Optical switching and inversionless amplification controlled by state-dependent alignment of molecules

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Abstract

Switching anisotropic molecules from strongly absorbing to strongly amplifying through a transparent state is shown to be possible by the application of dc or ac control electric fields without the requirement of population inversion. It is based on decoupling of the lower level molecules from the resonant light while the excited ones remain emitting due to their state-dependent alignment. The amplification index may become dependent only on a number of excited molecules and not on the population of the lower state. A suitable class of molecules and applications in optoelectronics, fibreoptics and nanophotonics are outlined.

1. Introduction

The feasibility of manipulating the transparency of materials from a strongly absorbing state to amplifying via absolute transparency without a change of the populations of the coupled energy levels is an alluring prospect because of numerous important applications. Amplification of light is determined by the difference between the net absorption and stimulated emission of radiation. Usually, this requires a larger population of the upper energy level than the lower one. However, the amount of absorbed and emitted photons depends not only on the populations of the resonant energy levels but also on the probabilities of the induced transitions and on the distribution of the populations over the energy-degenerated states. Thus, population inversion is not the only option for the amount of coherently emitted light to prevail over the absorbed one. A variety of processes that may lead to nonreciprocity of emission and absorption were investigated in the early years of quantum electronics. Among them are those based on the difference in velocity distributions in the upper and lower states [1, 2], on nonlinear interference effects (NIE) at quantum transitions in the field of the auxiliary radiation [3, 4] in two-level
atoms [5], and the related feasibility of inversionless amplification in three-level atoms [6]. The feasibility of manipulating the transmission of light from attenuation to amplification without population inversion (AWI) via transparency based on NIE in three-level atoms was investigated in [7–9] and illustrated with numerical examples for transitions of Ne driven by a He–Ne laser at the adjacent transitions. It was experimentally proved for the transition 2p4–2s2 of Ne probed by the $\lambda = 1.15 \text{\mu m}$ He–Ne laser, whereas the adjacent transition 2s2–2p1 was driven by another $\lambda = 1.5 \text{\mu m}$ He–Ne laser [10]. Possible nonreciprocity at the transitions to the autoionizing states and related AWI were discussed in [11]. The feasibility of AWI for short pulses in a bichromatically excited double-lambda scheme was investigated in [12]. A review of later publications on AWI is given in [13]. An asymmetry in the absorption and emission lineshapes of a two-level system caused by the interaction with a thermostat is discussed in [14]. In this paper, a different type of nonreciprocity in the absorption and emission processes is explored. It is associated with the energy-level-selective alignment of molecules [15, 16]. An efficient optical switching from the strong absorption to the transparency and to AWI is shown to be achieved through decoupling of the lower level molecules from polarized resonant radiation, while the upper level molecules remain amplifying. A suitable class of molecules and applications in optoelectronics, fibreoptics and nanophotonics are discussed. The proposed feasibilities are illustrated with numerical models.

2. Absorption index controlled by external electric field

In the dipole approximation, the probability of induced transitions between an upper level $m$ and lower one $g$, $w_{mg}$, depends on the squared modulus of the projection of the dipole transition matrix element on a vector of the resonant electric field $E$ which causes such a transition, $|d_{mg}|^2 \cos^2 \theta$,

$$w_{mg} = B|E|^2 F(\omega), \quad B = 8\pi |d_{mg}|^2 \cos \theta / \hbar^2.$$  \hspace{1cm} (1)

Here, $B$ is the Einstein coefficient, $F(\omega)$ is the frequency dependence (the transition spectral form factor) and $\omega$ is the frequency of radiation. For a molecule, the angle $\theta$ is determined by its orientation in space and by its symmetry. Under the influence of an auxiliary control electric field $E_0$, the molecule turns towards the direction of the minimum of the interaction energy. Hence, the degree of alignment of a molecular medium depends on the alignment parameter, which is given by the ratio of interaction energy with the control field in the quantum state $j$ to the energy of thermal motion $kT$ that renders disorientation. However, the energy of such interaction $U_j$, and consequently the degree of orientation, can be different for molecules in the lower and upper energy levels. Therefore, in such a case, the probabilities of induced emission and absorption of polarized light, averaged over the molecules with different orientations, are not equal. This enables inversionless amplification of the polarized light through manipulating the difference in the orientation degree of the upper and lower level molecules with the aid of the auxiliary dc or ac control fields without a change in their energy-level populations.

