

Inversionless amplification of light by molecules selectively oriented over states in the field of laser radiation

A.V. Shamshurin, I.A. Kuzovatov, A.S. Tsipotan, V.V. Slabko

Abstract. A possibility of inversionless amplification of light by non-dipole molecules oriented state-selectively by means of superimposing of external laser radiation is considered. Various variants of mutual direction of electrical vectors of the probe and orienting fields are numerically analysed for specific ratios of the orientation parameters of the ground and excited states both in pulsed and stationary regimes. It is shown that the variant in which the signs of the orientation parameters of the ground and excited states are opposite is the most promising. The latter can be implemented in selection of the orienting field frequency.

Keywords: inversionless light amplification, orientation, non-dipole molecules, exponential fitting.

1. Introduction

In the analysis of the interaction of light being in resonance with the transition between energy levels, it is assumed that the probabilities of the induced-emitted and absorbed radiation are equal, which requires the formation of population inversion of levels in order to implement quantum amplification of light. Population inversion is, generally speaking, only a particular case, in which the emission power prevails over absorption and, consequently, the light is amplified. Note that the possibility of light amplification without population inversion is of fundamental importance for quantum electronics and has a great practical value associated, in particular, with the problem of generation of short-wavelength vacuum ultraviolet and X-ray radiations. Since the advent of the first laser, the possibility of inversionless light amplification has been attracting increased interest of researchers and remains relevant. A fairly complete review of works published before 2000 and dedicated to inversionless amplification is given in [1]. Many papers have been published on this problem, in which various specific amplification schemes are proposed. Attention is drawn to a group of publications made independently by different authors and dedicated to the suppression of resonant absorption due to the interference of various absorbing channels. It is worth noting the works by A.K. Popov [2, 3], O.A. Kocharovskaya [4, 5], S.E. Harris [6], V.G. Arkhipkin [7, 8], and a number of other authors, for example, work [9]. Note that Zibrov et al. [10] have experi-

mentally confirmed the basic idea embodied in these works. In addition, we can mention a series of works by L.A. Rivlin [11–13], in which the concept of amplification of gamma radiation in cooled ensembles of free nuclei with latent inversion due to the Mössbauer effect was formulated and justified, and also the work by N.P. Konopleva and A.M. Tumaykin [14], who propose a scheme of inversionless amplification of the probe field in a medium of two-level degenerate atoms in the presence of a magnetic field. In Refs [15–17], the possibility of inversionless amplification in three-level systems of various types is demonstrated.

The possibility of inversionless light amplification in a system of molecules state-selectively oriented in a constant external field was discussed in the approximation of smallness of the orientation time as compared to the lifetime of excited state [18, 19]. It was shown that in a constant orienting field (OF), due to the low value of a pre-breakdown OF, it is unlikely that a sufficient degree of orientation of simple molecules satisfying the above approximation could be obtained. These ideas were subsequently developed in [20–23], in which, based on a coupled system of Boltzmann kinetic equations for the ground and excited states, a possibility for inversionless light amplification in a system of dipole molecules, state-selectively oriented in an external OF (both constant and pulsed), was considered at an arbitrary ratio of the orientation time and the excited state lifetime. It is shown that the degree of selective orientation under a pulsed OF may be sufficient in some cases for inversionless amplification.

In this paper, we consider the possibility of inversionless light amplification in a system of non-dipole molecules state-selectively oriented in an external orienting field of laser radiation. This makes it possible to significantly increase the orientation parameter owing to the greater intensity of the pre-breakdown field. In addition, the possibility of increasing the selectivity over states due to a quasi-resonant orienting effect is considered. The schemes of inversionless amplification at different mutual directions of the OF and probe fields and different values of the orientation parameters of ground and excited states are analysed numerically.

2. Models and research methods

When a molecular medium is exposed to the action of external orienting fields E_0 , molecules are oriented in the direction corresponding to the minimum potential energy of their interaction with the OF [24, 25]. In this case, the orientation direction is determined both by the direction of the external field E_0 and the molecule symmetry. The degree of orientation of a medium as a whole depends on the orientation parameter, which represents a ratio of the energy U of molecule interac-

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Received 29 May 2018; revision received 24 July 2018
Kvantovaya Elektronika 48 (11) 1076–1082 (2018)
Translated by M.A. Monastyrsky

tion with field to the energy kT of thermal motion exerting a disorienting effect. Naturally, the energy U may be different for molecules in the ground and excited states; therefore, the degree of orientation by the external OF for the excited and unexcited molecules may differ. In this case, the probabilities of induced transitions with absorption and amplification of a plane-polarised probe field \mathbf{E} may also be different. Thus, by controlling the orientation degree in the ground and excited states, it is possible to implement the conditions for light amplification with a certain polarisation in the absence of population inversion [18, 19]. In this paper, we consider the state-selective orientation of molecules in the field of quasi-resonant laser radiation.

