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Classical theory of the molecule alignment in a laser field

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Abstract. In the classical mechanics framework the solution is given for the problem of the two-atomic molecule alignment in a field of linearly polarised laser pulse. The numerical results are presented for the Cl_2 molecule, together with the analytical results in the limit of a very short and strong laser pulse. The width of the angular distribution for molecular axes in field is demonstrated to depend on the radiation intensity as $S^{-1/4}$, the alignment along the polarisation direction is possible as well as in the transversal direction. The analysis of some experimental data is given.

PACS. 33.90.+h Other topics in molecular properties and interactions with photons

1 Introduction

The effect of ionisational dissociation of molecules in a strong laser field was discovered in a number of experimental studies some years ago (see *e.g.* [1–3]). The analysis of the anisotropy of angular distribution for the positive ions has demonstrated that the alignment of molecules along the laser polarisation direction can be one of possible causes for such anisotropy [4,5]. Another possible cause is the enhanced ionisation of aligned molecules [6–8]. A direct evidence for strong-field alignment was demonstrated in [9] where the stimulated Raman effect was observed in naphthalen trimers.

It should be noted that the time-independent quantum mechanical theory for the molecule orientation has long been known. In particular, the transition from the rotational spectrum of a free molecule to the pendular spectrum of a molecule in a laser field was discussed in [10]. These transitions are due to the librations of the molecular axis (determining the direction of the induced dipole moment) about the direction of the electric vector of the light wave. In this case the solution for the Schrödinger equation may be written in terms of the spheroidal functions, stretched or ablated, depending on the sign of the dynamic polarisability of the molecule. The same result has been recently derived in [11]. However, in most of the experiments the short laser pulses were used with the duration comparable or even less than the period of the free molecular rotation. So the time-independent theory does not hold for this case.

The numerical analysis of the time-dependent Schrödinger equation for the simplest molecule H_2^+ in the linearly polarised laser pulse has been carried out and some interesting results have been obtained in [12,13]. In the reference [12], an asymmetry was predicted for the upward and downward emission of ions depending on the relative phases of waves in a two-color laser field. A possibility for stabilisation of ions H_2^+ has been discovered in [13]. Unfortunately, the use of such approach to the description of more compound molecules involves essential difficulties in calculations.

In this article the classical mechanics is used to describe the molecule alignment in field. The stimulated Raman scattering of light with the transitions between the molecule rotation levels according to the selection rules, $|\Delta J| \leq 2$, is the microscopic reason for the collisionless alignment. Such transitions result in a preferable direction of the molecular axis (with the induced dipole moment along) which coincides with the polarisation direction of the linearly polarised field if the polarisability of the molecule is positive or which is perpendicular to this direction if the polarisability is negative. Evidently, it is sufficient for the validity of the classical mechanics, that the laser radiation spectrum width were larger than the distance between the rotational levels. Then the stimulated emission and absorption of photons (which for the Raman processes have different energies) can be provided by the photons of the incident laser radiation. This condition fulfills in experiments we have discussed above.

The classical approach used for this problem in recent papers [14–16], has enabled one to explain qualitatively the experimental results of references [14–16], which were obtained for CS_2, CO_2 molecules under the laser pulse duration ~ 35 ps. But when simultaneously treating the alignment and dissociation, even the classical problem becomes exceedingly complicated. For instance, the angular distribution dependence upon the important parameters of the laser radiation (*e.g.* intensity and the pulse duration) was therefore not obtained for the "pure" alignment in [14–16]. Bearing in mind the restrictions of the model, where the molecule alignment is considered separately

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of the accompanying ionisation, one can hope, however, that the results of this work will be helpful in the interpretation of the experimental data. Concerning the description of the process with the help of the classical mechanics, its advantages for understanding the behaviour of atoms and molecules in a laser field have been demonstrated by numerous authors (see, *e.g.* [17–19]).

The general equations are derived in the next section which describe the motion of a rotator in a time-dependent electromagnetic field. The equations of the motion are averaged over the rapid oscillations of the laser field on its carrier frequency. Therefore the temporal dependence of the field in these equations is due to only the temporal dependence of the laser pulse envelope. The validity of such averaging for polar molecules in the absence of the resonances[1] is determined by the condition (2). For the non-polar molecules such a condition is not derived by now. Therefore, one cannot estimate the error arising due to the averaging over the rapid field oscillations. Such estimation could be obtained only in a more accurate theory which should not use the averaging. The interaction of the rotator with the field is caused by the dynamic polarisability of the molecule. The hyperpolarisabilities are not taken into account. The validity of this approximation can be estimated roughly: the laser field strength should be much less than the atomic one. However, we hope that the qualitative features of the alignment process can be described adequately by the proposed theory even in the case of high laser field strengths which are comparable with the atomic field. The below comparison of the theory with the experiment verifies this statement.

