# Mechanism of preparation of an active medium for generation of a field with high moment of momentum

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A mechanism is proposed for preparation of an active medium from diamagnetic "molecule + field" systems with artificial symmetry in a biharmonic pump filed.

## 1. Background

As known,[1] radiation carries fluxes of energy, momentum, and moment of momentum away from a source. The value of moment of momentum (MM) depends on the vector of radiative energy flux **S** and on the structure of the field (expansion into a series over powers of the vectors **E**, **H**, and **k**). At an ordinary structure of the field, it acquires MM, which is $\mathbf{J}_p \approx \hbar$ only at the field strength $E \geq 10^9$ V/cm. Here **E** and **H** are the strengths of the electric and magnetic fields in CGSE units; $\mathbf{k} = 2\pi n/\lambda$ is the wave vector; $n$ is the refractive index of the medium; $\lambda$ is the radiation wavelength; $\hbar$ is the Plank constant (quantum of action and the unit of MM).

Tvorogov[2,3] was the first to mention the possibility for a field with high MM at a moderate strength to exist. Within the framework of quantum electrodynamics, he showed a version of the field structure, at which it can carry the MM $\mathbf{J}_p \gg \hbar$. However, in his papers it was not shown how to prepare an active medium for generation of the field with high MM.

The interest to radiation with $\mathbf{J}_p \gg \hbar$ is not only academic, but practical too. Such a radiation induces high magnetic moment **M** in a unit volume of a medium at the optical frequency $\omega$, thus changing medium reflectivity (absorptivity). Consequently, if we succeed in obtaining generation of coherent radiation with high MM, then measurement *technologies* can be radically renovated in many fields employing laser radiation, including diagnostics of the environment, interaction control, and prediction of light propagation in the medium.

In our earlier papers, [4,5] we proposed a new approach to preparation of the active medium from an ensemble of diamagnetic (potentially magnetic) "molecule + field" systems. This approach opens up the possibility of generating a field with high MM. An ensemble of "molecule + field" systems is understood in the ordinary treatment of the problem within the framework of the full quantum theory. In this paper,

we consider the mechanism for preparation of an active medium (source of radiation) from an ensemble of diamagnetic "molecule + field" systems.

## 2. Peculiarity of new approach to preparation of an active medium

The traditional approach to solution of laser physics problems involves frequency resonance between light and a quantum system (molecules, atoms) and the energy conservation law. The idea of preparing an active medium within the framework of this approach is based on combining the effects of absorption, spontaneous and stimulated emission. The controlled parameters are population of stationary levels (states), the state of molecule as a whole, and its wave function

$$\Psi(t) = \psi^0 \exp(-iWt/\hbar). \qquad (1)$$

In practice, a volume cavity with a positive feedback (PF) between the radiation generated at the frequency $\omega$ and the active medium at time $t$ is usually used for this purpose. The wave function (1) accounts for the wave properties of a molecule and describes the state of the molecule with the energy $W$ at time $t$. Here $\psi^0$ is the amplitude; $\omega = W/\hbar$ is the angular frequency characterizing the corresponding molecular energy level.

To provide for generation of the field with MM $\mathbf{J}_p \approx 200\hbar$ within this approach, it is needed to obtain the population inversion between the stationary rotational states $|0\rangle \leftrightarrow |J = 100, M_J\rangle$. Here $J$ and $M_J$ are the quantum numbers of the total angular momentum of a molecule and its projection onto the direction of polarization of the perturbing field.

As known,[6,7] the probability of rotational transitions in a molecule with $\Delta J = 100$ between stationary rotational levels is almost infinitely low even at the perturbing field strength $E \geq 10^9$ V/cm. Therefore, it is practically impossible to induce the population inversion between molecular rotational levels $|0\rangle \leftrightarrow |J = 100, M_J\rangle$ and, consequently, to generate the field with high MM.

The conclusion thus can be formulated that the problem of generation of a field with high MM should be solved within the framework of a new approach to preparation of the active medium. A possible way is to use acceleration of bonding electrons in a molecule by **E**-component of light (pumping field) at the time $t'_\kappa$ ($\kappa = 0, 1, 2, …, J$) and to control their motion by **H**-component at the time $t''_\kappa$, i.e., to use the optical analog of the Veksler synchronous acceleration effect[8] for a molecular rotator as a nonlinear system. This process can be initiated through phase resonance between light and molecule ("resonance capture of phase"), and accumulation of MM in a molecule can be followed through PF between its state and the contributions of **E**- and **H**-components of light field to the spatial motion of bonding electrons at certain instants $t_\kappa$.

The term bonding electrons is applied to the electrons confining atoms in a molecule. As known, the bonding electrons are the main electric and magnetic elements determining optical properties of a molecule.[9] By spatial motion of the bonding electrons we mean the motion of their orbital and spin subsystems.

The "resonance capture of phase" means the coincidence of the following products:

$$\omega(t''_\kappa)\, t''_\kappa \approx W(t'_\kappa)/\hbar\, t'_\kappa, \qquad (2)$$

where $\omega(t''_\kappa)$ and $W(t'_\kappa)$ are the carrier frequencies of the perturbing light and the quantum energy of a molecule at the moments $t''_\kappa$ and $t'_\kappa$ in time.

If molecules start from the rotational level $|J = 1\rangle$ and interact with light due to square Stark and Zeeman effects by the $V$-scheme

$$|1\rangle \leftrightarrow |0\rangle \leftrightarrow |J, M_J\rangle \qquad (3)$$

through the lowest rotational state $|0\rangle$, then the sought process can be obtained in the following way. The direction and value of the shift—splitting of rotational levels (3) of the molecule should be specified through the spatial structure and strength of the light field so that:

– the "resonance capture of phase" of light and molecule (2) keeps at least during the critical (from the viewpoint of comparison of intramolecular interactions with the separation between the levels of the main rotational transition $|1\rangle \leftrightarrow |0\rangle$) time $T \approx 1/B_e \approx \approx 10^{-11}$ s;

– potential magnetic (diamagnetic) and potential electric interaction energies are transferred through the channel

$$|1\rangle \rightarrow |0\rangle \Rightarrow |J, M_J\rangle \Rightarrow |0\rangle \Rightarrow ….. |J, M_J\rangle; \quad (4)$$

– variations of the electron polarizability $\alpha''(\omega) + + \sum\kappa\delta\alpha_\kappa$ and diamagnetic susceptibility $\chi_{dnJ}(\omega) + + \sum\kappa\delta\mu_\kappa$ are resonantly intensified and accumulated in the molecules simultaneously with the accumulation of MM $\mathbf{J}_p + \sum\kappa\delta\mathbf{J}_{p\kappa}$ for the time $T \approx 1/B_e$.

It is clear that this process can arise only in a weak range of pumping field parameters provided that

the optimal PF exists between the state of the molecule and the contributions of **E**- and **H**-components of the field to the spatial motion of bonding electrons at neighboring moments in time $t_\kappa$ and $t_{\kappa+1}$. In this case, parametric (phase dependent) interaction of molecular and field subsystems and, as a consequence, *self-organization* of "molecule + field" systems with *artificial symmetry* may prove possible. The molecular wave function (1) transforms into the form describing not only the state of the molecular and field subsystems, but also their interaction in space and time. Here $B_e = \hbar^2/(4\pi I_{bb})$ is the rotational constant of a diatomic diamagnetic molecule at the time $t''_{\kappa=0} \approx 0$; $I_{bb}$ is the molecular moment of inertia about the axis of revolution at the time the interaction with the field turns on $t''_{\kappa=0} \approx 0$.

## 3. Mechanism of self-organization in an ensemble of "molecule + field" diamagnetic systems

Consider a system consisting of the following subsystems: quantum field, electron motion in molecule, and rotation of molecule as a whole, as well as interactions of the field with electrons and the electron motion with molecular rotation. Whenever necessary, we pass, in physical considerations, from quantum characteristics of the field, electrons, and molecules to the corresponding classical analogs. Besides, as the rotation of the molecule is nearly classical and the molecule-rotator is a nonlinear system, we should use the results and methods of nonlinear dynamics. In this formulation of the problem, the field and molecules form a single quantum system in the sense that if the wave function of such a system is constructed, then it should include variables of both the field and molecule.

Below we give physical reasons for obtaining the active medium from an ensemble of diamagnetic "molecule + field" systems at a moderate energy flux of the perturbing field **S**.

The idea of preparing an active molecular medium within the framework of the new approach is based on non-additive summation of the diamagnetic component of the magneto-optic (Faraday) effect and its electro-optic analog at high $\omega^*_{s\kappa}$ and low $\Omega^*_{\kappa,\kappa+1}$ frequencies of the field generated in molecules in the form of orbital electron current under the exposure to the pumping wave. As a pumping wave, we take two linearly polarized waves at the frequencies $\omega_1$ and $\omega_2$ propagating along the $X$ axis.

We assume that this electro-magneto-optic (EMO) mechanism provide for self-organization in diamagnetic molecules of the medium under the exposure to the biharmonic ($\omega_1$ and $\omega_2$) pump field due to cooperation of the dynamic Veksler,[8,11] Stark,[7,9,10] and Zeeman[10] effects. This turns on the hysteresis mode of accumulation of EMO characteristics, diamagnetic

energy $\sim H_0^2$, potential electric energy $\sim E_0^2$, and MM $\Sigma \kappa \delta \mathbf{J}_{p\kappa}$ by bonding electrons of molecule up to *critical values*. The hysteresis mode leads to evolution of a certain set of diamagnetic molecules in the external field into an ensemble of diamagnetic "molecule + field" systems for, on the average, $T \approx 1/B_e \approx 10^{-11}$ s.

When the period $T \approx 1/B_e$ terminates, the active medium of the ensemble of "molecule + field" diamagnetic systems self-excites and "initiate" generation of radiation with high MM at the *cooperative* frequency $\omega_p \approx \omega_{s\kappa}^* \approx \omega_{0J}$. Here $\omega_{0J} = = B_e J(J+1)$ and $\omega_{01} = 2B_e$ is the frequency of the main $|0\rangle \leftrightarrow |J=1\rangle$ and overtone $|0\rangle \leftrightarrow |J, M_J\rangle$ molecular rotational transitions neglecting the energy of centrifugal stretching $\Delta W_J = -D\,[J(J+1)]^2$; $D \approx 10^{-4} B_e$ is the constant of centrifugal stretching of the molecule.

Let us explain how the diamagnetic susceptibility, MM, and diamagnetic energy increase, taking as an example diamagnetic atoms (Ag, Ar, Bi), in alternating (at the frequency $\Omega$) magnetic field. In Section 4 we justify and in Section 5 present the conditions for initiation and intensification of the EMO mechanism and, correspondingly, EMO effect for diamagnetic molecules ($N_2$, $CO_2$, AgCl) starting from the rotational level $|J=1\rangle$ and interacting by the $V$-scheme (3) with the linearly polarized biharmonic ($\omega_1$ and $\omega_2$) optical radiation propagating along the $X$ axis.

Diamagnetism is the property of an atom (molecule) in the magnetic field to magnetize in the direction opposite to the direction of the external magnetic field (**H**). Currents inducing diamagnetism in isolated atoms (molecules) arise due to the Lorentz force

$$\mathbf{F} = e\mathbf{E} + e\,[\mathbf{v}\,\mathbf{H}]/c,$$

which affects the electron having the mass $m_e$ and the charge $e$ and moving in the nucleus-fixed coordinate system with the velocity $v$ in the perturbing electric **E** and magnetic **H** fields.

Consider how the Lorentz force affects the motion of the free electron and the electron in an atom.[11,12]

If $\mathbf{E} = 0$, $\mathbf{H} \neq 0$, and the force **F**, with which the homogeneous constant magnetic field affects the free electron, is perpendicular to the vectors **v** and **H**, then it does no work, but only distorts the electron trajectory (Larmor circle), not changing its energy, i.e., the Lorentz force **F** only controls the electron motion.