The potential energy of interaction of a non-isotropically polarised molecule with the electric field \mathbf{E}_0 in the absence of a constant dipole moment is defined through the components of the tensor of electric polarisation of the molecule in the j th energy state (α_{ik}^j) [19, 24],

$$U_j = -\alpha_{ik}^j E_{ik}^* E_{0i} / 2. \quad (1)$$

We consider axially symmetric molecules and assume that the direction of the electric dipole moment of transition between the ground and excited states coincides with the direction of the symmetry axis, which constitutes the angle ϑ_0 with the electric field \mathbf{E}_0 [18–20]. In this case, the orientation distribution function f_j of a non-isotropically polarised axially-symmetric molecule in the presence of the electric field only depends on the angle ϑ_0 and, in the absence of a constant dipole moment, is given by the expression

$$f_j(\Theta, E_0) = f_j(\vartheta_0, E_0) = A_j \exp[\pm q_j \cos^2 \vartheta_0]. \quad (2)$$

Here, $A_j^{-1} = \int \exp[-U_j(\Omega, E_0)/kT] d\Omega$ is the normalising factor; T is the temperature; k is the Boltzmann constant $d\Omega = \sin \vartheta d\vartheta d\varphi$ is the solid angle element; $q_j = (\alpha_{33}^j - \alpha_{11}^j) \times |E_0|^2 / (2kT)$ are the dimensionless orientation parameters of the molecule polarisability ellipsoid; and α_{33}^j and α_{11}^j are the principal values of the polarisability tensor along and across the symmetry axis, respectively, for a molecule in the j th state. Assuming that $\alpha_{33}^j \gg \alpha_{11}^j$, the component α_{33}^j of the polarisability tensor for a molecule in the j th electronic state at the OF frequency ω_0 can be determined by the expression [26]

$$\alpha_{33}^j = \frac{e^2}{m} \sum_l \frac{F_{lj}}{\omega_{lj}^2 - \omega_0^2}. \quad (3)$$

Here, ω_{lj} and F_{lj} are the frequency and oscillator strength of the transition from state j to state l ; e is the charge of the electron; and m is the mass of the electron.

A distinctive feature of the orientation of non-dipole molecules as compared to dipole ones is the dependence of the sign of the orientation parameter q_j both on the molecule structure and the OF frequency ω_0 . In particular, for molecules elongated along the symmetry axis (with positive optical anisotropy), $\alpha_{33}^j > \alpha_{11}^j$ and the parameter q_j is positive at not too high OF frequencies ($\omega_0 < \omega_{lj}$). For disk-shaped molecules (with negative optical anisotropy), we have $\alpha_{33}^j < \alpha_{11}^j$ and $q_j < 0$ at $\omega_0 < \omega_{lj}$ [24, 25].

It is known that the breakdown intensity for pulsed OF radiation at optical frequencies is much higher than that for a constant one. As was shown in work [20, 21], the orientation parameter for dipole molecules in the schemes in question cannot exceed unity for a constant OF even at the pre-break-

down \mathbf{E}_0 values. Although the \mathbf{E}_0 value can be increased by an order of magnitude in pulsed fields [27], this is, as a rule, insufficient. At the same time, the orientation parameter of the molecule polarisability ellipsoid in the field of pulsed laser radiation, the magnitude of which is lower than the breakdown one can significantly exceed unity (see references in papers [18, 19]).

We assume that the OF frequency ω_0 lies far from the transition frequency at which the amplification of the probe field \mathbf{E} with the frequency ω occurs. Let us dwell on two cases of mutual orientation of \mathbf{E}_0 and \mathbf{E} , depending on the relation between q_1 and q_2 .

According to [18, 19], the amplification (absorption) factor $\alpha > 0$ ($\alpha < 0$) defined by the Bouguer law, for the case of molecules oriented in the external field \mathbf{E}_0 and plane-polarised probe radiation \mathbf{E} , has the form:

$$\alpha = N\sigma_0 \int_{\Omega} [n_2 f_2(\Omega, E_0) - n_1 f_1(\Omega, E_0)] \cos^2 \vartheta d\Omega. \quad (4)$$

Here, $\sigma_0 = [(8\pi)^2 / (c\hbar)] \omega |d_{21}|^2 F(\omega)$ is the absorption–emission cross section of the molecule, whose electro-dipole transition moment is oriented parallel to the polarisation plane; d_{21} is the matrix element of the electro-dipole transition moment in the coordinate system associated with the molecule; $F(\omega)$ is the frequency form-factor; ω is the probe field frequency; ϑ is the angle between the direction of the electro-dipole transition moment d_{21} and the electric field vector \mathbf{E}_0 ; N is the concentration of particles; n_1 and n_2 are relative populations of the ground and excited energy levels ($n_1 + n_2 = 1$) stipulated by the external pump source, the type of which is not specified here; $d\Omega = \sin \vartheta d\vartheta d\varphi$ is the solid angle element; and $f_1(\Omega, E_0)$ and $f_2(\Omega, E_0)$ are the normalised-to-unity statistical distribution functions of molecular orientation in the ground (1) and excited (2) states, determined by the energy $U(\Omega, E_0)$ of molecular interactions with the external electric OF \mathbf{E}_0 .