The results of numerical solution of equations derived in such a way are presented in Section 3 for the molecule of Cl_2 as an example. The analytical study of the equations of motion in the limit of a very strong field is carried out in Section 4. In our study we consider the laser field as strong if it exceeds some crucial value determined by the inertia momentum of the molecule. Since this momentum is proportional to the masses of nuclei and the atomic field is determined by the electron mass, therefore this crucial field is much less than the atomic one. Section 5 deals with analytical considerations of the problem in the case when the alignment of molecules appears in the plane perpendicular to the polarisation vector of the field. A special form of the time dependence is used here for the laser pulse envelope which enables us to derive a closed expression for the angular distribution of molecules. The discussion of the experimental data is given in the last section.

2 Lagrange equation

Let the z-axis of the laboratory reference frame be directed along the electric field vector of the linearly polarised laser wave, $F(t)$ is the envelope of the electric vector in the laser pulse, ω_l is the laser field frequency. Let ϕ and θ to be the angles determining the orientation of the 2-atomic molecule axis. Taking into account only the terms quadratic to the field strength the potential energy of the molecule, averaged over the field period $2\pi/\omega_l$, takes the form (*cf.* [22]):

$$\mathsf{U}(\theta, \phi; t) = -\frac{1}{4}F^2(t)[\alpha_\parallel(\omega_l)\cos^2\theta + \alpha_\perp(\omega_l)\sin^2\theta].$$

Here $\alpha_{\parallel,\perp}$ are the longitudinal and transversal dynamical polarisabilities of the molecule in respect to its axis. In this approximation the frequency of the laser radiation ω_l arises only in the components of the dynamic polarisability $\alpha_{\parallel,\perp}$.

The Lagrange function determining the rotation of the molecule in the field is (*cf.* [23,24]):

$$\mathcal{L}(\theta, \phi; t) = \frac{I}{2}(\dot\phi^2\sin^2\theta + \dot\theta^2) + \frac{F^2(t)}{4}[\alpha_\parallel(\omega_l)\cos^2\theta + \alpha_\perp(\omega_l)\sin^2\theta], \qquad (1)$$

where I is the inertia momentum.

Deriving the Lagrange function (1) it was assumed implicitly that the molecule is a homonuclear one. This Lagrange function can be used for heteronuclear molecules with a constant dipole momentum $\mathbf{d}$, if the following condition is satisfied

$$\mathbf{dF} \ll \hbar\omega_l. \qquad (2)$$

This condition provides the elimination of the terms linear to the field because of the rapid oscillations of the field with the frequency ω_l [25].

Since the ϕ coordinate is cyclic, the momentum projection onto the z-axis is conserved:

$$M_z = I\dot\phi\sin^2\theta. \qquad (3)$$

The molecular dynamics is determined by the Lagrange equation, which with an account for the equation (3) reads as

$$\frac{\mathrm{d}^2\theta}{\mathrm{d}t^2} = \frac{M_z^2}{I^2}\frac{\cos\theta}{\sin^3\theta} - \frac{\beta F^2(t)}{2I}\sin\theta\cos\theta, \quad \beta \equiv \alpha_\parallel - \alpha_\perp. \qquad (4)$$

The numerical analysis of the equation (4) without the first term in its right-hand side was given in [26].

For qualitative analysis of the problem we consider firstly the time-independent limit supposing that the field amplitude F does not depend on time. In this case the energy integral corresponds to the equation (4)

$$E = \frac{I}{2}\dot\theta^2 + U(\theta), \quad U(\theta) = \frac{M_z^2}{2I\sin^2\theta} + \frac{1}{4}\beta F^2\sin^2\theta. \qquad (5)$$

It is easy to see that the potential energy $U(\theta)$ has its infinite maxima at $\theta = 0, \pi$, if $M_z \neq 0$. The minimum

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[1] Alignment of molecules in resonant laser field was studied in [20,21].

of the free molecule potential energy corresponds to $\theta = \pi/2$. A free molecule axis oscillates harmonically about this minimum:

$$\cos\theta = \left(1 - \frac{M_z^2}{2IE_0}\right)^{1/2} \cos\omega_0 t = \sin\gamma\cos\omega_0 t. \qquad (6)$$

Here γ is the angle between the total angular momentum direction of the molecule $\mathbf{M}$ and the z-axis: $M_z = M\cos\gamma$, $E_0 = M^2/2I$, $\omega_0 = \sqrt{2E_0/I}$ is the frequency of free rotations. Then

$$\dot\theta = \omega_0 \frac{\sin\gamma}{\sin\theta} \sin\left[\arccos\left(\frac{\cos\theta}{\sin\gamma}\right)\right]. \qquad (7)$$

With the field strength increase for $\beta < 0$, the minimum at $\theta = \pi/2$ deepens. It means that for $\beta < 0$ the molecules will be aligned perpendicularly to the electric field. This fact is physically clear since for $\beta < 0$ the energy of a molecule oriented along field increases.