If $\mathbf{E} = 0$, $\mathbf{H} \neq 0$, and the velocity of the free electron $v$ is directed at an angle $\gamma$ to the vector **H** in the homogeneous constant magnetic field, then under the effect of the force $\sim (e/c)\,vH\sin\gamma$ this velocity consists of two parts. Namely, the electron moves with the velocity $v_\parallel$ along **H** and with the Larmor velocity $v_\perp$ normally to **H**. The Lorentz force plays the role of a centripetal force $\mathbf{F} = m_e v_\perp^2/R_L$. In this case, the electron moves along a Larmor spiral having the radius

$R_L = cm_e v_\perp/eH = v_\perp/\Omega_L$, and the Larmor (cyclotron) angular rate of electron rotation (precession of the electron orbit) is $\Omega_L = eH/m_e c$. As this takes place, the center of the circle of the Larmor spiral shifts along the force line $H$ with the velocity $v_\parallel$.

If $\mathbf{E} = 0$ and $d\mathbf{H}(z, t)/dt \neq 0$, then the motion of the electron in the atom potential (field) leads to the increase of MM and diamagnetic energy of the atom. Both additions result from generation of the circulating electric field $\mathbf{E}_y'$ inside the atom due to magnetic induction at variation of the inducing magnetic field $d\mathbf{H}(z, t)/dt$ (Ref. 12). According to Faraday law, the contour integral of $\mathbf{E}_y'$ over a closed contour $\Gamma$ is equal to the rate of variation of the magnetic flux through the contour. If we take a circle with the radius $r$ and the center coinciding with the atom nucleus as the contour $\Gamma$, then the mean tangential electric field (orbital electron current)

$$\mathbf{E}_y'\, 2\pi r = -d(\mathbf{H}(z,\,t)\,\pi\,r^2)/dt$$

with the strength

$$\mathbf{E}_y' = (-\,r/2)\,d\mathbf{H}(z,\,t)/dt$$

is generated on this contour due to magnetic induction. This induced electric field affecting the atom electron increases its orbital speed $v_\perp = r\,\Omega_L$ and creates the moment of force $-e\,\mathbf{E}_y'\,r$, which must be equal to the rate of MM variation

$$d\mathbf{J}/dt = (er^2/2)\,d\mathbf{H}(z,\,t)/dt. \qquad (5)$$

Integrating over time starting from the zero field, we obtain that the additional MM imparted to the electron for the period of variation of the field $\mathbf{H}(z, t)$ is equal to

$$\Delta\mathbf{J} = er^2/2H_0$$

and connected with the precession of the electron orbit along the Larmor spiral of the radius $r_L \approx r$

$$v_\perp = r\,\Omega_L = \sqrt{2}\,\Delta\mu\,H_0/m_e \qquad (6)$$

at the Larmor frequency $\Omega_L = eH_0/2m_e$ ($\Omega_L \approx 10^{13}$ Hz in the atom at $H \sim 10^6$ E, CGSE units).

The additional MM $\Delta\mathbf{J}$ produces the increment $\Delta\mu$ of the diamagnetic susceptibility $\chi_d(\Omega)$ that, consequently, leads to the increase of the diamagnetic energy $W_{dn}$ of the atom. Since the vectors of magnetic moment $\mathbf{\mu}$, and the moment of momentum $\mathbf{J}_p$ for the orbital motion of the electron are related by the equation $\mathbf{\mu} = -\mathbf{J}_p e/2m_e$ and directed normally to the orbit plane, the value of the diamagnetic moment induced by one electron proves to be equal to

$$\Delta\mu = -\Delta\mathbf{J}_p\,e/2m_e = (-e^2\,r_e^2/4m_e)\,H_0 =$$
$$= (-e^2\,<r^2>/6m_e)\,H_0$$

and is observed as a contribution to diamagnetic susceptibility $\chi_d(\Omega)$ having negative sign and being

temperature independent. Here $<r^2>$ is the mean square separation of the electron from the atom nuclei (from the center of gravity (c.g.) of molecule); $H_0$ is the field strength amplitude. The minus sign means that the direction of the induced moment $\Delta\mu$ is opposite to the magnetic field (according to the Lenz's law), i.e., *the induced orbital current* is directed so that it "resists" the magnetic field $\mathbf{H}(z, t)$ increase. The orbital electric current $e\mathbf{E}'_y$ lives as long as the field $\mathbf{H}(z, t)$ is present in the atom, because the energy of the currents inside atom (molecule) does not dissipate in an atom (molecule) being in a stationary state.[9,12]

Thus, the induced orbital current caused by precession of the electron orbit with the Larmor frequency $\Omega_L$ (first-order Zeeman effect[9]) leads to the diamagnetic energy (the spin current makes no marked contribution) caused by the second-order Zeeman effect.[9]

The induced diamagnetic energy of the atom (molecule)

$$W_{dn} = -\mu_B\, e/(4\hbar c) <r^2> H_0^2$$

is considered as a potential magnetic energy of the electron, because it explicitly depends only on the electron position in the plane $XY$ ($r^2 = x^2 + y^2$) on the $n$th orbit. Here $\mu_B = e\hbar/(2m_e c)$ is the Bohr magneton, in erg / Gs.

The effect of increase of the atomic diamagnetic susceptibility $\Delta\mu$ can be intensified at the Larmor frequency $\Omega_L \approx \Omega$, if the technique of Rabi-resonance enhancement is used.[10]

The diamagnetic energy was neglected when considering properties of isolated atoms (molecules), because it is small as compared to Zeeman energy linearly dependent on $\mathbf{H}$ and connected with the precession of the electronic magnetic moment $\mu$ at the Larmor frequency $\Omega_L$. For a set of atoms (molecules), the Zeeman energy vanishes (although approximately) at averaging of the square distance from the electron to the atomic nuclei (to the molecular c.g.),[10] and the role of the diamagnetic energy, in particular, in the interaction of a molecule with the perturbing field, increases.

The significance of the diamagnetic energy increases considerably under conditions allowing the increase of the radius of the Larmor spiral of the induced orbital current $r \approx r_L$ and accumulation of MM by bonding electrons, for example, due to intensification of not only precession (low-frequency) motion of the electron orbit, but also nutation (high-frequency) motion of electrons through superposition of low- and high-frequency orbital electron perturbations of a set of molecules.

The nutation motion of the electron orbit in the external field arises due to rotation of the molecule-rotator as a whole and always accompanies the precession motion of electrons about the vector $\mathbf{H}$. The nutation motion of the electron orbit manifests itself in

oscillation of some axis $K(t)$, whose period decreases, as the angular rate of the natural rotation of the molecule-rotator increases (by analogy with a top[11]):

$$T_N \approx 2\pi I_z(t)/(I_{bb}\,\omega_{0J}). \qquad (7)$$

The nutation motion of the electron orbit was earlier ignored in problems of laser magnetic spectroscopy because of quick damping.[13] Here $I_z(t)$ is the moment of inertia with respect to the symmetry axis $K(t)$ passing through the c.g. and coinciding with the molecular axis $R'$ at the time, when the interaction of the molecule with the field turns on $t''_{\kappa=0} \approx 0$.

We assume that if the conditions are created for superposition of high- and low-frequency quantum fluctuations of molecular energy (directions of orbital and rotational MM), as well as for the PF between the state of molecules and contributions of $\mathbf{E}$- and $\mathbf{H}$-components of the pumping field by the $V$-scheme to the precession-nutation (PN) motion of the bonding electrons at neighboring $\kappa$th and $(\kappa + 1)$th time intervals, then the increase of the effective radius of the PN motion $r \approx r_L \to R^*$ self-organizes due to the quantum component of the pumping field, i.e., the EMO mechanism self-organizes in diatomic molecules: molecules resonantly (at the frequencies $\Omega^*_{\kappa,\kappa+1}$ and $\omega^*_{s\kappa}$) enhance and accumulate the MM $\mathbf{J}_p + \Sigma\kappa\delta\mathbf{J}_{p\kappa}$, diamagnetic susceptibility $\chi_{dnJ}(\omega) + \Sigma\kappa\delta\mu_\kappa$, and diamagnetic energy $\Sigma\kappa\delta W_{Z,\kappa} \sim H^2$, as well as the electron polarizability $\alpha''(\omega) + \Sigma\kappa\delta\alpha_\kappa$ and the potential electric energy $\Sigma\kappa\delta W_{S,\kappa} \sim E^2$. The field $\mathbf{E}'_y$, $\mathbf{H}'_z$ at the low $\Omega^*_{\kappa,\kappa+1}$ and high $\omega^*_{s\kappa}$ frequencies is generated in molecules (orbital electron current) under the effect of biharmonic ($\omega_1$ and $\omega_2$) pumping wave.

It is clear that the conditions for superposition of low- and high-frequency orbital electron perturbations, as well as for the PF between the state of molecules and contributions of the pumping field to the PN motion of the bonding electrons at neighboring $\kappa$th and $(\kappa + 1)$th time intervals *must be provoked* by relation of the low- and high-frequency motions of the perturbing field strength in space and time.

# 4. Justification of conditions for generation and intensification of PN motion of electrons in diamagnetic molecules under the exposure to a biharmonic pump wave

## 4.1. Preparation of parameters of the biharmonic pump wave

To compose and justify the relationship between the low- and high-frequency motions of the perturbing field strength in space and time, let us present two linearly polarized waves propagating along the $X$ axis

through amplitude-phase (AP) modulation of the resulting strength of the biharmonic pump field[14]:

$$\mathbf{E}(y,\, t) = e^{-ij\theta}\, e^{i\varphi}\, [E_1 \exp -i\, (kx + \alpha_y - \omega_1 t + \psi_1) +$$

$$+ E_2 \exp -i\, (kx + \alpha_y - \omega_2 t + \psi_2)] \equiv$$

$$\equiv e^{-ij\theta}\, e^{i\varphi}\, A(x,\, t) \exp i\Phi_s(x,\, t), \qquad (8a)$$

$$\mathbf{H}(z,\, t) = e^{-ij\theta}\, e^{i\varphi}\, [H_1 \exp -i\, (kx - \omega_1 t + \psi_1) +$$

$$+ H_2 \exp -i(kx - \omega_2 t + \psi_2)] \equiv$$

$$\equiv e^{-ij\theta}\, e^{i\varphi}\, A'(x,\, t) \exp i\Phi'_s(x,\, t). \qquad (8b)$$

Hereinafter, the spatial coordinates are sometimes omitted for simplicity.