Let the excited state polarisability be higher than that of the ground state, $q_2 > q_1$, then the parallel orientation of the orienting and amplified (probe) fields, $\mathbf{E} \parallel \mathbf{E}_0$, is optimal. In this case, the coefficient α ($\alpha > 0$ refers to amplification, and $\alpha < 0$ refers to absorption) appears as

$$\alpha = N\sigma_0 \int_0^{2\pi} d\varphi \int_0^{\pi} [n_2 A_2 \exp(q_2 \cos^2 \vartheta) - n_1 A_1 \exp(q_1 \cos^2 \vartheta)] \cos^2 \vartheta \sin \vartheta d\vartheta. \quad (5)$$

In the opposite case, when $q_1 > q_2$, the orthogonal geometry of orienting and probe fields, $\mathbf{E} \perp \mathbf{E}_0$, is optimal. In this case, the amplification factor is expressed as:

$$\alpha = N\sigma_0 \int_0^{2\pi} d\varphi \int_0^{\pi} [n_2 A_2 \exp(q_2 \cos^2 \vartheta) - n_1 A_1 \exp(q_1 \cos^2 \vartheta)] \sin^3 \vartheta d\vartheta. \quad (6)$$

When the OF with an intensity \mathbf{E}_0 is applied to the medium, the molecule orientation is described by the orientation distribution function $f_j(\Omega, E_0)$, which in general case is determined by the lifetimes τ_1^{eff} and τ_2^{eff} of the ground and excited states, respectively, and also by the time τ_0 of orientational relaxation, which depends both on the molecule parameters and the parameters of a medium in which the molecules

are placed. The lifetimes of the ground and excited states are defined as:

$$\frac{1}{\tau_1^{\text{eff}}} = w_{12}^{\text{ind}}, \quad \frac{1}{\tau_2^{\text{eff}}} = w_{21}^{\text{ind}} + w_{21}^{\text{sp}}, \quad \frac{\tau_2^{\text{eff}}}{\tau_1^{\text{eff}}} = \frac{n_2}{n_1}. \quad (7)$$

Here, $w_{12}^{\text{ind}} = w_{21}^{\text{ind}}$ and w_{21}^{sp} are the probabilities of induced and spontaneous transitions, respectively.

By using (5) and (6), Popov and Slabko [18, 19] obtained an analytical expression for the amplification factor α in terms of the generalised second-order Langevin functions for the stationary case in the approximation $\tau_2^{\text{eff}} \gg \tau_0$. Naturally, in the approximation accepted above, the functions $f_j(\Omega, E_0)$ have the Boltzmann form, because, during the lifetime of the excited state, molecules spend most of the time in thermalised state. In our work, the nonstationary regime for an arbitrary ratio between τ_0 and τ_2^{eff} is considered. It is obvious that this consideration requires a more sophisticated model based on a system of kinetic equations to be used.

To determine the distribution functions $f_1(\Omega, E_0)$ and $f_2(\Omega, E_0)$ and then the amplification factor α with taking into account expressions (5) and (6), we use a system of nonstationary kinetic equations [22, 23]

$$\begin{aligned} 6\tau_0 \frac{\partial f_1}{\partial t} - \frac{1}{\sin \vartheta} \frac{\partial}{\partial \vartheta} \left\{ \sin \vartheta \left[\frac{\partial f_1}{\partial \vartheta} + f_1 \frac{\partial}{\partial \vartheta} \left(\frac{U_1(t)}{kT} \right) \right] \right\} \\ = -\frac{6\tau_0}{\tau_1^{\text{eff}}} f_1 + \frac{6\tau_0}{\tau_2^{\text{eff}}} f_2, \\ 6\tau_0 \frac{\partial f_2}{\partial t} - \frac{1}{\sin \vartheta} \frac{\partial}{\partial \vartheta} \left\{ \sin \vartheta \left[\frac{\partial f_2}{\partial \vartheta} + f_2 \frac{\partial}{\partial \vartheta} \left(\frac{U_2(t)}{kT} \right) \right] \right\} \\ = -\frac{6\tau_0}{\tau_2^{\text{eff}}} f_2 + \frac{6\tau_0}{\tau_1^{\text{eff}}} f_1. \end{aligned} \quad (8)$$

Let us supplement system (8) with boundary conditions (9), initial conditions (10), and the normalisation condition for the distribution functions at the initial time moment (11):

$$\left. \frac{\partial f_1}{\partial \vartheta} \right|_{\vartheta=0} = \left. \frac{\partial f_2}{\partial \vartheta} \right|_{\vartheta=0} = 0, \quad \left. \frac{\partial f_1}{\partial \vartheta} \right|_{\vartheta=\pi} = \left. \frac{\partial f_2}{\partial \vartheta} \right|_{\vartheta=\pi} = 0, \quad (9)$$

$$f_1(0, E_0) = g_1(\vartheta), \quad f_2(0, E_0) = g_2(\vartheta), \quad (10)$$

$$2\pi \int_0^\pi [g_1(\vartheta) + g_2(\vartheta)] \sin \vartheta d\vartheta = 1. \quad (11)$$