In the presence of a control field, the amplification index $\alpha > 0$ for a linear-polarized probe radiation, $E$, is given by

$$\alpha = \sigma_0 \int \left[ n_m f_m(\Theta, E_0) - n_g f_g(\Theta, E_0) \right] \cos^2 \theta \, d\Theta.$$  \hspace{1cm} (2)

Here, $n_m$ is the orientation-integrated population of the upper and $n_g$ of the lower level, $d\Theta = \sin \theta \, d\Theta \, d\phi$ is the element of solid angle and $\sigma_0 = 8\pi |d_{mg}|^2 \omega F(\omega) / c\hbar$ is the absorption cross-section for molecules, whose transition dipole moment is aligned along $E$. Excitation of molecules can be accomplished in any common way, such as incoherent excitation or
others employed, e.g., for dye and solid-state lasers. The functions \( f_{m,g}(\Theta, E_0) \) depict the distributions of molecules over orientations in the corresponding levels. It is readily seen that their difference provides the feasibility of AWI. If the energy level lifetime exceeds significantly the time required to set the orientation balance, they are given by the Boltzmann distribution

\[
f_j(\Theta, E_0) = A_j \exp\left(-U_j(\Theta, E_0)/kT\right).
\]

Here, \( A_j^{-1} = \int \exp\left(-U_j(\Theta, E_0)/kT\right) d\Theta \) is a scale factor, \( T \) is the temperature, \( k \) is the Boltzmann constant and \( j = \{g, m\} \). The potential energy of a molecule coupled with the electric field \( E_0 \) can be presented as

\[
U_j = -\mu_j^i E_0^i - \alpha_{jk}^i E_0^k E_0^i/2.
\]

Here, \( \mu_j^i \) is the \( i \)th component of the permanent dipole moment and \( \alpha_{jk}^i \) is the component of the tensor of electrical polarizability, both for the molecules in the energy level \( j \); and \( E_0 \) is the \( i \)th component of the control field. The first term in equation (4) represents the energy of molecules without a centre of symmetry, which possess a permanent dipole moment. The second term depicts the interaction energy with the dipole induced by the field \( E_0 \). This term presents the alignment caused by either a strong dc or ac, e.g., laser field. For a review of the laser alignment, including recent experiments, see [26–30] and references therein.

Consider axial-symmetric molecules and further assume the directions of both the permanent \( \mu \) and the transition moment \( d_{mg} \) are aligned along the symmetry axis of the molecule, which makes an angle \( \theta_0 \) with the control field \( E_0 \). Then the interaction energy (4) and distribution functions (5) become dependent only on \( \theta_0 \):

\[
f_j(\Theta, E_0) = f_j(\theta_0, E_0) = A_j \exp\{p_j \cos \theta_0 + q_j \cos^2 \theta_0\}.
\]

Following [17, 18], we introduce the alignment parameters attributed to the permanent dipole orientation \( p_j \) and to the polarizability ellipsoid \( q_j \) as

\[
p_j = \mu_j^i E_0^i/kT, \quad q_j = (\alpha_{33}^j - \alpha_{11}^j)|E_0|^2/2kT.
\]

Here, \( \alpha_{33}^j \) and \( \alpha_{11}^j \) are the principal values of the polarizability tensor along and across the symmetry axis, accordingly, for the molecule in level \( j \). They are calculated as [17, 18]

\[
\alpha^j = \frac{2}{\hbar} \sum_i \frac{\omega_{lj}^j |d_{lj}^j|^2}{\omega_{lj}^j - \omega_0^j} = \frac{e^2}{m} \sum_i \frac{f_{lj}}{\omega_{lj}^j - \omega_0^j},
\]

where \( d_{lj} \) is the transition dipole moment along or across the molecule symmetry axis accordingly between the corresponding levels, \( f_{lj} \) is the corresponding oscillator strength, \( \omega_{lj} \) is the transition frequency and \( \omega_0 \) is the frequency of \( E_0 \).