The vector $\mathbf{E}(t)$ rotates within the polarization diagram. The polarization diagram is an ellipse (at $\varphi \neq 0$), whose size varies in time as $\cos\Delta\omega t / \cos\omega_s t$, and the orientation ($\theta$) and shape ($\varphi$) depend on the amplitude of the vectors $\mathbf{E}_1$ and $\mathbf{E}_2$ and on the half-sum of the initial phases of the biharmonic waves

$$\psi_s = (\psi_1 + \psi_2)/2;\ \ \Delta\omega = (\omega_1 - \omega_2),\ \ 2\omega_s = (\omega_1 + \omega_2).$$

In Eqs. (8):

− $\theta$ and $\varphi$ are the angles of orientation and ellipticity of the polarization diagram. The orientation angle $\theta$ (depends on the angle $\gamma$ between the vectors $\mathbf{E}_1$ and $\mathbf{E}_2$) and the ellipticity angle $\varphi$ (depends on the amplitude ratio $\mathbf{E}_1/\mathbf{E}_2$) determine the degree of inhomogeneity of the field strength (8a) and (8b) along the axes $Y$ and $Z$ at its homogeneous distribution along the axis $X$;

− $\alpha_y$ and $\alpha_z$ are the phase coefficients[15] characterizing the state of polarization of the field strength $\mathbf{E}(t)$ along the $Y$ and $Z$ axes at the time $t$ for the wave propagating along the $X$ axis, $\alpha = \alpha_y - \alpha_z$. The resulting radiation is linearly polarized in two cases: (a) at $\mathbf{E}(z, 0) = 0$ and (b) at $\mathbf{E}(y, 0) = \mathbf{E}(z, 0)$ and $\alpha = 0, \pi$ (Ref. 15). An example of the case (b): if at time $t = 0$ we have $\mathbf{E}(y, 0) = \mathbf{E}(z, 0)$ and $\alpha = 0$ [$\alpha = \pi$], then $\mathbf{E}(y, 0)$ and $\mathbf{E}(z, 0)$ are in phase (antiphase). In this case, the plane of radiation polarization divides the angle between the axes $y$ and $z$ [$y$ and $-z$], and the oscillations themselves are in quadrants ($y, z$) and ($-y, -z$) [($y, -z$) and ($-y, z$)]. The resulting radiation is elliptically polarized, if for $\mathbf{E}(y, t)$ and $\mathbf{E}(z, t)$ we assume $\alpha = \pm\pi/2$. The second case is known less than the first one, but it is just its modification that is used to change the state of polarization of the pumping wave (8) in the molecular medium and the state of molecules (8) in the field;

$$A(t) = E_0\, f(t)^{1/2} \text{ and } f(t) = [1 + m\cos2\Delta\phi(t)]$$

are slowly ($T_{am} \approx 4\pi c/\Delta\omega$) varying envelope of the amplitudes of $E$- and $H$-components of the resulting strength of the biharmonic field (RSBF) at the frequencies $\omega_1$ and $\omega_2$ and the function modulating the RSBF amplitude with the frequency $\sim\Delta\omega/2$; $A'(t) = H_0\, f'(t)^{1/2}$ is the envelope of the field amplitudes $H_1$ and $H_2$;

$$m = 2E_1 E_2/E_0^2 \text{ and } \beta = (E_1^2 - E_2^2)/E_0^2$$

are the depth of the amplitude modulation and the index of the phase modulation of the RSBF; $E_- = (E_1 - E_2)$, $E_+ = (E_1 + E_2)$; $E_0 = (E_1^2 + E_2^2)^{1/2}$;

$$2\Delta\phi(t) = (\Delta\omega t + \Delta\psi);\ \Phi_s(t) = (\omega_s t + \psi(t) + \psi_s);$$

$$\Delta\psi = (\psi_1 - \psi_2) \text{ and } 2\psi_s = (\psi_1 + \psi_2)$$

are the difference and the sum of the initial phases of the biharmonic waves;

$$\psi(t) = \arctan\,[(E_-/E_+)\,\tan(\Delta\omega t + \Delta\psi)/2] +$$

$$+ (\pi/2)\,\mathrm{sign}[(E_-/E_+)\,\sin(\Delta\omega t + \Delta\psi)/2] \times$$

$$\times \{1 - \mathrm{sign}\,[\cos(\Delta\omega t + \Delta\psi)/2]\}$$

is the instantaneous value of the additional phase change of the RSBF

$$\mathrm{sign}[a] = 1 \text{ at } a > 0 \text{ and } = -1 \text{ at } a < 0;$$

$$-\omega(t) = d\Phi_s(t)/dt = \omega_s + \Omega(t) \text{ and } \omega^*_{s\kappa} = \omega_s + \Omega(t''_\kappa)$$

is the instantaneous value of the carrier frequency of the RSBF and its value at the moment $t''_\kappa = \kappa T_{s,\kappa}/2$ (at the time of intersection of the axis $X$); $\Omega(t) = d\psi(t)/dt$;

$$\Omega(t'_\kappa) = \beta\Delta\omega/2/[1 + m\cos2\Delta\phi(t'_\kappa)]$$

is the deviation of the instantaneous carrier frequency $\omega(t)$ from its mean value $\omega_s$ at the time $t'_\kappa = (\kappa - 1/2)\, T_{s\kappa}/2$, when the value of the electric vector achieves its maximum value $\mathbf{E}(t'_\kappa) = \pm\mathbf{E}_{\max}$ (at $\omega_1/\omega_2 > 1$);

$$T_{s\kappa} = 2\pi/\omega^*_{s\kappa} =$$

$$= 2\pi/\{\omega_s + (\beta\Delta\omega/2)/[1 + m\cos2\Delta\phi(t''_\kappa)]\} \approx 10^{-15}\,\mathrm{s}$$

is the period of variation of the vector $\pm\mathbf{E}(y, t)$ direction in the plane $YX$ [variations in the direction of rotation of the magnetic induction vector $\mathbf{B}(z, t)$ in the plane $ZX$ about the vector $\pm\mathbf{E}(y, t)$] at the time $t''_\kappa$, when the absolute value of the vector $|\mathbf{E}(y, t)|$ achieves its minimum value $\mathbf{E}(t''_\kappa) = \mathbf{E}_{\min}$ (at $\omega_1/\omega_2 > 1$). Hereinafter the period $T_{s\kappa} \approx (t''_\kappa - t''_{\kappa+2})$ is called the "plane" period because of additively combining the results of action of $\mathbf{E}$- and $\mathbf{H}$-components of the field (8) on the medium;

$\kappa = 0, 1, 2, \ldots, J$ is the number of the "plane" period $T_{s\kappa}$ of two accelerations (time $t'_\kappa$ and $t'_{\kappa+1}$) of the vector $\mathbf{E}(y, t)$, as well as two controls (time $t''_\kappa$ and $t''_{\kappa+1}$) of the vector $\mathbf{H}(z, t)$;

$$T_{vp} \approx \Sigma\kappa T_{s\kappa} \approx J(J + 1)T_{s\kappa}/2$$

is the set of "plane" periods $T_{s\kappa}$ having the meaning of the duration of the RSBF wave packet;

$$\Omega^*_{\kappa, \kappa+1} = \omega(t'_\kappa) - \omega(t'_{\kappa+1})$$

is the deviation of the carrier frequency $\omega_s$ in between the moments $t'_\kappa$ and $t'_{\kappa+1}$.

The deviation $\Omega^*_{\kappa,\kappa+1}$ plays the role of a generator connecting the changes in the vectors $\mathbf{E}_+$ and $\mathbf{E}_-$ ($\mathbf{H}_+$ and $\mathbf{H}_-$) at the frequency $\Delta\omega$ in the medium at the neighboring time moments $t'_\kappa$ with the RSBF phase change in the same medium at the instantaneous carrier frequency $\omega^*_{s\kappa}$ for the time interval

$$10^{-15} \text{ s} \approx \pi/\omega_s \ll \Delta t \ll 4\pi/\Delta\omega \approx 10^{-11} \text{ s}. \quad (9)$$

The sign and value of the deviation $\Omega^*_{\kappa,\kappa+1}$ characterize *the direction and depth (duration) of the relation* of the waves of electric and magnetic induction at the frequencies $\omega_1$ and $\omega_2$ in the molecular medium with the field generated in molecules by the field at the frequency $\omega^*_{s\kappa}$ within the coherency volume

$$V \approx \pi R_0^2 \, \Delta L. \quad (10)$$

Here $R \le R_0 \approx \sqrt{(v_{\mathrm{ph}}\, 2\pi\Delta L/\omega_s)}$ is the radius of the medium layer "trapped" by the RSBF (8) in the plane $YZ$ at the entrance into the medium $X = 0$; $\Delta L \approx v_{\mathrm{ph}}\pi/\delta\omega$ is the length of transient coherence of the pumping field; $\delta\omega \approx \delta\omega_1 \approx \delta\omega_2 \ll |\omega_1 - \omega_2|$ is the width of spectral components of the pump field; $v_{\mathrm{ph}} = c/n$ is the phase velocity of the resulting wave (8) in the medium; $n = \sqrt{\varepsilon\chi}$, $\varepsilon$, $\chi = \chi_p + \chi_d$ are the refractive index, permittivity, and the sum of paramagnetic and diamagnetic susceptibilities of the medium at the instantaneous carrier frequency $\omega(t)$ varying with the period $T_{s\kappa}$ about its mean value $\omega_s$ in the time interval (9).

If $(E_1/E_2)^2 = 1$, then the deviation $\Omega^*_{\kappa,\kappa+1} = 0$ ($\psi(t) = 0$) and the ellipticity angle $\varphi = 0$, and the resulting polarization diagram for the vector $\mathbf{E}(r, t)$ is shaped as a plane (line) at any $t_\kappa$. This case has been thoroughly studied in laser spectroscopy: the effects associated with forced oscillation of the population difference between levels and phasing of intramolecular motions at the phase difference $\Delta\omega$ of the pumping field were discovered.[9]

If $(E_1/E_2)^2 \ne 1$, then the deviation $\Omega^*_{\kappa,\kappa+1} \ne 0$ ($\psi(t) \ne 0$) and the ellipticity angle $\varphi \ne 0$, and the resulting polarization diagram on the $X$ axis is shaped as a plane (line) only at certain moments $t_\kappa$. At other moments, the polarization diagram has the shape of an ellipse (circle), whose spatial orientation $\theta$ is determined by the angle $\gamma$ between the vectors $\mathbf{E}_1$ and $\mathbf{E}_2$, i.e., the angle $\gamma \ne 0$ determines the orientation of the polarization diagram $\theta$ with respect to the axis $X$ and thus determines some axis $X'$ of symmetry of the pumping wave (8) in the medium.

The deviation $\Omega^*_{\kappa,\kappa+1} \ne 0$ may be a synergetic generator of the state of polarization of the light wave (8) in a molecular medium and the state of every molecule in the volume (10) only under very favorable conditions set by restrictions imposed on the absolute values of $\omega_1$ and $\omega_2$, frequency ratio $\omega_1/\omega_2 > < 1$, intensity ratio $(I_1/I_2) \ne 1$, and the angle $\gamma$ between vectors $\mathbf{E}_1$ and $\mathbf{E}_2$, as well as the energy flux vector of the pumping field $\mathbf{S}$. Physically, the synergetic generation of the process of interaction between molecules and the traveling light wave (8) is caused by the following.

If the deviation $\Omega^*_{\kappa,\kappa+1} \ne 0$, then the instantaneous carrier frequency $\omega(t) = \omega_s + \Omega(t)$ varying *symmetrically* about its mean value $\omega_s$ varies *asymmetrically* relative to time moments $t''_\kappa$ (with respect to the axis $X$). As a consequence, at every "plane" half-cycle $T_{s\kappa}/2$ of the time interval (9) an *uncompensated* deviation step $\delta\Omega^*_{\kappa,\kappa+1} \ne 0$ arises, as well as the asymmetry in the distribution of molecular perturbations in the plane $YZ$ with respect to the axis $X$.

The uncompensated step $\delta\Omega^*_{\kappa,\kappa+1} \ne 0$ of deviation $\Omega^*_{\kappa,\kappa+1} \ne 0$ of the carrier frequency $\omega(t)$ is an active component of the period $T_{s\kappa}$ of oscillations of $\mathbf{E}$- and $\mathbf{H}$-components of the RSBF. Because of the square Stark and Zeeman effects, it leads to the increase of the potential energy of the molecule due to the uncompensated electric $\delta W_{S,\kappa}$ and diamagnetic $\delta W_{Z,\kappa}$ energies at every half-cycle $T_{s\kappa}/2$.

It is clear that the step of energy increase in a molecule at every next period $T_{s\kappa+1}$ depends nonlinearly on the value of the energy increase at the previous period $T_{s\kappa}$, and the increase of the diamagnetic energy $\delta W_{Z,\kappa+1}$ depends on the increase in the electric energy $\delta W_{S,\kappa}$ (or vice versa at other frequency ratio $\omega_1/\omega_2$). As a consequence, the "plane" period $T_{s\kappa}$ transforms into the "volume" period $T'_{s\kappa}$, within which the precession and nutation motions of the bonding electrons twice exchange energy and MM with the orbital motion of the electrons and rotation of the molecule as a whole.

