 z} \\
\mathbf{u}_2(z) &= \mathbf{u}_2(0)e^{i\kappa_0 \lambda_2 z}
\end{align*}
\]
with electric fields and not intensities.

\[
\begin{align*}
\lambda_1 & : \frac{E_{1x}}{E_{1y}} \approx -\tan(\theta) \\
\lambda_2 & : \frac{E_{2x}}{E_{2y}} \approx \frac{1}{\tan(\theta)} 
\end{align*}
\]  
(38)

Since these are real numbers the polarizations are linear and the directions are for \( \lambda_2 \) along the transition dipole moment and for \( \lambda_1 \) perpendicular to it. Since the stable solution is the eigenvector whose eigenvalue has the lowest imaginary part, \( \lambda_1 \) is stable for absorption and \( \lambda_2 \) is stable for gain.

**Analysis when \( R \to \infty \)**

In the other limit where \( R \to \infty \) the phase retardation dominates over the absorption which gives the following approximation of the eigenvalues;

\[
\begin{align*}
\lambda_1 & \approx \sqrt{n_{D_1}^2 + i\chi_{abs} \cos^2 \theta } \\
\lambda_2 & \approx \sqrt{n_{D_2}^2 + i\chi_{abs} \sin^2 \theta } 
\end{align*}
\]  
(39)

Here the stable solution depends on the transition dipole moment direction given by \( \theta \). If \( \theta < 45^\circ \), \( \lambda_2 \) is stable while if \( \theta > 45^\circ \), \( \lambda_1 \) is stable for absorption and the reverse is true for gain. Using Eq. (32) we can solve for the eigenvectors in the limit when \( R \to \infty \).

\[
\begin{align*}
\lambda_1 & : (n_{D_1}^2 + i\chi_{abs} \cos^2 \theta )E_{1x} + (i\chi_{abs} \cos \theta \sin \theta )E_{1y} \\
& \approx (n_{D_1}^2 + i\chi_{abs} \cos^2 \theta )E_{1x} \\
& \Rightarrow E_{1y} \approx 0 \\
\lambda_2 & : (i\chi_{abs} \cos \theta \sin \theta )E_{2x} + (n_{D_2}^2 + i\chi_{abs} \sin^2 \theta )E_{2y} \\
& \approx (n_{D_2}^2 + i\chi_{abs} \sin^2 \theta )E_{2y} \\
& \Rightarrow E_{2x} \approx 0 
\end{align*}
\]  
(40)

From these equations it is clear that the eigenvector corresponding to \( \lambda_1 \) has almost no \( E_{1y} \) component and is therefore along \( D_1 \) and the reverse is true for \( \lambda_2 \).

**Analysis of \( \text{Pr}^{3+}:Y_2\text{SiO}_5 \)**

In 0.05% \( \text{Pr}^{3+}:Y_2\text{SiO}_5 \) \( R \) is quite high so we can use the approximations made in the previous section for when \( R \to \infty \). Continuing from Eq. (39) we can write the imaginary part of \( \lambda_i \) as;

\[
\begin{align*}
\text{Im}(\lambda_1) & \approx \frac{1}{2} \sqrt{2n_{D_1}^4 + \chi_{abs} \cos^4 \theta - 2n_{D_1}^2} \\
\text{Im}(\lambda_2) & \approx \frac{1}{2} \sqrt{2n_{D_2}^4 + \chi_{abs} \sin^4 \theta - 2n_{D_2}^2} 
\end{align*}
\]  
(41)