For the numerical analysis of system (8)–(11), we use an original efficient computational algorithm, which is based on the exponential fitting method [28, 29] and implemented with the use of a first-order explicit computational scheme:

$$\begin{aligned} \frac{f_{1i}^{j+1} \exp[(\eta_1 + \eta_2)\tau] - f_{1i}^j}{\exp[(\eta_1 + \eta_2)\tau] - 1} (\eta_1 + \eta_2) \sin \vartheta_i - (\eta_1 + \eta_2) f_{1i}^j \sin \vartheta_i \\ + \frac{kT}{W} L_{1h} f_{1h}^j = -\eta_1 f_{1i}^j \sin \vartheta_i + \eta_2 f_{2i}^j \sin \vartheta_i, \\ \frac{f_{2i}^{j+1} \exp[(\eta_1 + \eta_2)\tau] - f_{2i}^j}{\exp[(\eta_1 + \eta_2)\tau] - 1} (\eta_1 + \eta_2) \sin \vartheta_i - (\eta_1 + \eta_2) f_{2i}^j \sin \vartheta_i \end{aligned} \quad (12)$$

$$+ \frac{kT}{W} L_{2h} f_{2h}^j = -\eta_2 f_{2i}^j \sin \vartheta_i + \eta_1 f_{1i}^j \sin \vartheta_i.$$

Here, $\eta_1 = 6\tau_0/\tau_1^{\text{eff}}$; $\eta_2 = 6\tau_0/\tau_2^{\text{eff}}$; and L_{1h} and L_{2h} are the exponential difference operators [22].

In constructing the exponential difference scheme (12), the exponential fitting method was applied to approximate the differential expressions both in spatial variables and in time. Conditional stability and monotonicity of the constructed scheme (12) have been proved. Due to high speed of the physical processes of laser generation, the stability condition turns out not cumbersome.

To solve the original system (8)–(11), we extend the normalisation condition (11) to any time moment t :

$$2\pi \int_0^\pi [f_1(t, \vartheta) + f_2(t, \vartheta)] \sin \vartheta d\vartheta = 1. \quad (13)$$

One of the most important aspects of numerical simulation is conservatism of the difference schemes applied. The exponential difference scheme (12) has the property of conservatism, namely, for the numerical solution in accordance with scheme (12), the difference analogue of the normalisation condition (13) is valid for any computational time layer $t = t^j$:

$$\sum_{i=0}^n f_{1i}^j \sin \vartheta_i + \sum_{i=0}^n f_{2i}^j \sin \vartheta_i = \frac{1}{2\pi}.$$

A detailed presentation and theoretical justification of this computational algorithm for the numerical study of the systems of kinetic equations are given in work [22].

3. Stationary case

The main goal of the study in a stationary case is to evaluate the impact of the ratio of the orientational equilibrium time and the excited state lifetime, $\tau_0/\tau_2^{\text{eff}}$. The interaction energy U_j in (8) does not depend on time, and so we have to put the time derivative equal to zero and supplement system (8) with boundary conditions

$$\left. \frac{\partial f_1}{\partial \vartheta} \right|_{\vartheta=0} = \left. \frac{\partial f_2}{\partial \vartheta} \right|_{\vartheta=0} = 0, \quad \left. \frac{\partial f_1}{\partial \vartheta} \right|_{\vartheta=\pi} = \left. \frac{\partial f_2}{\partial \vartheta} \right|_{\vartheta=\pi} = 0 \quad (14)$$

and a normalisation condition for the distribution functions at the initial time moment [20, 21]

$$2\pi \int_0^\pi [f_1(\vartheta) + f_2(\vartheta)] \sin \vartheta d\vartheta = 1. \quad (15)$$

When solving the system of equations (8), (14), and (15) for molecules stretched along the symmetry axis, the dependence of the dimensionless amplification α' ($\alpha' = \alpha/N\sigma_0$) on the orientation parameter $|q_1|$ at $q_2 > q_1 > 0$ with allowance for relation (5) is described by the curves shown in Fig. 1a. For disk-shaped molecules, $q_1 < q_2 < 0$, the dependences of the coefficient α' on the parameter $|q_2|$ are presented in Fig. 1b. Here, as mentioned above, $\mathbf{E} \parallel \mathbf{E}_0$ is the optimal mutual orientation of the fields.