The "volume" period $T'_{s\kappa}$ plays the role of a prepared AP loop (memory) of RSBF oscillations that is similar to the hysteresis cycle of magnetization of paramagnetic materials.[11] A step $\delta\Omega^*_{\kappa,\kappa+1} \ne 0$ plays the role of an active component of the scale $s_\kappa$ of filling the space between the planes $YOZ$ and $YLZ$ along the axis $X$ over the cross section $2R_0$ by the light energy flux $\mathbf{S}$ within the time interval (9). The square of this scale is equal to

$$s_\kappa^2 = v_{\mathrm{ph}}^2 \, (t'_\kappa - t''_\kappa)^2 - R_\kappa^2.$$

where $t'_\kappa = t''_\kappa - R_\kappa/v_{\mathrm{ph}}$ is the lag of the induced waves;

$$R_\kappa^2 = (ax' - ax'')^2 + (y' - y'')^2 + (z' - z'')^2$$

is the square distance between the moment of the electron acceleration $t'_\kappa$ and the instant $t''_\kappa$, when the electron affects all other charges; $(t'_\kappa - t''_\kappa) \approx T'_{s\kappa}/4$.

Since the amplitude $A(r, t)$ includes the half-difference $\Omega^*_{\kappa,\kappa+1}/2 \approx \Delta\omega/2$, and the phase $\Phi_s(r, t)$ includes the half-sum $\omega_s$ plus the half-difference $\Delta\omega/2$ of the pumping field frequencies ($\omega^*_{s\kappa} \approx 2\omega_s/2 + \Omega^*_\kappa$), when the field frequencies $\Omega^*_{\kappa,\kappa+1}/2$ and $\omega^*_{s\kappa}$ fall in

resonance with the frequencies of molecular transitions by the $V$-scheme (3) at the moments $t'_\kappa$ and $t''_\kappa$, the correlation arises between variations of the amplitude $A(r, t)$ and the phase coefficient $\alpha = \alpha_y - \alpha_z$ of the RSBF (8) and the variations of the quantum energy (orientation of the orbital and rotational MM) of the molecule. As a consequence, the nonlinear relation arises between variations of the step $\delta\Omega^*_{\kappa,\kappa+1} \neq 0$ at neighboring AP loops $T'_{s\kappa}$ with the resonance shifting-splitting of the levels of the $V$-scheme (3) in the molecular phase space. Thus, the step $\delta\Omega^*_{\kappa,\kappa+1} \neq 0$ "unites" the space (10) and time (9) into a single set connected by linear-nonlinear interactions between the molecules of the medium and $E$- and $H$-components of the RSBF (8) and the field $(E'_y, H'_z)$ generated in molecules at low $\Omega^*_{\kappa,\kappa+1}/2$ and high $\omega^*_{s\kappa}$ frequencies.

Thus, the deviation $\Omega^*_{\kappa,\kappa+1} \neq 0$ of the carrier frequency $\omega(t)$ of the RSBF (8) may become a synergetic generator connecting the natural motion of subsystems of every molecule in the volume (10) with the precession and nutation motion of the electron orbits through the level $|0\rangle$ as a bifurcation point. The bifurcation process is constructed due to correlation of the transverse and longitudinal variations of the amplitude $A(r, t)$ and the phase coefficient $\alpha = \alpha_y - \alpha_z$ of $E$- and $H$-components of the RSBF (8) in the medium molecules (10). The deviation step $\delta\Omega^*_{\kappa,\kappa+1} \neq 0$ allows us to control intensification of the PN motion of the bonding electrons in molecules through nonlinear intensification of transverse (over the cross section $2R_0$) and longitudinal (along the axis $X$) spatial interactions of $E$- and $H$-components of the RSBF (8) with the molecules of the medium.

The step $\delta\Omega^*_{\kappa,\kappa+1} \neq 0$ depends on the inhomogeneity and absolute value of the RSBF along the $Y$ and $Z$ axes, i.e., not only on the selected absolute values of $\omega_1$ and $\omega_2$ and the frequency ratio $(\omega_1/\omega_2 > < 1)$, as well as the ratio $(I_1/I_2) \neq 1$, but also on the angle $\gamma \neq 0$ between the vectors $E_1$ and $E_2$ and the energy flux of the pumping field $S$.

## 4.2. Justification of the initial scale of AP loop of the RSBF oscillation

The initial scale $s_{\kappa=0}$ of the AP loop $T'_{s\kappa}$ must be commensurable with the "distance," at which the molecule having received the energy increments $\delta W_{S,\kappa}$ and $\delta W_{Z,\kappa}$ via the bonding electron and, respectively, the increments $\delta\alpha_\kappa$, $\delta\mu_\kappa$, $\delta J_{p\kappa}$ for the half-cycle $T'_{s\kappa}/2$ has time to redistribute them in its phase space and is ready to receive the following energy increment at the next half-cycle $T'_{s\kappa+1}/2$. It is clear that the commensurability of the sum of AP loops making up the duration of the RSBF wave packet

$$T'_{vp} \approx \Sigma\kappa T'_{s\kappa} = J(J + 1) T'_{s\kappa}/2, \qquad (11),$$

and the time $T \approx 1/B_e \approx 10^{-11}$ s determine the speed of preparation of AP loops in the molecular medium under the effect of $E$- and $H$-components of the RSBF (8). The rate of preparation of the AP loop $T'_{s\kappa} \approx 10^{-15}$ s determines the speed of *self-organization* of the ensemble of diamagnetic "molecule + field" systems in the volume (10), as well as the time of initiation of generation of the radiation with high MM at the *cooperative* frequency $\omega_p \approx \omega_{0J} \approx \omega^*_{s\kappa}$. Let us justify the scale $s_{\kappa=0}$ of the AP loop through "credible" reasoning using the language of semi classical mechanics based on two circumstances.

First, the electron polarizability $\alpha_{0J}(\omega)$ of diatomic molecules (most of diatomic molecules) is positive for the ground state and the optical frequencies $\omega$, whereas the diamagnetic susceptibility $\chi_{dnJ}(\omega)$ is negative. This is caused by the fact that diamagnetism arises due to the increase of the kinetic energy of the electron because of the increment of $v_\perp$ at the orbit (5), whereas the electron polarizability arises due to the shift of the electron density to the region of lower potential electric energy, i.e., to the molecular c.g. $(\delta r \approx r - r_e \approx 0)$. Here $2r_e$ is the internuclear distance corresponding to the equilibrium configuration of electrons in the ground rotational-vibrational-electronic state of an unperturbed diatomic molecule.

Second, the electron polarizability and diamagnetic susceptibility of molecules-rotators have the opposite dependence on $J$ at the optical frequencies: $\alpha_{0J}(\omega_1,\omega_2,\Delta\omega/2,\omega_s) \sim 1/J$ and $\chi_{dnJ}(\omega_1,\omega_2,\Delta\omega/2,\omega_s) \sim J$ (Ref. 9). Therefore, the variation of electron polarizability of molecules-rotators depends mostly on their resonance with the frequency $\sim\Delta\omega/2$ of the amplitude modulation of $E(y, t)$-component of the RSBF (8) and with the frequency $\Omega^*_{\kappa,\kappa+1}$ of the field $E'_y$ induced in molecule due to $dH(z, t)/dt$. And variation of the diamagnetic susceptibility of molecules-rotators depends mostly on their resonance with the frequency $\sim\Delta\omega/2$ of the amplitude modulation of $H(z, t)$-component of the RSBF (8) and with the frequency $\omega^*_{s\kappa}$ of the field $H'_z$ induced in the molecule due to $dE(y, t)/dt$.

As a consequence, the spectral line corresponding to the transition $|1\rangle \leftrightarrow |0\rangle$ shifts-splits under the effect of $E(y, t)$-component of the field (8a) mostly in phase with the effect of $E'_y$ component at the frequency $\Omega^*_{\kappa,\kappa+1} \approx \Delta\omega$ on the $\pi$- and $\sigma_\pm$-components arranged symmetrically (in the first approximation) at the distances $\sim E_0$ relative to the spectral line. In this case, the levels of the transition $|1\rangle \leftrightarrow |0\rangle$ shifts-splits by $\delta\omega_S \sim (e^2/2m_e^2\,\omega_{0J}^2)\,E_0^2$ because of the square Stark effect arising due to anharmonicity of electron rotation in molecule. The levels shift to the region of the minimum potential electric energy, thus increasing the electric field strength in a small volume $\delta V' \approx \delta r\,\pi(r_e/2)^2$ near the molecular c.g.

As the electron polarizability of a molecule (unperturbed) at the level $|0\rangle$ achieves its maximum[7]

$$\alpha''(\omega) = (\alpha_\parallel + 2\alpha_\perp)/3, \qquad (12)$$

the shift of just this level determines the mean increase $\delta\alpha_\kappa$ (because of the uncompensated shift of the levels $\sim E_0^2$) for every half-cycle $T'_{s\kappa}/2$. Here $\alpha_\parallel$ and $\alpha_\perp$ are the longitudinal and transverse components (relative to the molecular axis $R'$) of the tensor of electron polarizability $\alpha(\omega)$ at the optical frequencies $\omega$ calculated at $\delta r \approx (r - r_e) \approx 0$ by summing up the dipole moments over all intermediate RV states of all ($n$) electron terms, except for the ground one;

$$\alpha_{\parallel,\perp} = (2/\hbar) \sum |d_{0\Lambda}^{\parallel,\perp}|^2 \, \omega_{\Lambda 0}/(\omega^2 - \omega_{\Lambda 0}^2),$$

and $d_{0\Lambda}^\parallel$ and $d_{0\Lambda}^\perp$ are the projections of the dipole moment of a molecule onto its interatomic axis $R'$ and onto one of the directions normal to the axis $R'$; $\omega_{\Lambda 0}$ and $\omega$ are the frequencies of the electronic transition of a molecule and the perturbing optical field.

The refractive index $n$ of the medium near the transition $|1\rangle \leftrightarrow |0\rangle$ depends on the coincidence of the frequency of amplitude modulation $\Delta\omega/2$ of $\mathbf{E}(y, t)$-component of the RSBF (8a) with the frequency of this transition $\omega_{01}$ at certain moments in time $t_\kappa$; therefore, under the effect of the field difference $(E_0 - E'_y) \approx E_0$ the value of $n$ also varies (dispersion curve). The dispersion curve at the transition $|1\rangle \leftrightarrow |0\rangle$ sharpens or smoothes for the linearly polarized wave (8) depending on the amplitude ratio $(E_1/E_2)^2 \gtrless 1$ and the frequency ratio $(\omega_1/\omega_2 \gtrless 1)$ of its spectral components.

Besides, the spectral line corresponding to the transition $|0\rangle \leftrightarrow |J, M_J\rangle$ shifts-splits under the effect of $\mathbf{H}(z, t)$-component of the field (8b) mostly in phase with the effect of $\mathbf{H}'_z$-component at the frequency $\omega^*_{s\kappa}$ on the $\sigma_\pm$-components of each of $M_J$-sublevels. These sublevels are arranged symmetrically (in the first approximation) at the distances $\delta\omega_Z \sim \pm eH_0/2m_e$ (Larmor frequency) about the line corresponding to the transition $|0\rangle \leftrightarrow |J, M_J\rangle$. As a consequence, the natural molecular frequencies corresponding to the right and left electron orbital rotation shift in different directions under the effect of the magnetic field $H_0$. Thus, the relation establishes between the Zeeman and Faraday effects, i.e., the electron motion shifts in the direction of the $X$ axis of propagation of $\mathbf{H}(z, t)$-component of the field (8b), and its polarization plane turns (Faraday effect) on the average by the angle

$$\psi = (\varphi_+ - \varphi_-)/2 = \omega_s \, \Delta L (n_- - n_+)/2c =$$
$$= \pi\Delta L \, (n_- - n_+)/\lambda_{0s}. \qquad (13)$$

The effect of rotation of the polarization plane of the pump wave depends on the ratio $d/\lambda \sim 10^{-3}$ ($kr \ll 1$). Here $\varphi_+ = \omega(t - x/v_+)$ and $\varphi_- = \omega(t - x/v_-)$, $v_+ = c/n_+$ and $v_- = c/n_-$, $n_+$ and $n_-$ are the penetration depth $x = L$, phase velocities, and refractive indices for the right-hand and left-hand circularly polarized waves; $\omega/c = 2\pi/T$; $d = 2r$ is the molecule size; $k = 2\pi/\lambda_{0s}$ is the wave number, $\lambda_{0s}$ is the wavelength in vacuum.