For $\beta > 0$ the bifurcation appears in the phase plane of the system when the field strength increases: the equilibrium state for $\theta = \pi/2$ splits into two new equilibrium states and a saddle point with a two-loop separatrix passing through. This behaviour of the system is caused by the local potential energy maximum which appears at $\theta = \theta_{\max} \simeq \pi/2$. The location of the minimum near $\theta \simeq 0$ is determined by the relation

$$\theta_{\min} = \left(\frac{M_z^2}{I\beta F^2}\right)^{1/4}, \qquad (8)$$

if the field is strong enough, so that

$$\theta_{\min} \ll 1. \qquad (8')$$

A similar minimum appears also near $\theta \simeq \pi$. So, if the energy of a molecule is

$$E < \frac{M_z^2}{2I} + \frac{1}{4}\beta F^2,$$

then for $\beta > 0$ the molecules align along the field polarisation vector. The width of the angular distribution smoothly depends on the field intensity, $S = F^2$, and on the difference of polarisabilities, β, according to the equation (8), as $(\beta S)^{-1/4}$. It should be however noted that due to the maxima of the potential energy at $\theta = 0, \pi$, the angular distribution function for the molecular axes has minima for $\theta = 0, \pi$, if $M_z \neq 0$. These minima are due to the conservation of the angular momentum components along the linearly polarised field strength direction. They are discussed below in detail.

The critical field strength at which the bifurcation occurs for $\beta > 0$, is determined by the equation:

$$F = F_c \equiv \left[\left(4E - 2M_z^2/I\right)/\beta\right]^{1/2}. \qquad (9)$$

When the relation (9) holds, the minimum of the free rotation potential energy disappears and the maximum is beginning to arise due to the field action. So the stationary point (9) corresponds to the "indifferent" equilibrium.

3 Dynamics of alignment

There are no difficulties for the numerical solution of equation (4). To obtain the angle distribution function $P(\theta)$ the histograms of solutions $\theta(t)$ with the step $\sim 3°$ was plotted with random initial values θ_0, M_z and the initial energy of a molecule E_0. Since the quantity $\cos\theta_0$ has a simple (harmonic) temporal dependence (6), the distribution $P(\theta_0)$ may be derived from the distribution for the harmonic oscillator coordinate:

$$P(\theta_0) = \frac{1}{\pi}\begin{cases} \sin\theta_0(\sin^2\gamma - \cos^2\theta_0)^{-1/2}, & \cos^2\theta_0 < \sin^2\gamma, \\ 0, & \cos^2\theta_0 \geq \sin^2\gamma. \end{cases} \qquad (10)$$

The γ angle is supposed to have a uniform distribution:

$$P(\gamma) = \frac{1}{2}\sin\gamma,$$

and the value $\dot\theta_0$ is determined according to the equation (7). Because of the singularity of the function (10) at $\sin^2\gamma = \cos^2\theta_0$, the averaging over γ, θ_0 is carried out with the use of the Gauss-Chebyshev algorithm [27].

Some results of calculations are presented in Figure 1 for the distribution $P(\theta)$ at different time moments after the laser pulse have begun to act. The total number of initial conditions for the equation (4) which were used for plotting the curves of Figure 1 was about 50 000. The parameters of the molecule $\mathrm{Cl_2}$ studied in [28] were taken for the numerical values of molecular parameters. The inertia momentum of this molecule is $I = 10^{-38}$ g cm^2. The static polarisability difference has been taken for the difference of the longitudinal and transversal polarisabilities [29]: $\beta = 2.6 \times 10^{-24}$ cm^3. The Gauss form was used for the laser pulse:

$$F(t) \sim \exp(-t^2/2\tau^2),\ \tau = 100 \text{ fs},$$

with the power density of the flux of 10^{15} W/cm^2. These data correspond to the experimental setup described in [28]. The initial energy of the molecule E_0 has been assumed to have a Boltzmann distribution with temperature 300 K. For comparison with experiment the angle θ was considered in the range of $-\pi/2 \leq \theta \leq \pi/2$. For $t = 0$ the width of the distribution function at the half-height (without account for the central drop of the function $P(\theta)$) is 36.5°, that agrees satisfactorily with the experimental value of 38° [28].

Of course, the quantitative agreement between the theory and the experiment is not of great significance here, because of the approximations used in deriving the equations of motion. However, it can be stated that the proposed theory provides at least a correct qualitative interpretation for the experimental results.

4 Analytical considerations in a very strong field

To understand qualitatively the numerical results presented above, it is useful to consider the solution of equation (4) in a very strong field, the numerical estimation