3. Molecules with large ground-state permanent dipole moment

Consider axial-symmetric molecules whose alignment is primarily determined by the permanent dipole moment. If such a moment in the lower state is larger than in the upper one \( (\mu_g > \mu_m) \), the alignment degree in the lower state is larger than for molecules in the upper state. Consequently, the orthogonal orientation of the probe \( E \) and control \( E_0 \) fields is advantageous for suppression of the absorption and for enhancement of the emission. The orientation-averaged absorption/amplification index in the case of orthogonal orientation of
Figure 1. Scaled amplification index $\alpha_\perp/\alpha_g$ versus alignment parameter $p_g = \mu_g E_0/kT$. $n_m = 0.8 n_g$. $\mu_g = 4 \mu_m$. The orientation of the molecule symmetry axis $C$ is depicted in the inset.

Figure 2. Scaled amplification index versus alignment parameter $p_g = \mu_g E_0/kT$ and ratio of the excited- and ground-state permanent dipole moments $n_\mu = \mu_\mu/\mu_g$ for the case of $\mu_g > \mu_m$ and orthogonal orientation of the control and probe fields. $n_m = 0.8 n_g$.

The probe and the control fields are calculated as

$$
\frac{\alpha_\perp}{\sigma_\alpha_n} = \int_0^{2\pi} d\varphi \cos^3 \varphi \int_0^\pi d\theta_0 \sin^3 \theta_0 \left[ \frac{n_m}{n_g} A_m \exp\{p_m \cos \theta_0\} - A_g \exp\{p_g \cos \theta_0\} \right] = \frac{n_m L(p_m)}{n_g L(p_g)} - \frac{L(p_g)}{p_g},
$$

where $L(p_j)$ is the Langevin function [17–20]

$$
L(p_j) = \left(1 - \frac{2}{p_j}\right)^{-1} \int_0^{2\pi} d\varphi \int_0^\pi d\theta_0 A_j \exp\{p_j \cos \theta_0\} \cos^2 \theta_0 \sin \theta_0 = \frac{\int_0^\pi d\theta_0 \exp\{p_j \cos \theta_0\} \cos \theta_0 \sin \theta_0}{\int_0^\pi d\theta_0 \exp\{p_j \cos \theta_0\} \sin \theta_0} = \coth p_j - \frac{1}{p_j}.
$$

Figures 1–3 depict features of the optical switching from absorption to inversionless amplification in this case. Figure 1 illustrates the case where the upper-state permanent dipole moment, $\mu_m$, is four times smaller than the one in the lower state, $\mu_g$, $\mathbf{E}_\perp \mathbf{E}_0$, and the
population of the upper energy level is 20% less than that of the lower one. It shows that the absorption decreases with increasing control field $E_0$ and with decreasing temperature. The sample becomes transparent in the range of the orientation parameter $p_\parallel$ between 1 and 2. Then the probe radiation is amplified at any strength of $E_0$ above this threshold value (see also figure 2). The threshold value depends on the population ratio, $n_m/n_g$ (see figure 3). The amplification index reaches its maximum for the interval of $p_\parallel$ between 5 and 10. In the given specific case, the maximum value of the amplification factor makes up $\alpha_{max} \approx 0.1\sigma_0n_g = 0.1\sigma_0n/1.8$, where $n$ is the total molecular number density. Because the absorption index at the control field and the excitation of the upper level turned off is $\alpha(E_0 = 0) = -n\sigma_0/3$, this corresponds to a pretty strong AWI. Maximum values of AWI grow with a decrease of the ratio $\mu_m/\mu_g$ (figure 2) and with an increase of the amount of excited molecules, $\eta_n$ (see figure 3).