Since the refractive index $n$ depends on the degree of coincidence between the carrier frequency $\omega^*_{s\kappa}$ of $\mathbf{H}(z, t)$-component of the field (8b) and the frequency $\omega_{0J}$ of the transition $|0\rangle \leftrightarrow |J, M_J\rangle$, the value of $n$ (dispersion curve) also varies under the effect of the field difference $(\mathbf{H}_0 - \mathbf{H}'_z)$ The dispersion curve at the transition $|0\rangle \leftrightarrow |J, M_J\rangle$ sharpens or smoothens for $\mathbf{H}(z, t)$-component of the wave (8b) depending on the amplitude ratio $(H_1/H_2)^2 \gtrless 1$ and the frequency ratio $(\omega_1/\omega_2 \gtrless 1)$ of its spectral components.

The square Zeeman effect (diamagnetic effect) arising as a consequence of the Larmor *precession* of the electron magnetic moment $\delta\mu_\kappa$ induced by the perturbing magnetic field $(\mathbf{H}_0 - \mathbf{H}'_z) \approx \mathbf{H}_0$ is proportional to the mean area occupied by the electron density in the plane $ZY$. The energy of shifting-splitting of the level $|J, M_J\rangle$ is equal (in the first approximation) to the mean value of the total diamagnetic energy of all electrons of a molecule calculated for the $\tilde{n}$th electron orbit:

$$\delta W_{\tilde{n}JMJ} = \chi_{d\tilde{n}J}(\omega) \, H_0^2.$$

The diamagnetic susceptibility of molecules $\chi_{dnJ}(\omega)$ increases with the increase of $J$ and $n$ (Refs. 9 and 10) or, in other words, as the orbit radius of the bonding electrons $r_e$ increases ($\tilde{n} = 1, 2, 3, \ldots$).

From the Fresnel equation (13) it follows[10,11] that rotation of the polarization plane of the pump wave allows us to create a detectable difference of $10^{-6}$ between $n_+$ and $n_-$ of the molecular medium at the distance $\Delta L$. In our case, the difference $\Delta n_\pm \approx 10^{-6}$ must be generated by the wave (8) at the distance $\Delta L$ for the time equal to the difference of the periods of revolution of molecules-rotators between the neighboring pairs of rotational levels:

$$T' \approx T_{J,J-1} - T_{J+1,J} = 2\pi/\omega_{01} =$$
$$= 1/(2B_e) \approx 10^{-11} \text{ s.} \qquad (14)$$

Then the difference $\Delta n_\pm \approx 10^{-6}$ can be read-out (selected) and the uncompensated increase $\delta W_{Z,\kappa}$ ($\delta\mu_\kappa$ and $\delta\mathbf{J}_{p\kappa}$) can be generated by the same wave (8) for the following $T' \approx 1/(2B_e)$ s.

It is clear that the scale $s_{\kappa=0}$ of the AP loop of RSBF oscillations, at which generation of the increments $\delta W_{Z,\kappa}$ ($\delta\mu_\kappa$ and $\delta\mathbf{J}_{p\kappa}$) and $\delta W_{S,\kappa}$ ($\delta\alpha_\kappa$) begins in molecule, must compare with the internuclear distance in the molecule $d \approx s_{\kappa=0}$. Therefore, the smaller is $T'_{s\kappa}/2$, the higher is the accuracy of the initial scale of the AP loop, within which the molecules can take the acceptable part of the energy flux of the pumping field $\mathbf{S}_{opt}$ to provide for these increments in the energetically favorable directions of their phase space.

### 4.3. The lowest state $|0\rangle$ as a point of bifurcation of natural rotation of bonding electrons in molecule on the mode of their stable PN motion

The rotational molecular state $|0\rangle$ may become a bifurcation point of intramolecular motions for the following reasons.

1. The molecular MM at the level $|0\rangle$ is much less than the photon quantum of action $J = 0 \ll \hbar = \ell' + s'$ and oriented arbitrarily about the immovable coordinate system. Therefore, the molecular rotation dynamics at the level $|0\rangle$ must include both classic (nonlinear) and quantum (linear) elements. Since the molecule-rotator is a nonlinear system in the classic approximation, small variations of the external perturbations of the orbital electron motion in a molecule as a classic rotator being at the level $|0\rangle$ may lead to drastic changes in its state as a quantum system; $\ell'$ and $s'$ are the orbital (spatial) and the spin photon moments.

2. The state $|0\rangle$ is a reference point in the molecular phase space (in the rigid rotator approximation) of the difference between molecular periods of revolution between the neighboring pairs of rotational levels $T'$ (14).

The value of $T'$ is constant for the unperturbed molecule and has the meaning of the order parameter for intramolecular interactions, including interactions of the electron motion with rotation of the molecule as a whole. The energy of centrifugal stretching $\Delta W_{J=1} = -4D$ ($D \approx 10^{-4} B_e$) bounds the molecular order parameter from below for the synergetic generator.

3. The position of the level $|0\rangle$ on the potential curve can be specified using an infinitely small volume $\delta V' \approx \delta r \, \pi (r_e/2)^2$ near the c.g. of the unperturbed molecule, in which the minimum potential energy of rotation of the molecule as a whole is concentrated. The volume $\delta V'$ plays a principal part in the transformation of the relation between the orbital (spatial) and spin electron subsystems, the structure of rotational levels, and the probability of transitions between them for three reasons.

*First*, the molecular volume $\delta V'$ meeting the condition $kr \ll 1$ is the initial scale of longitudinal (along the $X$ axis) contributions of the perturbing pump field to MM of the incipient spherical wave about the $X'$ symmetry axis (Ref. 16).

*Second*, the molecular volume $\delta V'$ characterizes the initial scale $s_{\kappa=0}$ of its transverse (along the radius $R \leq R_0 \approx \sqrt{v_{ph} \, 2\pi \Delta L / \omega_s}$) interactions with **E**- and **H**-components of the RSBF within the duration of the wave packet (11).

*Third*, the local instability of molecular rotation at the level $|0\rangle$ may give rise (and intensify) interactions of the orbital and spin electron subsystems and the orbital $\ell'$ and spin $s'$ photon subsystems of the perturbing field (8) in the volume $\delta V'$ at the frequencies $\Omega^*_{\kappa,\kappa+1}/2 \approx \Delta\omega/2$ and $\omega^*_{s\kappa} \approx 2\omega_s/2 + \Omega^*_\kappa$.

In other words, the perturbing field (8) may provoke local instability of molecular rotation at the frequency $\Omega^*_{\kappa,\kappa+1}/2$, resonance frequency $\omega_{01}$ of oscillation of the positions of the levels $|1\rangle \rightarrow |0\rangle$, as well as the nonlinear relation between the molecular rotation and the rotation of electrons at the frequency $\omega^*_{s\kappa}$ and the resonance frequency $\omega_{0J}$ of oscillation of the position of the levels $|0\rangle \rightarrow |J, M_J\rangle$. As a consequence, the resonance interaction between the electron and photon subsystems arises in the molecule. During the wave packet (11), this interaction generates:

– potential (field) for motion of nuclei with deceleration and neutralization of the energy of centrifugal stretching $\Delta W_J$;

– autophasing of PN motion of the bonding electrons with the accumulation of MM, uncompensated electric energy $\delta W_{S,\kappa}$, and diamagnetic energy $\delta W_{Z,\kappa}$ at the neighboring $\kappa$th and ($\kappa$+1)th time intervals up to critical values.

### 4.4. Autophasing of PN motion of bonding electrons of diamagnetic molecules-rotators in the cross $E(y, t)$ and $H(z, t)$ fields as a tool for self-organization of the EMO mechanism

1. The principle of autophasing was proposed by Veksler in 1944 (Ref. 10) as a means for an infinitely long preservation of the resonance between the motion of charged relativistic particles (electrons, ions) inside cyclotron accelerators by changing the strength of high-frequency fields: accelerating electric field $\mathbf{E}(t)$ and controlling magnetic field $\mathbf{H}(t)$. The phases of particle motion in the accelerator are captured by the *equilibrium phase* $+\varphi_0$ of the accelerating electric field $E_0 \cos\varphi_0$ and the controlling magnetic field; $E_0$ is the amplitude of the accelerating field strength.

This principle employs the dependence of the time interval $\partial t$ between two neighboring events of acceleration on the energy of particles $W$. Consider the situation that $\partial t/\partial W > 0$, i.e., $\partial t$ increases with the increasing kinetic energy $W$ (velocity $v$) of a particle. Let $\varphi_0$ be the phase of the field in the accelerating gap ("equilibrium phase"), the particle 0, if in this gap, moves exactly in resonance with the accelerating field and picks up the specified speed $v + \Delta v$ and the energy $W_0 = eE_0 \cos\varphi_0$. If the particle 2 moves quickly, i.e., falls in the leading phase

$$\varphi_2 > \varphi_0 > \varphi_1 > 0,$$

then it acquires the energy $eV_0 \cos\varphi_2$ less than the equilibrium one and arrives earlier ($\partial t$ "decreases") in the next accelerating gap, and the phase of arrival of this particle approaches the equilibrium phase $\varphi_0$. On the contrary, the lagged particle 1 acquires excess energy, $\partial t$ increases, the particle arrives later in the

accelerating gap and its phase approaches the equilibrium phase too. Thus, all particles being in some area near the phase $\varphi_0$ (phase capture area) oscillate about the phase $\varphi_0$ and accelerate. In this case, the total energy $W$ of "captured" particles takes the value close to the resonance one

$$W_{\text{res}} = c\,e\,<B>\,q/\omega_y,$$

if the frequency of the accelerating field $\omega_y$ changes rather slowly in time at the mean magnetic induction $<B>$; $c$, $e$ and $q = 1, 2, ..., N$ are the speed of light, charge of a particle, and an integer number (frequency multiplicity) showing how many times the frequency $\omega_y$ exceeds the frequency of particle circulation $\Omega_p$ in the resonance accelerator.

2. Oraevskii et al.[17] found an optical analog of the Veksler autophasing and proposed it for coherent excitation of high vibrational levels of anharmonic molecules-oscillators and for preparation of the dynamic (overexcited) vibrational state through neutralization of the effect of anharmonism $\delta\omega_{\text{anh}}$. The idea of the proposed model is that oscillations of an anharmonic oscillator are subject to perturbation from the electric field, whose strength is modulated by the phase $\Omega'' = d\phi(t)/dt \approx \delta\omega_{\text{anh}}$ with the amplitude $\Omega' = dA(t)/dt$, and the carrier frequency is in resonance with the transition between the lower pair of levels 1 and 2 ($\omega \approx \omega_{21}$).

The Veksler principle for the molecule-oscillator is essentially based on the coincidence of the difference between the neighboring periods of oscillation of its nuclei with the period of their anharmonic shift:

$$T_{q,q-1} - T_{q+1,q} = \Delta T_{\text{anh}} = 2\pi/\delta\omega_{\text{anh}},$$

and $T_{q+1,q}$, according to Ref. 17 means the time of the $q$th cycle of rotation of a charged relativistic particle about the magnetic field direction. Thus, this relation physically means that the time difference of two sequential cycles is a constant value independent of the particle energy, i.e., of $q$. It becomes possible to *"turn on"* one of the coherent mechanisms of interaction between the field and the medium, which manifests itself in the pulsation character acquired by the variation of the probability of level population.