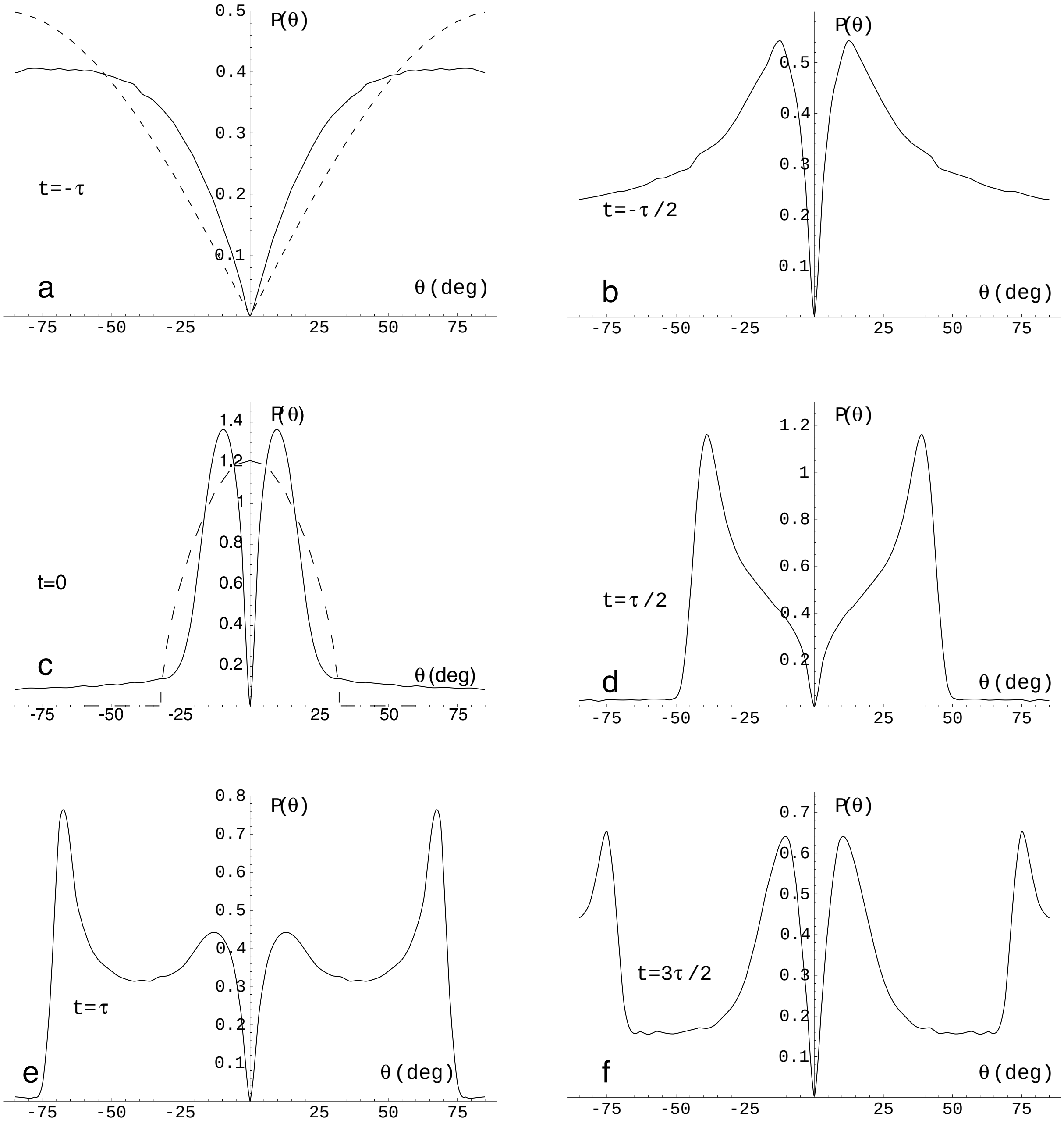

Fig. 1. Time dependence for the angular distribution of molecular axes. The molecular parameters are nearly those for the molecule Cl_2, $\tau = 100$ fs, the peak intensity of radiation is 10^{15} W/cm^2. The dashed line for $t = -\tau$ corresponds to the field-free distribution of molecular axes $P(\theta_0) = (1/2)|\sin\theta|$. The dashed line for $t = 0$ is the asymptotic distribution function (19). The hole at $\theta = 0$ is due to the maximum of the molecular potential energy (5) at this point.

of which will be given below (Eq. (13)). In a very strong field the oscillations of the molecular axis in the potential well with the minimum at the point (6) can be considered to have a rate essentially higher compared with the variation of electric field amplitude. The replacement $\sin\theta \to \theta,\ \cos\theta \to 1$, is possible in equation (4) for this case according to (8′), and the solution in the quasistatic limit [24], may be written similarly to the well-known quantum mechanical WKB-approximation:

$$\theta^2(t) = \frac{1}{\omega(t)}\left\{\sqrt{\frac{4M_z^2}{I^2}+\kappa^2}+\kappa\sin\left[\int_{t_0}^{t}\omega(t')\mathrm{d}t'+\varphi\right]\right\}, \tag{11}$$

$$\omega^2(t) = 2\beta F^2(t)/I. \tag{12}$$

This quasistatic approximation (11) is valid under the following condition:

$$\omega(t)\tau \gg 2\pi, \tag{13}$$

where τ is the pulse duration. Note that the frequency of the molecular axes oscillations (12) does not depend on the M_z value.