At $\mu_m = 0$ and $p_\parallel \to \infty$ equation (8) reduces to

$$\alpha_\perp = n_m\sigma_0/3, \quad (\alpha_\perp/\sigma_0n_g = \eta_n/3).$$

In this case, amplification is determined only by the upper-state molecules and does not depend on the population of the lower state. This is because the lower-state molecules are aligned orthogonal to the probe field and, therefore, are not coupled with this field. In contrast, the excited molecules are decoupled from the control field $E_0$. Their orientation remains isotropic and, hence, the averaged projection of the transition dipole moment $d_{mg}$ on the probe field $E$ is not equal to zero.

4. Molecules with large excited-state permanent dipole moment

In the alternative case, $\mu_m > \mu_g$, the polarization $\mathbf{E}$ along $E_0$ appears advantageous because it ensures stronger coupling of the probe field with the upper-state molecules than with the ground-state ones. The orientation-averaged absorption/amplification index is calculated...
Figure 4. Scaled amplification index $\alpha_{\uparrow\uparrow}/\sigma_g$ ($\sigma_g = n_g \sigma_0$) versus alignment parameter $p_g$ for $n_m = 0.8n_g$, $\mu_m = 4\mu_g$. The orientation of the molecule symmetry axis $C$ is depicted in the inset.

Figure 5. Scaled amplification index versus alignment parameter $p_g = \mu_g E_0/kT$ and ratio of the excited- and ground-state permanent dipole moments $\eta_{\mu}^{-1} = \mu_E / \mu_m$ for the case of $\mu_m > \mu_g$ and parallel orientation of the control and probe fields. $n_m = 0.8n_g$.

Accordingly as

$$\frac{\alpha_{\uparrow\uparrow}}{\sigma_0 \mu_g} = \int_0^{2\pi} d\phi \int_0^\pi d\theta_0 \cos^2 \theta_0 \sin \theta_0 \left[ \frac{n_m}{n_g} A_m \exp \{ p_m \cos \theta_0 \} - A_g \exp \{ p_g \cos \theta_0 \} \right]$$

$$= \frac{n_m}{n_g} \left[ 1 - 2 \frac{L(p_m)}{p_m} \right] - \left[ 1 - 2 \frac{L(p_g)}{p_g} \right].$$

(10)

Figures 4–6 depict the switching features in this case, which include inversionless amplification. They display a different dependence of the amplification index on the alignment parameter and on the upper-state populations than in the previous case. Figure 4 depicts the case when the permanent electric dipole moment in the upper state is four times larger than that in the lower state. All other parameters remain the same as in figure 1. Besides switching from absorption to amplification within a relatively narrow interval of the small alignment parameters $p_g \approx 0.05$, figure 4 displays a second interval of more flat switching behaviour in the vicinity of $p_g \approx 6.5$. Figures 5 and 6 show that with $\mu_g \rightarrow 0$, the amplification may

The natural text is a continuation of the provided text, discussing the effects of varying alignment parameters on the amplification index, and includes mathematical expressions and diagrams to illustrate these effects.
become comparable with the absorption corresponding to the control field and excitation of the upper level turned off. The figures also show that the transparency or one and the same magnitude of the amplification index can be achieved for different sets of population and alignment parameters.

5. Molecules without permanent dipole moment

Figures 1–6 depict optical switching with a dc control electrical field for molecules that possess permanent electric dipole moment. Similar conclusions are applied to optical switching and AWI induced by an ac field $E_0$ due to the difference of the induced dipole moments (polarizabilities) in the upper and lower states for axially-symmetric molecules without permanent electric dipole moment. In such a case, equations (8) and (10) take the form

$$\frac{\alpha_{\|}}{\sigma_0 n_g} = \int_0^{2\pi} d\varphi \cos^2 \varphi \int_0^{\pi} d\theta_0 \sin^3 \theta_0 \left[ \frac{n_m}{n_g} A_m \exp\{q_m \cos^2 \theta_0 \} - A_g \exp\{q_g \cos^2 \theta_0 \} \right]$$

$$= \frac{n_m}{n_g} \left[ 1 - L_2(q_m) \right] - \left[ 1 - L_2(q_g) \right], \quad (11)$$