If there is no relaxation of the levels 1 and 2, then their population varies periodically with the Rabi frequency $\Omega_{21} = E_0 d_{21}/\hbar$. At the moments $t = \tau_1$ such that $\Omega_{21}\tau_1 = (2\kappa + 1)\pi$ ($\kappa = 0, 1, ...$), the system is completely inverted – all molecules are at the upper level 2. If at the moment $\tau_1$ the radiation frequency varied stepwise and became equal to the transition frequency $\omega_{32}$ of the upper-lying pair of levels, then the molecules "captured" by the pulse start to oscillate again, but already between the levels 3 and 2 with the Rabi frequency $\Omega_{32} = E_0 d_{32}/\hbar$. Selecting the phase $\Phi(t) = \omega t + \phi(t)$ of turning-on of the next pulse according to the condition $\Omega_{32}\tau_2 = \pi(2\kappa + 1)$, we can transfer molecules to higher and higher energy levels.

Thus, a set of phased-in pulses with the frequencies $\omega_{21}$, $\omega_{32}$, $\omega_{43}$, ... or periodic change of the phase $\phi(t)$ of the field strength with the frequency of anharmonism of the molecule-oscillator allows, in principle, the cascade mechanism of populating high vibrational levels to be realized. The effect of anharmonism of the molecule-oscillator is neutralized due to regular tuning of the pump field frequency through the Rabi frequency to the frequency of neighboring $q$th and $(q + 1)$th vibrational transitions.

If the pump field strength has, in addition to the frequency modulation, amplitude modulation with the frequency $\nu$

$$E(t) = 1/2\,E_0\,g(t)\,\{\exp i\,[\omega t + \phi(t)] + \text{c.c.}\} \approx$$

$$\approx 1/2\,E_0\,g(t)\,[\Sigma C_n \exp i\,(\omega + \tilde{n}\delta\omega_{\text{anh}})\,t + \text{c.c.}],$$

then the modulation of the Rabi frequencies corresponding to different vibrational transitions of the anharmonic oscillator is complemented with the modulation of the "friction coefficient" with the frequency $\nu$; $g(t) = (1 + m\cos\nu t)$ is the function modulating the amplitude; $\Omega_0 = E_0 C_0 d_{q+1,q}/\hbar$ is the Rabi frequency (in the first approximation) at the transition between the $q$th and $(q + 1)$th vibrational levels.

In this case, the coherent mechanism of interaction between the field and the medium causes undamped oscillations of the diagonal elements of the density matrix $\rho_{mm}$ for the time $t$ exceeding the period of system relaxation. The pulsation spectrum consists of combination frequencies $\nu_{q\tilde{n}} = \nu_q \pm \tilde{n}\nu$, and the pulsation frequencies (Rabi frequencies) are multiple of the modulation frequency $\nu$. The maximum stock of vibrational quanta $\varepsilon$ per oscillator (averaging for the period of field pulsations) depends on the parameter $\Omega_0/\nu > 1$. If the depth $m$ and the frequency ($\tilde{n}$ and $q$) of the modulation are chosen such that $\nu_q = \tilde{n}\nu$, then it becomes possible to excite parametrically a quantum oscillator so that it finds itself in an anomalous (dynamic overexcited) equilibrium state. In this case, the component for the probabilities $\rho_{mm}$ increases and the mean number of vibrational quanta $\varepsilon$ per quantum oscillator exceeds $N/2$.

3. To make clear the idea of self-organization of the EMO mechanism and, consequently, the EMO effect, let us complement manifestation of the Lorentz force $\mathbf{F}$ (Section 3) with the version, when the free electron is subject to the effect of slowly varying cross fields $d\mathbf{E}(y, t)/dt \neq 0$ and $d\mathbf{H}(z, t)/dt \neq 0$ (Ref. 12). In this case, the electron moving along the $X$ axis in the direction of linearly polarized field $\mathbf{E}(y, t)$ accelerates, and the magnetic field bends its trajectory more weakly according to the Lorentz force $\mathbf{F}$. When the vector $\mathbf{E}(y, t)$ changes the direction, the electron moving against the field $\mathbf{E}(y, t)$ decelerates and its trajectory is more and more bent by the magnetic field $\mathbf{H}(z, t)$ along a cycloid. As a result, we have the electron "drift" in the direction ($\mathbf{E} \times \mathbf{H}$). This electron

drift is a superposition of the uniform motion with the velocity $v_p = cE_\perp/H$ and the circular one. Let us imagine an observer moving along the $X$ axis with a constant speed $v_p$. In the observer's reference system, the field $\mathbf{H}$ transforms into a new field $\mathbf{H}'$ plus the electric field $\mathbf{E}'$ directed downward. If the observer velocity $v_p$ is selected such that the total electric field is zero $(\mathbf{E}(y, t) - \mathbf{E}' = 0)$, then the observer sees the electron moving along a circle, i.e., the observed motion is circular plus transport in the direction $X$ with the drift velocity $v_p = cE_\perp/H$.

We do not know any research on electron motion in the potential (field) of a molecule under the effect of slowly (quickly) varying cross fields $d\mathbf{E}(y, t)/dt \neq 0$ and $d\mathbf{H}(z, t)/dt \neq 0$. Below, based on the physical reasons, we propose the idea of self-organization of the EMO mechanism in a set of diamagnetic molecules under the effect of quickly varying cross fields $d\mathbf{E}(y, t)/dt \neq 0$ (8a) and $d\mathbf{H}(z, t)/dt \neq 0$ (8b) produced from the fields at the optical frequencies $\omega_1$ and $\omega_2$.

The idea of self-organization of the EMO mechanism in molecules is as follows. If the rotation of molecular electrons is subject to perturbation from $\mathbf{E}$- and $\mathbf{H}$-components of the RSBF (8a), which is amplitude-modulated at the frequency $\Omega^*_{\kappa,\kappa+1}/2 = \omega_{10} \approx$ $\approx \Delta\omega/2$ resonance to the transition $|1\rangle \leftrightarrow |0\rangle$ (instants $t'_\kappa$) and the carrier frequency $\omega^*_{s\kappa} \approx (2\omega_s/2) + \Omega^*_\kappa$ is in resonance with the transition $|0\rangle \leftrightarrow |J, M_J\rangle$ (moments $t''_\kappa$) at the optimal spatial-energy configuration of the RSBF (see Section 4.1), then the Lorentz force $\mathbf{F}$ acting, according to the low-high-frequency rotational push-pull principle must arise in the diamagnetic molecules.

The low-high-frequency rotational push-pull principle is autophasing of the PN motion of the bonding electrons in a molecule during the time interval (9). Autophasing determines the potential (field) for motion of nuclei with deceleration and neutralization of the energy of centrifugal stretching $\Delta W_J = -D [J (J + 1)]^2$ for the time $T \approx 1/B_e$ s. As a consequence, in diamagnetic molecules from the volume (10), the transfer of the interaction energy by the $V$-channel (4) self-organizes along with the accumulation of MM by molecules $\boldsymbol{J}_p = m_e v_\perp r \approx m_e \Omega_L r_L^2 \to m_e \Omega_L R^{*2} \approx$ $\approx \sum_\kappa \delta J_{p\kappa} \approx K_p(t''_{\kappa=J}) \approx \hbar 2(J + 1)$ and with the formation of the ensemble of diamagnetic "molecule + field" systems on the average for the time $T \approx 1/B_e$ s. Here $R^* \approx R_0$ is the effective radius of PN motion of the bonding electrons. The radius of the Larmor orbit $r \approx r_L \to R^*$ increases because of the quantum component of the low-high-frequency perturbation of the orbital electron motion by the pump field (8) if the approximate equation (17) is valid (see below).

4. Let us justify the conditions of autophasing and growth of the effective radius of the PN motion of the bonding electrons in a molecule as follows. Consider the coherency volume (10) of the medium consisting of

diamagnetic molecules and the causes for appearance of autophasing of the PN motion of the bonding electrons as a tool for transfer of the interaction energy through channel (4), self-organization of the EMO mechanism and the ensemble of the diamagnetic "molecule + field" systems.

The volume (10) of a medium consists of closed contours $\Gamma$ of the radius $R \leq R_0$ in the plane $YZ$ with the center on the $X$ axis. The AP loops of RSBF oscillations having the duration $T'_{s\kappa}$ join the contours. In molecules from the volume (10), in which the electron orbits have the radii $r$, the tangential electric field

$$\mathbf{E}'_y \, 2\pi \, r_0 \approx - \, d(\mathbf{H}(z, t) \, \pi \, r_0^2)/dt$$

is generated due to magnetic induction from $d\mathbf{H}(z,t)/dt$. The strength of this field is

$$\mathbf{E}'_y \approx - \, r_0/2 \, d\mathbf{H}(z, t)/dt.$$

The induced electric field $\mathbf{E}'_y$ circulating in molecules consists of the low-frequency $\Delta\omega/2$ $(\Omega^*_{\kappa,\kappa+1}/2)$ and high-frequency $\omega_s$ $(\omega^*_{s\kappa})$ parts [similar parts are present in the circulating field $\mathbf{H}'_z$ induced in molecules due to $d\mathbf{E}(y, t)/dt)$].

The low-frequency induced electric field $\mathbf{E}'_y$ acting on the bonding electrons according to Eq. (5) produces the moment of force $- e\mathbf{E}'_y \, r_0$, whose value is equal to the rate of change of the molecular MM

$$d\mathbf{J}_p/dt \approx (er^2/2) \, d\mathbf{H}(z, t)/dt. \qquad (15)$$

If we integrate Eq. (15) starting from zero field, then for the half-cycle of the amplitude modulation of the field strength (8b) $T_{am}/2 \approx 2\pi/\Delta\omega \approx 10^{-11}$ s the molecule must receive the extra MM $\Delta\mathbf{J}_p$ and, consequently, the extra diamagnetic moment

$$\Delta\mu \approx -\Delta\mathbf{J}_p \, e/2m_e \approx (-e^2 \, r^2/4m_e) \, H_0. \qquad (16)$$

It is clear that $\Delta\mu$ is very small because of the small mean strength $H_0$, absence of resonance energy (MM) exchange between molecules and the pump field, in particular, at the frequencies $\Omega^*_{\kappa,\kappa+1} \approx \Delta\omega$ (for $\mathbf{E}'_y$) and $\omega^*_{s\kappa} \approx (2\omega_s/2) + \Omega^*_\kappa$ (for $\mathbf{H}'_z$), as well as because of the absence of PF between the state of the molecule and the contributions of $\mathbf{E}$- and $\mathbf{H}$-components of the field (8) to the PN motion of the bonding electrons at the neighboring $\kappa$th and $(\kappa + 1)$th time intervals.

The situation can be changed drastically, if making use of the fact that the energy of the induced electric current and magnetic flux does not dissipate in the molecule being in the stationary state, for example, $|J = 1\rangle$ (Refs. 9 and 12). This allows us to produce:

– resonance energy (MM) exchange between the molecules and the field (8) at low frequency $\Omega^*_{\kappa,\kappa+1}/2 =$ $= \omega_{01} \approx \Delta\omega/2$ (through the field $\mathbf{E}'_y$) at time $t'_\kappa$ with the participation of precession of the bonding electrons

on the orbit at the Larmor frequency $\Omega_L$, as well as at high frequency $\omega_{sk}^* \approx \omega_{0J}$ (through the field $\mathbf{H}_z'$) at the moments $t_\kappa''$ with the participation of nutation of the electron orbit about the vector $\mathbf{K}(t_\kappa)$ at the cooperative frequency of the signal (generation) $\omega_p$;

– PF between the molecular states and the contributions of $\mathbf{E}$- and $\mathbf{H}$-components of the field (8) to the PN motion of electrons along the orbit within the neighboring $\kappa$th and $(\kappa + 1)$th time intervals.