As can be seen from Fig. 1a, for molecules stretched along the symmetry axis, amplification replaces absorption in a narrow region in the approximation $\tau_2^{\text{eff}} \gg \tau_0$ [curve (1)] described in works [18, 19]. It is also shown that even at $\tau_2^{\text{eff}} > \tau_0$ [curve (2)], the amplification effect can be attained, which

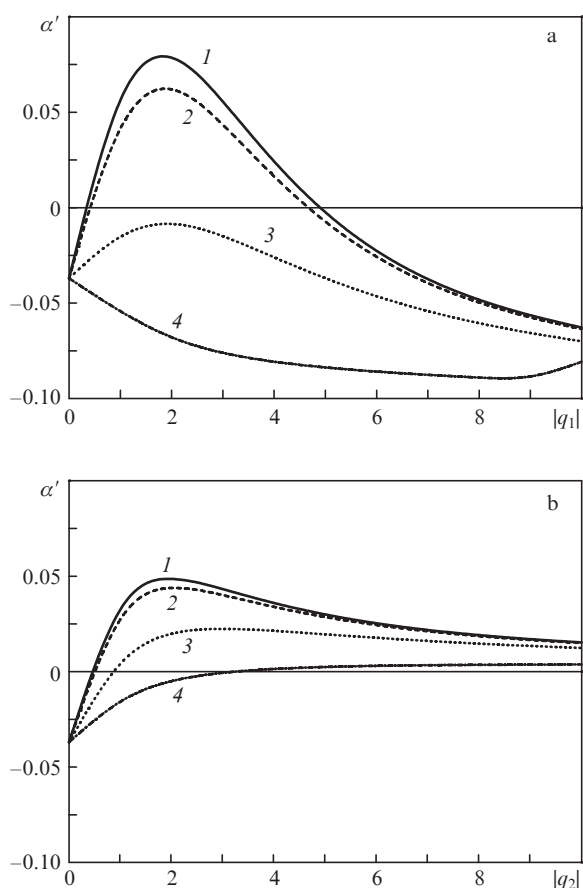


Figure 1. Dependences of the dimensionless coefficient α' on the orientation parameter $|q_j|$, obtained at $\mathbf{E} \parallel \mathbf{E}_0$ for (a) $q_2 > q_1 > 0$, $|q_2|/|q_1| = 4$ and (b) $q_1 < q_2 < 0$, $|q_1|/|q_2| = 4$. Calculations were performed at $n_2/n_1 = 0.8$ for $\tau_0/\tau_2^{\text{eff}} = (1) 0.01$, (2) 0.1, (3) 1.0, and (4) 10.0.

does not impose such rigid restrictions as in works [18, 19]. In the case $\tau_2^{\text{eff}} \leq \tau_0$ [curves (3) and (4)], amplification does not occur in the entire domain where q_j is defined. Thus, for given parameters, the amplification region is limited to a rather narrow interval of q_j values. This is due to the fact that in the selected range of q_j values, the orientation distribution functions of molecules differ significantly. At the same time, a further increase in the OF ($U/kT \gg 1$) results in the levelling of these differences, so that absorption begins to prevail over amplification.

However, as can be seen from Fig. 1b, for disk-shaped molecules, amplification replaces absorption in a wide range. Indeed, the molecule symmetry axis is orthogonal to the OF. Qualitatively, the situation can be explained as follows: at large q_1 values, all molecules in the ground state turn out completely oriented so that the projection of the electro-dipole transition moment being parallel to the symmetry axis onto the direction of the probe field \mathbf{E} equals to zero. Consequently, molecules in the ground state do not absorb the probe field. At the same time, the orientation degree of molecules in the excited state is less than that in the ground state since $|q_1| > |q_2|$, while the orientation distribution function of molecules in the ground state is steeper than that in the excited state, and the average value of the projection of the transition electro-dipole moment onto the \mathbf{E} direction is not zero, which leads to its amplification. This also leads to the fact that the distribution function steepness increases even

more with increasing q_j , and amplification can be attained even at large orientation parameters.

A similar situation is observed at $q_1 > q_2 > 0$ and $q_2 < q_1 < 0$ for orthogonal ($\mathbf{E} \perp \mathbf{E}_0$) geometry of the orienting and amplified fields.

Of greatest interest may be the variant in which the sign of the parameter q_j is different in the ground and excited states. This is possible if the signs of the exit from the resonance of the OF frequency ω_0 relative to the electron transition ω_{ij} of the molecule in states 1 and 2 are different [see expression (3)]. Indeed, if in the excitation of molecule to state 2, the transition frequency between the working energy level 2 and any other excited level j satisfies the condition $\omega_{ij} < \omega_0$, while the transition frequencies satisfy the condition $\omega_{ij}, \omega_{12} > \omega_0$, then $q_2 < 0 < q_1$ for elongated molecules and $q_2 > 0 > q_1$ for disk-shaped ones. In this case, the OF causes an orienting effect on molecules in one of the working states (2 or 1), and a disorienting effect – in the other.

Figure 2a shows the dependences of the dimensionless coefficient α' on q_1 for $q_1 > 0$, $q_2 < 0$, $|q_1| = |q_2|$ at $\mathbf{E} \perp \mathbf{E}_0$ [see expression (6)], similar dependences on q_2 for $q_1 < 0$, $q_2 > 0$, $|q_1| = |q_2|$ at $\mathbf{E} \parallel \mathbf{E}_0$ [see expression (5)] are presented in Fig. 2b.

As can be seen from Fig. 2, amplification can be attained in a wide range of q_j values. Note that the dependences shown in this Figure are an evidence of the effect of the OF frequency ω_0 on amplification and indicate a significant advantage of this scheme in implementing inversionless amplification.