In \( \text{Pr}^{3+}:Y_2\text{SiO}_5 \) the exponential decays of the intensity along \( D_1 \) and \( D_2 \) are measured and they are \( \alpha_{D_1} = 3.6 \pm 0.5 \text{ cm}^{-1} \) and \( \alpha_{D_2} = 47 \pm 5 \text{ cm}^{-1} \) respectively. Given that the eigenvectors calculated in Eq. (40) are along \( D_1 \) and \( D_2 \) respectively one can, using Eq. (30) write the following equation;

\[
\begin{align*}
\text{Im}(\lambda_1) & = \frac{\alpha_{D_1}}{2\omega_0} \\
\text{Im}(\lambda_2) & = \frac{\alpha_{D_2}}{2\omega_0} \\
k_0 & = \frac{2\pi}{\lambda_0} 
\end{align*}
\]  
(43)

where the factor of 1/2 comes from that we now deal with electric fields and not intensities. \( k_0 \) is the wave number and \( \lambda_0 \) is the wavelength of the incoming wave in vacuum (not to be confused with \( \lambda_1 \) and \( \lambda_2 \) which are the eigenvalues of \( M \)). Using Eq. (42) and Eq. (43) one can calculate the angle \( \theta \) from \( D_1 \) to the transition dipole moment axis and the absorption susceptibility \( \chi_{abs} \) for a \( \text{Pr}^{3+}:Y_2\text{SiO}_5 \) crystal using \( \lambda_0 = 605.977 \text{ nm} \), \( n_{D_1} = 1.7881 \) and \( n_{D_2} = 1.809 \).

\[
\begin{align*}
\theta & = 74.6 \pm 1.9^\circ \\
\chi_{abs} & = (8.82 \pm 0.8) \times 10^{-4} 
\end{align*}
\]  
(44)

An approximation of \( \theta \) and \( \chi_{abs} \) in Eq. (42) can be made by assuming that \( \text{Im}(\lambda_1) \ll n_{D_1} \) and that \( n_{D_1} \approx n_{D_2} \approx n_{bg} \) (which is the case for \( \text{Pr}^{3+}:Y_2\text{SiO}_5 \)). This results in;
\[
\theta \approx \arctan\left(\frac{(\text{Im}(\lambda_2))^2 n_D^2}{(\text{Im}(\lambda_1))^2 n_D^2}\right)^{1/4}
\]

\[
\approx \arctan\left(\frac{\text{Im}(\lambda_2)}{\text{Im}(\lambda_1)}\right) = \arctan\left(\frac{\alpha D_2}{\alpha D_1}\right)
\]

and

\[
\chi_{\text{abs}} \approx 2\text{Im}(\lambda_1)n_D_1 + 2\text{Im}(\lambda_2)n_D_2
\]

\[
= \frac{\alpha D_1 n_D_1}{k_0} + \frac{\alpha D_2 n_D_2}{k_0} \approx \frac{(\alpha D_1 + \alpha D_2) n_{\text{bg}}}{k_0}
\]

(Eq. 45) and (Eq. 46) are only valid in the case where \( R \to \infty \) but allows us to use (Eq. 20) and (Eq. 21) for the case of \( \text{Pr}^{3+}:Y_2\text{SiO}_5 \) which gives values quite close to the more correct values obtained in (Eq. 44).

Continuation of analysis when \( R \to 0 \)

In the other case where \( R \to 0 \) we only have one axis with absorption (see (Eq. (37))) and therefore in (Eq. (43)) we replace \( \alpha D_2 \) by \( \alpha_{\text{dipole}} \) that measures the absorption along the transition dipole moment axis for \( \lambda_2 \) whose eigenvector points in that direction and set \( \text{Im}(\lambda_1) = 0 \). \( \theta \) is now given by the angle from the crystal axis to the axis of which the absorption is measured. Assuming that \( \chi_{\text{abs}} \gg n_D^2 \), the following proportionality between \( \chi_{\text{abs}} \) and \( \alpha_{\text{dipole}} \) can also be calculated using (Eq. (37))

\[
\chi_{\text{abs}} \approx \frac{1}{2} \left(\frac{\alpha_{\text{dipole}}}{k_0}\right)^2
\]

(Eq. 47)

This shows that the electric susceptibility \( \chi_{\text{abs}} \) does not always scale linearly with the absorption coefficient \( \alpha_{\text{dipole}} \).