To produce the resonance energy (MM) exchange between molecules and the pump field at frequencies $\Omega_{\kappa,\kappa+1}^* / 2 = \omega_{01} \approx \Delta\omega / 2$ $(\mathbf{E}_y')$, $\omega_{sk}^* \approx \omega_{0J}$ $(\mathbf{H}_z')$, it is worth using the classic and quantum components of perturbation of the electron orbital motion by the pump field. Toward this end, let us find the density of photons $N_f$ (energy flux $\mathbf{S}_{opt}$) of the pump field and density of molecules $N_r$ in the volume (10) $(N_f \le N_r)$ such that the *cooperative* (collective, vacuum) Rabi frequency[18]

$$\Omega_c = (2\pi N_r \,| \, d_{0\Lambda}^{\|,\perp} |^2 \, \omega_{0J} / \hbar V)^{1/2},$$

the Larmor frequency $\Omega_L$, and the frequency of the transition $|1\rangle \leftrightarrow |0\rangle$ are related as follows

$$\omega_{10} \approx \sqrt{(\Omega_L^2 + \Omega_c^2)}. \tag{17}$$

The approximate equation (17) accounts for the classic (through $\Omega_L$) and quantum (through $\Omega_c$) components of the interaction between the molecules and pump field, and the former mostly affects the Q-factor of the transition $|1\rangle \leftrightarrow |0\rangle$, whereas the latter one affects the frequency of the energy (MM) exchange between the molecules and the field at the transition $|0\rangle \leftrightarrow |J, M_J\rangle$.

Since the cooperative Rabi frequency $\Omega_c$ characterizes the frequency of energy (MM) exchange between the molecules and the field by the $V$-scheme (3) in the absence of an external field $(\mathbf{S} = 0)$ (Refs. 18 and 19), the condition (17) means the parity dependence of the probability of molecular transition by the $V$-channel (4) on the density of molecules $N_r$ and on the density of photons $N_f$ in the volume (10). In this case, the cyclotron frequency $\Omega_L$ reflects the extra rotation of the bonding electrons along the Larmor spiral, and the cooperative Rabi frequency represents the process of stretching of the Larmor spiral $r \approx r_L \to R^* \approx R_0$, i.e., the process of growth of the effective radius of the PN motion of the bonding electrons $r \approx r_L \to R^* \approx R_0$ due to the quantum component of the energy flux $\mathbf{S}_{opt}$.

To make the PF between the molecular state and the contributions of $\mathbf{E}$- and $\mathbf{H}$-components of the field (8) to the PN motion of electrons in the neighboring $\kappa$th and $(\kappa+1)$th periods $T_{sk}'$, we use the adaptation of the distribution of the pump field (8) and the intramolecular field in space and time through the uncompensated step $\delta\Omega_{\kappa,\kappa+1}^* \ne 0$ of the deviation $\Omega_{\kappa,\kappa+1}^* \ne 0$ of the carrier frequency $\omega(t)$. Toward this end, let us relate the variations of $\mathbf{E}$- and $\mathbf{H}$-

components of the RSBF (8) at the neighboring moments in time $t_\kappa'$ and $t_{\kappa+1}'$ with the Larmor $\Omega_L$ and cooperative $\Omega_c$ frequencies of motions of the electron orbit by Eq. (17) and those at the neighboring moments $t_\kappa''$ and $t_{\kappa,\kappa+1}''$ with the period of nutation motion $T_N$ by Eq. (7). Equation (7) provides for generation of the moment of inertia $I_z(t)$, as well as the value and orientation of the artificial MM $\mathbf{K}(t_{\kappa=0}'') \approx$ $\approx \delta\mathbf{J}_{p\kappa=0} \to \mathbf{K}_p$ $(t_{\kappa=J}'')$ of the diamagnetic "molecule + filed" systems about the $X'$ axis during the wave packet (11).

Adaptation of the field distributions is efficient, if the intensity ratio is $(I_1/I_2) \approx 5.8$ $(\varphi \ne 0)$, and the angle $\gamma \ne 0$ between the vectors $\mathbf{E}_1$ and $\mathbf{E}_2$ makes inhomogeneity of the field (8) over the cross section $R_0$ to be such that variations of the amplitude $A(r, t)$ and, correspondingly, the position of the levels of adjacent transitions (3) in the cross section $R_0$ for every $T_{sk}'/2$, show maximum correlation with the variations of the phase coefficient $\alpha = \alpha_y - \alpha_z$ of the RSBF. Then, for every half-cycle $T_{sk}'/2$, molecules can receive optimal portions of the uncompensated energy, MM, polarizability, and susceptibility, redistributing them into the energy efficient directions of their phase space.

At $(I_1/I_2) \approx 5.8$ the deviation is equal to doubled main frequency of molecular rotation $\Omega_{\kappa,\kappa+1}^* \approx 4B_e =$ $= 2\omega_{10}$; therefore, each of the vectors $\pm \mathbf{E}(y, t)$ and $\pm \mathbf{H}(z, t)$ (8), alternating its direction twice during every period of the AP loop $T_{sk}'$, accelerates twice and controls the motion of the bonding electrons, and the following molecular parameters can increase by the optimal values: energy $(2\delta W_{S,\kappa}$ and $2\delta W_{Z,\kappa})$, electro-magneto-optical parameters $(2\delta\alpha_\kappa$ and $2\delta\mu_\kappa)$, and MM $2\delta\mathbf{J}_{p\kappa}$. The frequency ratio $(\omega_1/\omega_2) > < 1$ determines the direction of accelerations and controls of the electron motion (direction of PF between the molecular state and the increment) and, correspondingly, the direction of transfer of the interaction energy through (back) the channel (4) at every next period $T_{sk}'$.

If at $(I_1/I_2) \approx 5.8$ we take the frequency ratio $(\omega_1/\omega_2) > 1$ and $\gamma \ne 0$, then the PF arises in the medium molecules between their states at the next period $T_{sk}'$ and the increments at the previous period $T_{sk-1}$ (analog of the Veksler autophasing effect[10] for the molecule-rotator as a nonlinear system). The interaction energy $(2\delta W_{Z,\kappa-1}$ and $2\delta W_{S,\kappa-1})$ resonantly increases and it is transferred from the field to molecules as a result of the action of the PN electron motion on the state of molecules (analog of the Rabi-resonance intensification of Larmor precession of electrons in atoms[10]) and the molecules undergo the transition $|1\rangle \to |0\rangle$, regularly tuning the value and the direction of the shifting-splitting of the levels (3) by the $V$-channel (4) to resonance with the frequencies $\Omega_{\kappa,\kappa+1}^*$ and $\omega_{sk}^*$.

Thus, first, every diamagnetic molecule from the coherency volume (10) starts from the level $|1\rangle$ at its

own moment $t''_{\kappa=0}$ of its own $\Gamma$ contour and falls in its own AP loop of oscillations of the RSBF (8) with its own uncompensated step $\delta\Omega^*_{\kappa,\kappa+1} \neq 0$ of deviation $\Omega^*_{\kappa,\kappa+1} \approx 4B_e = 2\omega_{10}$ of the carrier frequency $\omega(t)$. The set of molecules acquires the property of a EMO trap (volume cavity), in which the rotational transitions (3) of every molecule have high Q-factor and are connected by the double parametric resonance with the pumping field (8) through the secondary waves at the initial frequencies $\omega_1$, $\omega_2$, and the generated fields at the frequencies $\Omega^*_{\kappa,\kappa+1}$ ($\mathbf{E}'_y$), $\omega^*_{s\kappa}$ ($\mathbf{H}'_z$).

Every molecule from the EMO trap proves to be involved in the "game" of cross (in the plane $YZ$) resonant (frequencies $\Omega^*_{\kappa,\kappa+1}$ and $\omega^*_{s\kappa}$) and nonlinear interactions with $\mathbf{E}$- and $\mathbf{H}$-components of the RSBF (8). This game leads to the transfer of the energy of shifting-splitting of molecular energy levels by the $V$-channel (4) about the axis $X'$.

Second, the process of cross interactions of $\mathbf{E}$- and $\mathbf{H}$-components of the field (8) with molecules prevails over other processes developing in the direction of the $X$ axis (rotational SRS and energy exchange between the fluxes $E_1$ and $E_2$ at oscillations of the Stark shift), if the frequency detuning of the fields exceeds the width of nonradiative transition $|1\rangle \leftrightarrow |0\rangle$ ($|\Delta\omega| \geq \gamma_{01}$, $\gamma_{01}$ depends on the density of molecules $N_r$), and the density of the energy flux $\mathbf{S}_{opt}$ of the RSBF provides for the optimal excess of the strength $E_0$ ($H_0$) over the threshold value $E_0 \geq E_{th}$ ($H_0 \geq H_{th}$).

# 5. Conditions for origin and development of EMO mechanism

Let the pumping field be represented in the form of the RSBF (8) with the amplitude $A(0, t)$ modulated at the frequency

$$\Omega^*_{\kappa,\kappa+1}/2 = \omega_{10} \approx \Delta\omega/2 \approx 10^{11} \text{ Hz} \qquad (18a)$$

resonant to the rotational transition $|1\rangle \leftrightarrow |0\rangle$ and with the phase

$$\Phi'_s(x = 0, t) = \omega_s t + (\alpha_y - \alpha_z) + \phi(x = 0, t)$$

corresponding to the instantaneous carrier frequency $\omega(t) = \omega_s + \Omega(t)$ resonant to the rotational transition $|0\rangle \leftrightarrow |J, M_J\rangle$ at the moments $t''_\kappa$:

$$\omega^*_{s\kappa} = \omega_s + \beta\Delta\omega/2/[1 + m\cos 2\Delta\phi(x = 0, t''_\kappa)] \approx$$
$$\approx \omega_{0J} \approx 10^{15} \text{ Hz}, \qquad (18b)$$

when it crosses the $X$ axis.

Let the diamagnetic molecules ($N_2$, $CO_2$, or AgCl) be *localized* on the rotational level $|1\rangle$ of the ground vibrational-electronic state with the rotational energy $W_{J=1} = \hbar 2B_e$ and with the angular momentum $\mathbf{J}_p = \hbar\sqrt{2}$. A small volume $\delta V' \approx \delta r\,\pi(r/2)^2$ near the molecular c.g. is considered as a space, in which the potential electric energy and, correspondingly, the

electric field strength can grow, whereas the degenerate rotational level $|J, M_J\rangle$ is considered as a potential source of $(2J + 1)$ angular momenta preserved in the form of $\sigma_\pm$-components of magnetic sublevels $M_J$.

Let the result of averaging of orbital perturbations at the frequencies $\Omega^*_{\kappa,\kappa+1}$ and $\omega^*_{s\kappa}$ over the initial orientations of molecules from the volume (10) for the period $T'_{s\kappa=0}$ give nonzero values of the electric and diamagnetic energy:

$$<\Omega^*_{\kappa,\kappa+1}> \approx \delta W_{S,\kappa=0} \approx \alpha''_{J=0}(\Omega^*_{\kappa,\kappa+1})\, E_0^2$$

and

$$<\omega^*_{s\kappa}> \approx \delta W_{Z,\kappa=0} \approx -\chi_{dnJ}(\omega^*_{s\kappa})\, H_0^2. \qquad (19)$$

From physical reasoning in Section 4, it follows that the energy of interaction of diamagnetic molecules with $\mathbf{E}$- and $\mathbf{H}$-components of the RSBF (8) can be transferred by the $V$-channel (4) due to autophasing of the PN motion of the bonding electrons at the moments $t'_\kappa$ and $t''_\kappa$, and the EMO mechanism (EMO effect) can self-organize in the molecular medium bounded by the volume (10), provided that the following conditions are fulfilled.