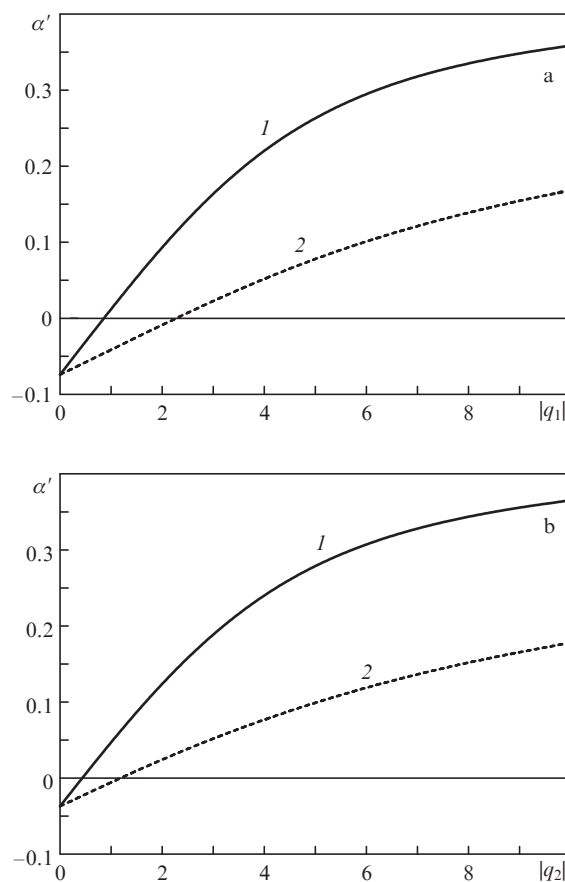


Figure 2. Dependences of the dimensionless coefficient α' on the orientation parameter $|q_j|$, obtained (a) at $\mathbf{E} \perp \mathbf{E}_0$ for $0 > q_2 = -q_1$ and (b) at $\mathbf{E} \parallel \mathbf{E}_0$ for $0 > q_1 = -q_2$. Calculations were performed at $n_2/n_1 = 0.8$ for $\tau_0/\tau_2^{\text{eff}} = (1) 0.01$ and (2) 1.0.

4. Nonstationary case

As shown below, at characteristic values of the electro-dipole moments of molecule transitions, q_j values may significantly exceed unity at the OF intensity E_0 , which can hardly be attained in the continuous-wave regime. Therefore, it is advisable to consider the case of pulsed orientation of molecules in the orienting field of laser radiation with a duration on an order of magnitude greater than τ_2^{eff} .

The following computational experiment was performed to study the nonstationary system (8) exposed to the action of the pulsed OF E_0 [see expression (2)] of rectangular shape. As noted above, provided the excited state polarisability is greater than that of the ground state ($q_2 > q_1$), parallel orientation of the orienting and amplified fields is optimal, $E \parallel E_0$. Figure 3 shows the time dependences of the dimensionless amplification factor α' [see expression (5)] under the action of a pulsed OF for molecules, both stretched along the symmetry axis [$q_2 > q_1 > 0$, curve (1)] and disk-shaped ones [$q_1 < q_2 < 0$, curve (2)].

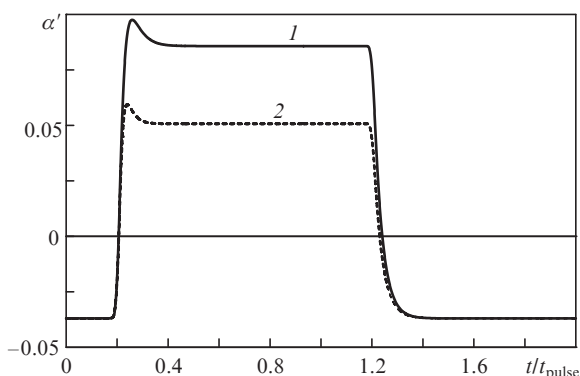


Figure 3. Effect of the orientation parameter on amplification at $E \parallel E_0$. Calculations were performed at $n_2/n_1 = 0.8$, $\tau_0/\tau_2^{\text{eff}} = 0.01$ for (1) $q_2 > q_1 > 0$ ($q_1 = 2.0$, $q_2 = 8.0$) and (2) $q_1 < q_2 < 0$ ($q_1 = -8.0$, $q_2 = -2.0$).