$$\frac{\alpha_{\perp}}{\sigma_0 n_g} = \int_0^{2\pi} d\varphi \int_0^{\pi} d\theta_0 \cos^2 \theta_0 \sin \theta_0 \left[ \frac{n_m}{n_g} A_m \exp\{q_m \cos^2 \theta_0 \} - A_g \exp\{q_g \cos^2 \theta_0 \} \right]$$

$$= \frac{n_m}{n_g} L_2(q_m) - L_2(q_g). \quad (12)$$

Here, $L_2(q)$ is the generalized Langevin function \[17–20\]

$$L_2(q) = \int_0^{\pi} d\theta_0 \cos^2 \theta_0 \sin \theta_0 \exp\{q \cos^2 \theta_0 \} = \left[ 2 \sqrt{q} e^{-q} \int_0^{\sqrt{q}} dt \exp[t^2] \right]^{-1} - \frac{1}{2q}. \quad (13)$$

Parallel orientation of polarizations of the control and probe fields is advantageous for molecules with larger parameter $q$ in the upper state ($q_g < q_m$), whereas the orthogonal orientation is advantageous in the opposite case, $q_m < q_g$. 

![Figure 6. Scaled amplification index versus ratio of the excited- and ground-state permanent dipole moments $\eta^{-1} = \mu_e/\mu_m$, and population ratio $\eta_n = n_m/n_g$ for the case of $\mu_m > \mu_g$ and parallel orientation of the control and probe fields. $p_x = 2$.](image-url)
The important feature of ac alignment is that the sign of the parameter $q$ depends not only on the structure of the molecules but also on the ratio of frequency $\omega_0$ of the control field and that of the strongest transitions contributing to the alignment, $\omega_{ij}$. For molecules stretched along the symmetry axis, $a_{33} > a_{11}$, and $q > 0$ at $\omega_0 < \omega_{ij}$. Alternatively, for disc-type molecules, $a_{33} < a_{11}$, and $q < 0$ at $\omega_0 < \omega_{ij}$. Consequently, in the latter case, the control field decreases the orientation-averaged projection of the induced moment on the vector of the control field. Therefore, the parallel orientation $\mathbf{E}_0 \parallel \mathbf{E}$ of the probe and control fields is optimal for the cases of $0 < q_g < q_m$ and $q_g < q_m < 0$, but in the first case the dependence $\alpha(|\mathbf{E}_0|^2)$ resembles figure 4, whereas in the second case—figure 1. In the opposite case, $q_g > q_m > 0$ and $0 > q_g > q_m$, the orthogonal orientation of the fields $\mathbf{E}_0 \perp \mathbf{E}$ is advantageous, and the dependence $\alpha(|\mathbf{E}_0|^2)$ is qualitatively analogous to figure 1, whereas in the second case to figure 4. The effect is strongest when the signs of $q$ are different in the upper and lower states. This may happen if the signs of the detuning of $\omega_0$ relative to the transitions dominantly contributing to the alignment are different for molecules in the upper and lower states. Then the control field aligns molecules along the direction of its polarization in one state, whereas there is a decrease in the number of such molecules in another coupled state.