The constants κ, φ are to be found basing on continuous transition between the values of $\theta(t_0)$, $\dot{\theta}(t_0)$ determined by (11) on the one hand, and the same values for a free molecular rotation on the other hand. The time moment t_0 for the matching procedure may be naturally chosen according to the condition $F(t_0) \simeq F_\mathrm{c}$, F_c being determined by (9) where E is considered equal to the rotation energy of the free molecule. The formulae derived in such a way are sufficiently complicated, so it is not convenient to use them for the further analytical calculations. Therefore we restrict ourselves here to the case of very short laser pulse with the duration lower than the rotation period of the free molecule:

$$\tau \ll 2\pi\sqrt{\frac{I}{2E_0}}\,. \tag{14}$$

The value in the right-hand side of the inequality (14) for the $\mathrm{Cl_2}$ molecule is 0.3 ns. When the inequality (14) holds, the molecule position does not change during the field-on time interval. Thus, the matching time moment t_0 may be taken so that the inequality

$$\omega(t_0) \gg \omega_0 \tag{14$'$}$$

is satisfied. Then it is easy to see that the continuity of $\theta(t_0)$ may be provided for

$$\begin{aligned} &\kappa = \omega(t_0)\theta^2(t_0) \gg 2|M_z|/I = 2\omega_0|\cos\gamma|,\\ &\varphi \simeq 0, \end{aligned} \tag{15}$$

if of course, $\theta(t_0)$ is not too small. Evidently, the small values of $\theta(t_0)$ occupy a small region in the set of initial values, and the inequality (15) should be treated in this sense. The continuity of $\dot{\theta}(t_0)$ may be provided by an appropriate choice of the t_0 value, since this value is not determined exactly. The corresponding equation is

$$\omega(t_0) = \frac{\dot{\omega}(t_0)}{\omega(t_0)} + 2\frac{\dot{\theta}(t_0)}{\theta(t_0)}\,. \tag{16}$$

Under the inequality (14) the last term in the right-hand side of (16) can be neglected. The corresponding results become independent of $\dot{\theta}(t_0)$. This fact is completely clear from the physical point of view. Indeed, for the short laser pulses the initial angular distribution $\theta(t_0)$ is "frosen" and the variation of these angles caused by the initial velocities $\dot{\theta}(t_0)$ becomes negligible.

Equation (16) without the last term in the right-hand side should be considered as a transcendental equation for t_0. The numerical analysis demonstrates that the solutions of this equation give the value for $F(t_0)$, exceeding slightly the value of F_c. This situation is quite satisfactory since the orientational interaction energy of the molecule with radiation becomes equal to the energy of free rotation beginning with the field value $F = F_\mathrm{c}$. Let us consider a numerical example. For the $\mathrm{Cl_2}$ molecule, in the field of intensity 10^{15} W/cm^2 the electric field amplitude equals the value of F_c at the time moment $t_\mathrm{c} = -2.2\tau$, and the time moment corresponding to the solution of equation (16) is $t_0 = -1.66\tau$. It is evident from Figure 1 that for such values of t_0 the initial angular distribution is unable indeed to change noticeably. The value of $\omega(t_0)/\omega_0 > 800$ enables the inequality (14$'$) to hold for the most part of the angles $\theta(t_0)$. Nevertheless, the value of the parameter $\omega(0)\tau \simeq 6.6$ does not allow to judge about the validity of inequality (13) which is thus the most restrictive one for the theory presented here.

The use of the values of constants (15) in equation (11) results in neglecting completely the quantity M_z. Thus this approximation leads to the removal of the central hole in the distribution function which has the width $\sim \omega_0^2\cos^2\gamma/\omega^2(t_0) \ll 1$. The corresponding formula has the form:

$$\theta^2(t) = \frac{\omega(t_0)\theta^2(t_0)}{\omega(t)}\left\{1 + \sin\left[\int_{t_0}^{t}\omega(t')\mathrm{d}t'\right]\right\}.$$

Assuming the $\theta^2(t)$ dependence to be harmonic, the distribution function may be written as:

$$P(\theta) = \frac{2\omega(t)\theta}{\pi\omega(t_0)\theta^2(t_0)}\left\{1 - \left[1 - \frac{\omega(t)\theta^2}{\omega(t_0)\theta^2(t_0)}\right]^2\right\}^{-1/2}. \tag{17}$$

The values of θ in (17) should provide the subradical expression to be positive. Otherwise, $P(\theta) = 0$ should be taken.

Assuming the isotrope distribution for the quantity $\theta(t_0)$ in (17) and averaging over it, we obtain:

$$\begin{aligned} P(0 \le \theta \le \theta_\mathrm{max}) &= \frac{\pi L^{(s)}(z)}{2\theta_\mathrm{max}},\\ \theta_\mathrm{max} &\equiv \frac{\pi}{2}\sqrt{\frac{2\omega(t_0)}{\omega(t)}}, z \equiv \theta/\theta_\mathrm{max},\\ L^{(s)}(z) &\equiv \frac{2}{\pi}\int_1^{1/z}\frac{\sin(\pi x z/2)}{\sqrt{x^2-1}}\mathrm{d}x\\ &\simeq \sqrt{1-z^2}\left(1 - \frac{\pi^2}{72} - \frac{\pi^2 z^2}{36}\right),\\ P(\theta_\mathrm{max} < \theta \le \pi/2) &= 0. \end{aligned} \tag{18}$$