As is seen from Fig. 3, the inversionless amplification is attained during a time interval on the order of orientation time. A sharp increase in amplification at the initial stage of switching on of the OF is due to the fact that $q_2 > q_1$, and therefore, for molecules in the excited state, the orientation distribution functions reach a quasi-stationary state faster

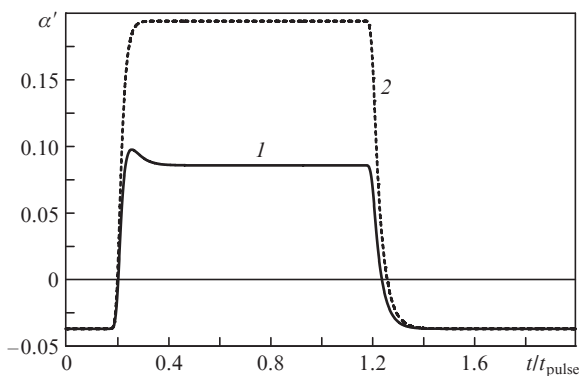


Figure 4. Effect of the orientation parameter on amplification at $E \parallel E_0$. Calculations were performed at $n_2/n_1 = 0.8$, $\tau_0/\tau_2^{\text{eff}} = 0.01$ for (1) $q_2 > q_1 > 0$ ($q_1 = 2.0$, $q_2 = 8.0$) and (2) $q_2 > q_1 = 0$ ($q_1 = 0$, $q_2 = 8.0$).

than for molecules in the ground state. Naturally, after the ground state molecules acquire a quasi-stationary orientation distribution function, the amplification coefficient is stabilised. This can be easily verified by setting the parameter q_1 equal to zero (Fig. 4).

Figure 5 demonstrates the time dependence of the dimensionless amplification factor α' [see expressions (5), (6)] under the action of a pulsed OF E_0 in the case when the signs of the parameter q_j in the ground and excited states are different.

As can be seen from Fig. 5, there is no sharp jump in amplification at the time moment of switching on of the OF. This is due to the fact that in this example $|q_2| = |q_1|$, and the equilibrium orientation distribution functions are attained at equal time intervals.

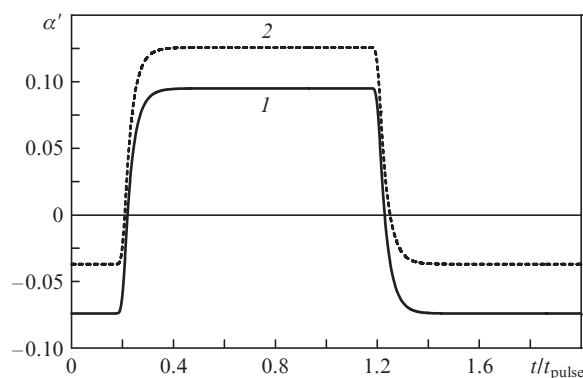


Figure 5. Effect of the orientation parameter on amplification. Calculations were performed at $n_2/n_1 = 0.8$, $\tau_0/\tau_2^{\text{eff}} = 0.01$ for (1) $q_1 = 2.0$, $q_2 = -2.0$, $E \perp E_0$ and (2) $q_1 = -2.0$, $q_2 = 2.0$, $E \parallel E_0$.

5. Discussion of results and estimates

The above consideration allows us to evaluate the conditions for the implementation of inversionless amplification by the example of electronic transition in J_2 iodine molecule, which is fairly well studied and has electronic transitions in the visible range [30, 31]. Figure 6 shows a diagram of three lower electronic levels from work [32].

Expression (3) makes it possible to estimate the values of the polarisability tensor components of a molecule in states 1 and 2, which are proposed for estimating the possibility of inversionless amplification. In this case, state 1 is the ground state, and state 2 is the excited working state.

Let us consider the most convenient variant for the experiment in which the OF has the same frequency ω_0 equal to the second-harmonic frequency of radiation from a neodymium laser with a wavelength $\lambda = 0.53 \mu\text{m}$. In this case, as can be seen from Fig. 6, the OF frequency is higher than the frequency of the resonant transition between levels 1–2 and less than the frequency of the transition between levels 2–3. This provides different signs of the orientation parameter in the ground and excited states, which makes amplification more advantageous compared to the case when the orientation parameters q_j have the same signs (see Fig. 2). The characteristic value of the components of the molecule polarisability tensor at a large output from resonance (the nonresonant case) constitutes $\sim 10^{-24} \text{ cm}^3$ [26].

By using expression (3), we can estimate the quasi-resonance value of the polarisability tensor of the J_2 molecule in

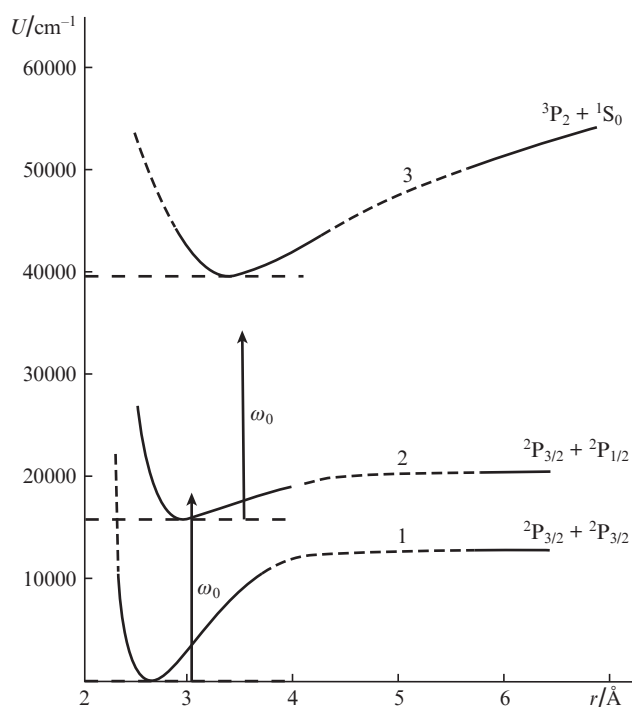


Figure 6. Diagram of potential energy levels for the J_2 molecule [32]; U is the potential energy, and r is the interatomic distance.