6. Suitable molecules and applications

A variety of dye-type molecules possess state-dependent permanent dipole moments and state-dependent polarizability tensors. For example, the molecule 3-6-diacetyl-amino-phthalimide does not possess a permanent dipole moment in the excited state ($\mu_m = 0$), whereas the ground state has $\mu_g \approx 5.5$ D [21]. The important requirement for realization of the proposed technique is that the lifetime in the excited state $\tau$ must exceed the time $\tau_0$ necessary to set the orientation equilibrium. The alignment time varies depending on the molecular size and on the viscosity $\eta$ of the solvent. For large organic molecules in solvents, the characteristic alignment time is $\tau_0 \approx 10^{-10}$–$10^{-12}$ s, for protein macromolecules it is $\tau_0 \approx 10^{-6}$–$10^{-8}$ s and $\tau_0 \approx 10^{-2}$–$10^{-4}$ s for big biomacromolecules. Hence, for the applications under consideration, the molecular mass must not significantly exceed $10^3$ atomic units, and their length should be several nanometres. For such compounds, a permanent electric dipole moment is usually of the order of $1–10$ D [17, 18, 21–23]. In the electrically resistant solvents, an alignment parameter of several units can be achieved for such molecules with a dc field $E_0$ below the breakdown threshold [24]. The breakdown threshold can be increased by the application of pulsed dc fields $E_0$ and by a decrease of the temperature of the solvents, which is possible by using cryogenic liquids or cooled gas. For example, at $\mu_g = 10$ D, $T = 70$ K, the parameter $p = 1$ is achievable at $E_0 \approx 28$ kV mm$^{-1}$. Such a strength of the control field is within the typical range used in laser Stark spectroscopy [25] and can be easily realized, e.g., in hollow fibres. A greater alignment is achievable for molecules having dipole moment of the order of $10^2$–$10^3$ D. However, their reorientation time $\tau_0$ may be comparable or longer than the lifetime in the exited state, which reduces the effect. Thus, optical switching and AWI may appear easier to achieve through orientation of molecules by an ac optical field. The polarizability anisotropy $a_{33}–a_{11}$ is $\approx 10^{-23}$ cm$^3$ even for large resonance detunings $|\omega_{ij} – \omega_0| \sim \omega_{ij}$. At a normal temperature $T = 300$ K, the optical alignment parameter reaches the magnitude $|q| \sim 1$ with the control radiation of $P \sim 10$–$100$ MW focused below the breakdown threshold. The magnitude of $q$ can be considerably increased and the strength of the required control field decreased by setting the frequency $\omega_0$ in the vicinity of the resonant frequencies of the adjacent optical transitions. Alignment of about 70% of molecules ($\langle \cos^2 \theta \rangle \sim 0.75$) has been recently achieved in experiments with short laser pulses [31], and various important applications in
optical physics are foreseen [26]. A possible alignment of linear molecules in hollow fibres is shown in [30].

Besides hollow fibres, the proposed technique can be implemented in nanophotonics and near-field optics, especially associated with plasmonic metamaterials. Such nanostructures as metal islands on a dielectric surface or colloidal metal fractal aggregates provide the presence of nanometre-scale spatial regions of very high concentration of local electric and electromagnetic fields, i.e., ‘hot’ spots. Accordingly, this leads to many-order enhancements for a variety of optical processes, such as photoluminescence, nonlinear absorption and refraction, Raman scattering and four-wave mixing. This facilitates new possibilities for optical microanalysis, spectroscopy of single molecules, and lasing and nonlinear optical processes down to a single molecule. The features associated with the inhomogeneity of the near fields and with the alignment of molecules adsorbed onto metal nanostructures are now opened for investigation, where the experiments carried out to date have provided encouraging results. Giant enhancement of optical effects including lasing at an ultra-low threshold and enhancement of the excitation efficiency up to $10^{11}$ have been observed in rhodamine 6G molecules added to silver colloidal nanoaggregates which were placed inside a quartz microcavity [32–35].

7. Conclusions

In summary, we propose to facilitate the optical switching from strong absorption to transparency, and further to amplification and lasing without the population inversion by the anisotropic molecules through a decoupling of polarized light from the absorbing molecules in the lower energy level while the upper-state molecules stay emitting. Such switching becomes feasible with the aid of a control dc electric or an ac optical field that aligns anisotropic molecules in upper- and lower-energy levels in a different way. The example of a suitable molecule, similar to dye molecules, is given, and the favourable conditions are discussed and numerically illustrated. The achievable magnitudes of amplification without population inversion may reach values that are determined only by the excited-state molecules and do not depend on the population difference. The outlined features promise novel applications of the alignment, such as in optoelectronics, fibreoptics and nanophotonics. Among them are advances in the design of optical micro- and nanoswitches and nanolasers operating on a small number of (or even on individual) molecules adsorbed on metal nanostructures that are placed inside a micro- or nanocavity or inside a hollow fibre. This offers novel applications for control over the combined evanescent and surface-enhanced radiative processes with high-$Q$ morphology-dependent resonances by the auxiliary dc or optical electric fields.

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