With account of the above approximation for the function $L^{(s)}(z)$, the distribution (18) is occurred to be normalised within a 2% inaccuracy. Assuming the angle θ to vary in the $[-\theta_\mathrm{max}, \theta_\mathrm{max}]$ range, the expression (18) may be rewritten as follows:

$$\begin{aligned} P(-\theta_\mathrm{max} \le \theta \le \theta_\mathrm{max}) = \frac{\pi}{4\theta_\mathrm{max}}&\left(1 - \frac{\theta^2}{\theta^2_\mathrm{max}}\right)^{1/2}\\ &\times\left(1 - \frac{\pi^2}{72} - \frac{\pi^2}{36}\frac{\theta^2}{\theta^2_\mathrm{max}}\right). \end{aligned} \tag{19}$$

As it is evident, the distribution (19) depends on a single parameter $\theta_{\max}$, determining the distribution function width. Similarly to the equilibrium position (8), $\theta_{\max} \sim (\beta S)^{-1/4}$. The distribution (19) is shown on the Figure 1c (dashed line) for the same values of the parameters that were used in the previous section. A 15% discrepancy between this function and the numerical calculations is due to that these parameters have not yet reached their asymptotic values.

5 Linear model for $\beta < 0$

As it was noted in Section 2, for $\beta < 0$, when the field is on, the bifurcations do not appear. The minimum of the free molecule potential energy $\theta = \pi/2$ in the field is only deepened without changing its position. It allows, within some assumptions, to derive an analytical solution of the problem.

In a strong field, for which the interaction energy exceeds essentially the free rotation energy, the first term in the right-hand side of (4) may be neglected. To meet the initial conditions describing the motion of a free molecule, the inequality (14) should also be fulfilled. Thus this section considers short laser pulses with high intensity.

It is convenient to take the potential energy minimum for the reference point of the angle θ, using the following substitution in (4):

$$\theta = \pi/2 - \chi, \qquad \chi \in [-\pi/2, \pi/2].$$

In addition, we linearise equation (4) in complete analogy with the linearisation of equation for the motion of a classical pendulum [24]: $\sin\theta \to 1$, $\cos\theta \to \chi$:

$$\frac{\mathrm{d}^2\chi}{\mathrm{d}t^2} = -\frac{\beta F^2(t)}{2I}\chi. \tag{20}$$

Equation (20) is linear and its analytical solution can be derived if we take

$$F(t) = F_0 \operatorname{sech}(t/2\tau) \tag{21}$$

for the field envelope. The function (21) provides an exponential decrease of the radiation intensity for $|t| \gg \tau$. Note in this regard, that a similar dependence $F(t) \sim \exp(-|t|/\tau)$ was assumed for the radiation intensity in reference [12].

The general solution of (20) with the right-hand side (21) may be written in terms of the Gauss hypergeometric functions [30]:

$$\begin{aligned}
&\chi(t) = c_1 f^{(-)}(t) + c_2 f^{(+)}(t),\\
&f^{(\pm)}(t) = F\left[\frac{1+a}{2}, \frac{1-a}{2}; 1; \frac{1}{2}\left(1 \pm \tanh\frac{t}{2\tau}\right)\right],\\
&a = \sqrt{1 + 4\Omega_0^2\tau^2}, \qquad \Omega_0^2 = 2|\beta|F_0^2/I,
\end{aligned} \tag{22}$$

$c_{1,2}$ being the integration constants. The quantity Ω_0 is similar to the frequency $\omega(0)$, determined in (12). But now the conditions (13) are not required for the validity of the used approximations.

The limit of expression (22) for $t \to -\infty$ may be determined with the use of equations 2.10(12–13) of reference [30]. So the constants $c_{1,2}$ may be related to the values of $\chi_0, \dot{\chi}_0$, determining the orientation of molecule before the field is on:

$$\begin{aligned}
\chi(t) &= f^{(+)}(t)\chi_0 + g(t)\tau\dot{\chi}_0,\\
g(t) &= \frac{\pi}{2\cos(\pi a/2)}\left[f^{(+)}(t)\sin\frac{\pi a}{2} - f^{(-)}(t)\right]\\
&\quad + \left[\psi(1) - \psi\left(\frac{1+a}{2}\right)\right] f^{(+)}(t),
\end{aligned} \tag{23}$$

$\psi(\cdot)$ is the logarithmic derivative of the Γ-function. The singularities of the function $g(t)$ at $a = 2n + 1$, where n is an integer, are removable. It can be easily demonstrated expressing the hypergeometric functions for these values of a in terms of the Jacobi polynomials which in this case are reduced to the Legendre polynomials [30]. Together with the condition (14) this fact allows us to neglect the term with $g(t)$ in the equation for $\chi(t)$. This approximation is equivalent to neglecting the second term in the right-hand side of equation (16). The numerical example considered in the preceding section demonstrates the accuracy of this approximation.