states 1 and 2. Assuming the F_{lj} values for transitions 1–2 and 2–3 to be the same and equal to 0.5, we obtain the quasi-resonant values $\alpha_{33}^{(1)} = -3.50 \times 10^{-23} \text{ cm}^3$, $\alpha_{33}^{(2)} = 1.82 \times 10^{-23} \text{ cm}^3$ which are an order of magnitude higher than the nonresonant ones. Then, according to (2), for the orientation parameter at $T = 300 \text{ K}$ we have $q_1 = -I_1 \cdot 3.54 \times 10^{-19}$, $q_2 = I_2 \cdot 1.84 \times 10^{-19}$; therefore, for $|q_1|, |q_2| > 1$, the minimal OF intensities are as follows: $I_1 = 2.82 \times 10^{11} \text{ W cm}^{-2}$, $I_2 = 5.43 \times 10^{11} \text{ W cm}^{-2}$. It is obvious that the OF intensity should be less than the breakdown threshold. For estimates, we use works [33–35], in which the data on the breakdown threshold for a number of molecular and atomic gases are presented. Thus, at a laser pulse duration $\tau_p = 50 \text{ ps}$ and a near-atmospheric pressure, the intensity of the breakdown threshold in these gases constitutes $\sim 10^{14} \text{ W cm}^{-2}$ and decreases as $1/\tau_p^{1/2}$. Then, when using J_2 vapours with a concentration significantly lower than that of the buffer gas, it can be assumed that the breakdown intensity for a mixture at $\tau_p = 5 \text{ ns}$ is close to $10^{13} \text{ W cm}^{-2}$. This is more than an order of magnitude higher than the OF intensity required for the experiment utilising both pico- and nano-second OF pulses. Note that the OF intensity can be reduced by 1–2 orders of magnitude when using frequency-tunable orienting radiation. Naturally, this greatly complicates the requirements to the experimental setup.

The version of the experiment under discussion is convenient because it allows the use of the Stokes component of the stimulated Raman scattering (SRS) of the OF as a pump source for the 1–2 transition. To ensure that the pump frequency corresponds to the maximum of electron–oscillation transition band, it is necessary to provide the Stokes shift by approximately $1000\text{--}2000 \text{ cm}^{-1}$ [36]. Given the relatively high OF intensity, the SRS of the intensity required for saturation of the 1–2 transition can be obtained in a number of media, which will practically ensure close equality of populations, $n_1 \approx n_2$. Thus, the proposed version may become a basis for the planned experiment.

6. Conclusions

The calculations, considerations, and estimates presented in this work allow us to make a conclusion on the possibility of inversionless amplification of radiation by state-selective orientation of molecules in the laser radiation field. The calculations of specific variants confirm the qualitative considerations that can be used to predict situations at different ratios between the q_j parameters, and also between the lifetimes of the excited states and orientation times. We should emphasise that the rigid restriction on the choice of molecules made in [18, 19], $\tau_2^{\text{eff}} \gg \tau_0$, is removed, and this allows us to expand the class of molecules to be used, including the use of protein macromolecules.

In the case of optical orientation, the polarisability anisotropy ($\alpha_{33}^j - \alpha_{11}^j$) is $\sim 10^{-24} \text{ cm}^3$ even at the resonance exits, $\omega_{lj} - \omega_0 \approx \omega_0$. At normal temperatures ($T \approx 300 \text{ K}$), the optical orientation parameter q_j can reach ~ 1 at the OF intensity below the breakdown threshold. It is of special importance to point out that the q_j value can be significantly increased as the ω_0 frequency approaches the optical transition frequencies, and, in addition, the selection of the ω_0 frequency may ensure the implementation of the conditions under which the orientation parameters in the ground and excited states have different signs. The latter makes it possible to obtain inversionless amplification at lower OF values and expands the class of molecular compounds that can be considered as objects for experimental studies.

It is obvious that the above estimates for the vapours of J_2 molecules can be used in experiment planning; however, the possibility of the practical use of a laser of this type, operating in the visible range, is questionable. Of practical interest may be a laser operating in the vacuum UV range, for example, a hydrogen laser [37] or its analogues. Indeed, by means of the quasi-resonant orientation of the upper excited state, lasing can be implemented using those transitions on which it has not yet been observed, or the lasing conditions at already known transitions can be improved.

Acknowledgements. V.V. Slabko is grateful for the support of the Ministry of Education and Science of the Russian Federation (Grant No. 3.6341.2017/VU).

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