To derive the distribution function, let us use the relation

$$P_t(\chi) = \frac{1}{2}\int_{-\pi/2}^{\pi/2} P(\chi_0)\delta(\chi(t) - \chi)\mathrm{d}\chi_0.$$

Here $\chi(t)$ depends linearly on χ_0 according to (23) with account for the above approximations. Assuming $P(\chi_0) = (1/2)\cos\chi_0$, according to the uniform distribution of the angle θ_0, we obtain:

$$\begin{aligned}
P(|\chi| < \chi_{\max}) &= \frac{\pi}{4\chi_{\max}}\cos\frac{\pi\chi}{2\chi_{\max}},\\
P(|\chi| > \chi_{\max}) &= 0,\\
\chi_{\max} &\equiv \frac{\pi}{2}|f^{(+)}(t)|.
\end{aligned} \tag{24}$$

Equations (24) make clear that the quantity $\chi_{\max}$ has the meaning of the distribution width. Let us note a property of the function (24):

$$\lim_{\chi_{\max}\to 0} P_t(\chi) = \delta(\chi).$$

The function $f^{(+)}(t)$ plot is given in Figure 2 determining $\chi_{\max}$ for some values of a. It is seen that the restrictions for $|f^{(+)}(t)|$ appear with the increase of a. However, as for large values of a the function $f^{(+)}(t)$ oscillates, the distribution function $\langle P(\chi)\rangle$ averaged over these oscillations takes a physical sense. To derive $\langle P(\chi)\rangle$, we use the Watson's asymptotic formulae for the hypergeometric functions (22) with $a \gg 1$, given in [30] (Eq. 2.3(17)). The result is

$$f^{(+)}(t) \simeq \sqrt{\frac{\cosh(t/2\tau)}{2\pi\Omega_0\tau}} \times \cos\left\{2\Omega_0\tau \operatorname{arccot}[\sinh(t/2\tau)] + \pi/4\right\}.$$

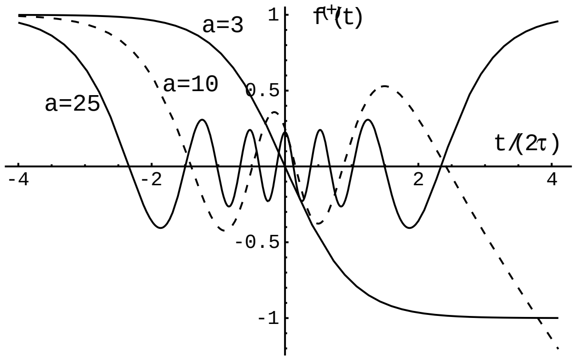

Fig. 2. The function $f^{(+)}(t)$ (22) graphs, which determines the distribution of the molecular axes in the case of $\beta < 0$, for different values of the parameter a.

The physical meaning of this expression is simply understood if we introduce the instantaneous frequency of the molecular axis oscillations relative to the field polarisation vector:

$$\Omega(t) = \Omega_0 \operatorname{sech}(t/2\tau) = \sqrt{2|\beta|F^2(t)/I}.$$

Then the formula may be rewritten as follows:

$$f^{(+)}(t) \simeq -(2\pi\Omega(t)\tau)^{-1/2}\cos\left(\int_{-\infty}^{t}\Omega(t')\mathrm{d}t' + \pi/4\right). \tag{25}$$

This expression is similar to the approximation used in the preceding section, since the large values of a correspond to the inequality (13).

Inserting (25) into (24), we carry out the averaging over the half-period $\pi/\Omega(t)$ considering $\Omega(t)$ to be constant

$$\langle P(|\chi| < \chi_{\max}^{(0)})\rangle = \frac{1}{2\chi_{\max}^{(0)}}\int_0^{\arccos z}\frac{\mathrm{d}x}{\cos x}\cos\left(\frac{\pi z}{2\cos x}\right),$$
$$\chi_{\max}^{(0)} \equiv (8\Omega(t)\tau/\pi)^{-1/2}, \qquad z \equiv |\chi|/\chi_{\max}^{(0)}.$$

By substituting $\sec x = y$, we transform this equation to the form similar to (18):

$$\langle P(|\chi| < \chi_{\max}^{(0)})\rangle = \frac{L^{(\mathrm{c})}(z)}{2\chi_{\max}^{(0)}},$$
$$L^{(\mathrm{c})}(z) \equiv \int_1^{1/z}\frac{\cos(\pi y z/2)}{\sqrt{y^2-1}}\mathrm{d}y. \tag{26}$$

The function $L^{(\mathrm{c})}(z)$ inside the interval $z \in (0,1]$ is approximated with the accuracy of $\sim 10\%$ by a simple logarithmic dependence $L^{(\mathrm{c})}(z) \simeq -\ln z$. Within this accuracy

$$\langle P(|\chi| < \chi_{\max}^{(0)})\rangle \simeq \frac{1}{2\chi_{\max}^{(0)}}\ln\frac{\chi_{\max}^{(0)}}{|\chi|}.$$

Higher accuracy may be obtained if we resolve the cosine function of the integrand in (26) in power series. The first three terms of this resolution give the equation

$$L^{(\mathrm{c})}(z) \simeq \left(1 - \frac{\pi^2 z^2}{16} + \frac{\pi^4 z^4}{2^{10}}\right)\ln\frac{1+\sqrt{1-z^2}}{z}$$
$$- \frac{\pi^2}{16}\left(1 - \frac{\pi^2}{96} - \frac{\pi^2 z^2}{2^{10}}\right)\sqrt{1-z^2},$$

which has the accuracy of $\sim 1.75\%$. Notice the logarithmic divergence of the averaged distribution function for $\chi \to 0$, which corresponds to the $\theta = \pi/2$ plane as written in usual coordinates. It is also easy to see that for $\Omega(t)\tau \gg 1$ the distribution function width determined by the value of $\chi_{\max}^{(0)}$ is $\sim (\beta S)^{-1/4}$, as it took place above.

6 Analysis of experimental data

As it was noted in Introduction, a lot of papers have been published with experimental data on anisotropy in directions of the molecular fragments departed due to the ionisation dissociation. Having no possibility to consider here all the results we shall only draw attention to some experiments related straightforwardly to the model presented.

It was noted in Section 3 that the total width of angular distribution, as obtained in numerical calculations in the frames of the classical mechanics for the molecule $\mathrm{Cl_2}$, agrees well with the experimental value determined for this quantity in reference [28]. Nevertheless, a qualitative difference exists between the calculations and the experiment [28]: the lack of the hole in the center of experimental curve for the angular distribution of the ions Cl. The minimum in the angular distribution of ions S at $\theta = 0$, which can be associated with “the hole”, was observed in [15] where the dissociation of the linear molecule $\mathrm{CS_2}$ was investigated (see also Fig. 2 in [16]).

One of the causes of this qualitative discrepancy between the experimental results [28] and [15,16] can be the difference between the intensities of the laser radiations: 10^{15} W/cm^2 in [28] and 10^{13} W/cm^2 in [15,16]. According to (8) the width of the hole decreases with the increase of the field intensity. The other reason for the difference between the theory and experiment [28] might be the following. Due to the ionisation, the surrounding free electrons and the nearest molecular ions influence on the orientation of the molecules, so that the axial symmetry of the problem is violated. Thus, the angular momentum component ceases to be an integral of motion, and the central hole of the distribution function disappears. The variation of the angular distribution of the ions $\mathrm{S^+}$ with variation of the ionised molecules density from 10^9 cm^{-3} to 10^{11} cm^{-3} was observed in experiment [16]. This variation was explained to be due to the variation of the ion density in the region of the laser light focusing. A similar effect of non-conserving angular momentum and its projection was recently studied theoretically for the Rydberg molecular states in [32,33]. However this theory cannot be transferred directly to the case of the molecule alignment in field which is considered here. The development of such a theory seems to be an independent problem.

The existence of the hole does not follow from the use of the classical mechanics since in quantum calculations the conservation of the angular momentum component must provide the same results. The only result of classical description which is changed taking into account the spatial quantisation of the molecular rotations, is that the probability of $M_z = 0$ value becomes nonzero. This probability is easily calculated in the limit of $M^2/2I \ll kT$ [31]:

$$P(M_z = 0) \simeq \frac{\hbar}{\sqrt{8IkT}} \cdot \qquad (27)$$

In other words, the angular distribution function for the molecular axes is not zero at $\theta = 0$ but equals to the value determined by equation (27). *E.g.* for the Cl_2 molecule we obtain $P(M_z = 0) \simeq 0.01$. Thus evidently, the quantum effects do not remove this anomaly in the molecular axes distribution function, maybe except for the lightest molecules.

In [16,34] the ionisational dissociation for the molecules NO_2 was investigated. The angular distribution observed for the O^+ ions qualitatively agrees with the results of this work for the case of $\beta > 0$, whereas the results for the NO^+ ions agree with the case of $\beta < 0$. This circumstance may be explained by the assumption that the O^+ and NO^+ ions are produced from different electronic states and the molecule NO_2 or its ion remains sufficiently long time in this state, so the molecule alignment occurs resulting in the observed anisotropy in the directions of the departed fragments. If it is so, results obtained in [16,34] demonstrate an important role of excited electronic states for a laser field-induced alignment of molecules.

The results obtained qualitatively in this work are confirmed by recent experiment [35], where the alignment of light molecules H_2, N_2 was observed in a field with the pulse duration of 50 fs. For the heavy molecules I_2, the anisotropy of the ion departure can be explained by dependence of the electron tunneling on the orientation of the molecule with respect to the laser beam polarisation [6,7]. However, the pulse duration of 15 fs is not enough for alignment even of light molecules [36].

The results presented above make clear that the anisotrope scattering of fragments in the ionisation dissociation of molecules is complicated substantially. The author hopes, nevertheless, that the classical theory presented in this article for the simplest 2-atomic molecules will help to understand some aspects of this phenomenon.

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