1. *Phases of forced oscillation* of the energy difference of the molecular rotational transitions (3) fall in resonance with the phases of fields induced in the molecule at the difference $\Delta\omega$ and sum $2\omega_s$ frequencies of the RSBF

$$(\Delta\omega/2 + \delta\omega)t \approx \Delta\phi_{10}(t) \text{ and } (\omega_s + \Delta\omega/2)t \approx \Delta\phi_{0J}(t). (20)$$

In this case, the efficiency of interaction of the molecules with $\mathbf{E}$- and $\mathbf{H}$-components of the RSBF (8) is determined by the "resonance capture of phases" of molecules and the field generated in them at the frequencies $\Omega^*_{\kappa,\kappa+1}$, $\omega^*_{s\kappa}$ with autophasing of PN motion of the bonding electrons, rather than spatial synchronism of secondary waves at the frequencies $\omega_1$ and $\omega_2$ (Ref. 20). "Resonance capture of phases" has the meaning of the local phase synchronism of secondary waves at the frequencies $\omega_1$, $\omega_2$ and fields generated in molecules at the frequencies $\Omega^*_{\kappa,\kappa+1}$, $\omega^*_{s\kappa}$.

2. *Intensity and frequency ratios* of the spectral components of the RSBF, as well as the angle $\gamma \neq 0$ between the vectors $\mathbf{E}_1$ and $\mathbf{E}_2$ must *have* the values

$$(I_1/I_2 \pm \Delta\omega/2\omega_s) \approx 5.8, \ (\omega_1/\omega_2) > 1, \ \gamma \approx \pi/6, \quad (21)$$

providing for the optimal AP loop (period $T'_{s\kappa}$) of oscillation of the RSBF in the medium: deviation $\Omega^*_{\kappa,\kappa+1} \approx 4B_e = 2\omega_{10}$ of the carrier frequency $\omega(t)$ at $\delta\Omega^*_{\kappa,\kappa+1} \neq 0$ and at the significant angles of ellipticity $\varphi$ and orientation $\theta$ of the polarization diagram of the wave (8) in the medium.

As a consequence, the bonding electrons in molecules at every period $T'_{s\kappa}$ are accelerated in the direction of the $Y$ axis at the frequency $\omega^*_{s\kappa}$ with in-phase tuning of shifting-splitting of levels of the transition $|1\rangle \leftrightarrow |0\rangle$ to the frequency $\Omega^*_{\kappa,\kappa+1}$. This

tuning is provided mostly by the dynamic Stark effect at the initial frequencies $\omega_1$, $\omega_2$ and the induced field $\mathbf{E}'_y$ at the frequency $\Omega^*_{\kappa,\kappa+1}$. The dynamic Zeeman effect at the initial frequencies $\omega_1$, $\omega_2$ and the induced field $\mathbf{H}'_z$ at the frequency $\omega^*_{s\kappa}$ provide mostly for control over electron motion in the direction of the $Z$ axis at the frequency $\omega^*_{s\kappa}$ with in-phase tuning of shifting-splitting of levels of the transition $|0\rangle \leftrightarrow |J, M_J\rangle$ to the frequency $\Omega^*_{\kappa,\kappa+1}$.

Molecules acquire the capability of *accumulating* the static electric field near the c.g. in the form of increments of polarizability $\alpha''(\omega) + \sum\kappa\delta\alpha_\kappa$ and the electric energy $\sum\kappa\delta W_{S,\kappa}$, as well as *filling* the source $|J, M_J\rangle$ by increments of MM $\mathbf{J}_p + \sum\kappa\delta J_{p\kappa}$, diamagnetic susceptibility $\chi_{\mathrm{d}nJ}(\omega) + \sum\kappa\delta\mu_\kappa$, and ==diamagnetic== energy $\sum\kappa\delta W_{Z,\kappa}$.

At the same time, the optimal PF arises between the state of molecules and the contributions of $\mathbf{E}$- and $\mathbf{H}$-components of the RSBF (8) to the transfer of the interaction energy by the channel (4) within double parametric resonance (20) (by analogy with Ref. 21). Molecules acquire the capability of receiving optimal increments $2\delta W_{S,\kappa}$ ($2\delta\alpha_\kappa$), $2\delta W_{Z,\kappa+1}$ ($2\delta\mu_\kappa$ and $2\delta J_{p\kappa}$) in the neighboring $\kappa$th and ($\kappa+1$)th periods $T'_{s\kappa}$, if the volume (10) is supplied with the optimal energy flux $\mathbf{S}_{\mathrm{opt}}$ of the field (8) at the optimal density of molecules $N_r$ (cm$^{-3}$).

3. *Energy flux* $\mathbf{S}_{\mathrm{opt}}$ of the pump field, angle, and density of molecules $N_r$ (cm$^{-3}$) must *provide* for the Eq. (17) to be valid, at which the molecules from the volume (10) can accumulate the critical MM $\mathbf{K}_p(t''_{\kappa=J}) \approx \sum\kappa\delta J_{p\kappa} \approx \hbar 2(J+1)$, as well as the critical value of the electric energy

$$(\delta W_0 + \delta W^*)/\hbar \le |F'| \le K_{\mathrm{am}} \sum\kappa |\delta W_{S,\kappa=0,1,2,...,J}/\hbar| \approx$$

$$\approx (4B_e + \delta\omega) \qquad (22\mathrm{a})$$

on the average for time $T'_{vp} \approx \sum\kappa T'_{s\kappa} \approx T \approx 1/B_e \approx 10^{-11}$ s by shifting the level $|0\rangle$ to the c.g. and the critical value of the ==diamagnetic== energy

$$(\delta W_0 + \delta W^*)/\hbar \le |F'| \le K_{\mathrm{am}} \sum\kappa |\delta W_{Z,\kappa=1,2,...,J}/\hbar| \approx$$

$$\approx (4B_e + \delta\omega) \qquad (22\mathrm{b})$$

by splitting the level $|J, M_J\rangle$ and shifting $\sigma_\pm$-components of its $M_J$-sublevels on the scale $s^2_\kappa = v^2_{\mathrm{ph}}(t'_\kappa - t''_\kappa)^2 - R^2_\kappa$.

As a consequence, every diamagnetic molecule from the coherency volume (10) acquires the capability of evolving into anisotropic diamagnetic "molecule + field" systems with artificial symmetry. Here $\delta W_0 \approx 4\hbar D|_{t=0}$ and $\delta W^* \approx \delta W_{S,\kappa=0}/\sqrt{(Q_1 Q_J)}$ are the energy of centrifugal stretching of molecule and the energy loss of a molecule for the relaxation of the acquired potential electric and ==diamagnetic== energy; $Q_J = c\,\nu_J/(\gamma_J P_r)$ is the Q-factor of the rotational level; $\gamma_J$ ($\mu$s · Torr)$^{-1}$ and $P_r$(Torr) are the rate of relaxation of molecular levels and the pressure of the

molecular gas; $F' \approx |1 - \beta/(1-m)|\Delta\omega/2$ is the area of the accelerating-controlling gap of the AP loop of oscillation of the RSBF (8); $K_{\mathrm{am}} = 1/[1 - m/m^*]$ is the coefficient of double-tuned parametric amplification of the PN motion of the bonding electrons of the molecule as a parametric amplifier.[21]

The value of $K_{\mathrm{am}}$ plays, in the first approximation, the role of PF. The efficiency of real PF depends on the degree of "resonance capture of phases" (20) and provides for the rate of generation of the signal at the *cooperative frequency*

$$\omega_p \approx 2\pi\,c\,A(J, j)|t''_{\kappa=J}\,J(J+1) \sim \omega_{0J} \qquad (23)$$

in molecule, evolution of molecules into the "molecule + + field" systems in the prepared state $|K(t''_{\kappa=J})M_J\rangle$ in the process of exhaustion of the depth of amplitude modulation $m \to m^* \approx \pi/\sqrt{(Q_1 Q_J)}$, the value $\mathbf{S}_{\mathrm{opt}} \to \mathbf{S}^*$, and the ratios $(E_1/E_2) \to (E_1/E_2)^*$, $(H_1/H_2) \to \to (H_1/H_2)^*$. Here $A(J, j)|t''_{\kappa=J} \approx B_e$ is the parameter of multiplet splitting, which depends on $J' = s' + j$, $j = \ell + s$ and determines the value of multiplet splitting of rotational levels in the prepared state $|K(t''_{\kappa=J})M_J\rangle$.

## 6. Dynamics of preparation of molecular coherence and generation of field with high MM

The process of preparation of the molecular coherence (EMO effect) due to the EMO mechanism in the molecular medium consists in the following.

Molecules are captured at $t''_{\kappa=0} \approx 0$ in AP loops of oscillations of the RSBF. Transferring the increments $2\delta\alpha_\kappa$, ($2\delta W_{S,\kappa-1}$) to the c.g., and $2\delta\mu_\kappa$ ($2\delta W_{Z,\kappa-1}$) to the level $|J, M_J\rangle$ by the channel (4) for every period $T'_{s\kappa} \approx 10^{-15}$ s, molecules make the rotational transition $|1\rangle \to |0\rangle$ in the hysteresis mode on the average for the duration of the wave packet $T_{vp} \approx \sum\kappa T'_{s\kappa} = = J(J+1)\,T'_{s\kappa}/2 \approx T \approx 1/B_e \approx 10^{-11}$ s, Eq. (1). As this occurs, the molecules lose their natural angular momentum $J_p = \hbar\sqrt{2} \to 0$, but acquire the artificial angular momentum $\mathbf{K}(t''_{\kappa=0}) \approx \delta\mathbf{J}_{p\kappa=0}$ ($\mathbf{K}(t''_{\kappa=0})\|X')$, whose absolute value increases $\delta\mathbf{J}_{p\kappa=0} \to \mathbf{K}(t''_{\kappa=J}) \approx \hbar 2(J+1)$ simultaneously with the change of its orientation about the $X'$ axis up to the angle $\theta$.

The coherent combination of the processes of decrease of $\mathbf{J}_p$ and increase of $\mathbf{K}(t''_{\kappa=0,1,2,...,J})$ in space and time is accompanied by the increase of the effective radius $r \approx r_{\mathrm{L}} \to R^* \approx R_0$ of the orbit of PN motion of molecular electrons due to generation of the field at the frequencies $\Omega^*_{\kappa,\kappa+1}$ ($\mathbf{E}'_y$) and $\omega^*_{s\kappa}$ ($\mathbf{H}'_z$). The natural rotation of the bonding electrons evolves into the stable PN motion, and the corresponding set of "captured" molecules evolves into the ensemble of "diamagnetic molecule + field" systems in the prepared superposition state $|K(t''_{\kappa=J})M_J\rangle$.

The energy acquired by molecules and the degree of alignment of projections of the artificial angular momentum $\delta \mathbf{J}_{p\kappa=0} \to \mathbf{K}(t''_{\kappa=1,2,...,J})$ about the $X'$ axis at every following period $T'_{s\kappa}$ depend on the energy acquired and alignment at the previous cycle $T'_{s\kappa-1}$. Just in this sense the molecular coherence "captures" and memorizes photons of the field (8), which intensify the interaction of the electron motion with molecular rotation, i.e., the electron motion in molecule, twisting (artificial internal rotation) about the c.g., prepares molecular coherence about the $X'$ axis in the form of ordered orbital perturbations at the frequencies $\Omega^*_{\kappa,\kappa+1}$, $\omega^*_{s\kappa}$ of the "diamagnetic molecule + field" system. The artificial symmetry of every "diamagnetic molecule + field" system is kept by the interaction of the electron motion with rotation via photons of the pump field.

The EMO effect can be interpreted as an electromagnetic *spiral phase structure* prepared of $(2J + 1)$ $\sigma_\pm$-components of Zeeman sublevels of the state $|J,M_J\rangle$ and the level $|0\rangle$ of molecules, as well as the induced fields at the frequencies $\Omega^*_{\kappa,\kappa+1}$ ($\mathbf{E}'_y$) and $\omega^*_{s\kappa}$ ($\mathbf{H}'_z$), or as *molecular coherence* prepared in the form of superposition of orbital perturbations of the bonding electrons of the ensemble of "diamagnetic molecule + field" systems. In this case, the level $|0\rangle$ and $\sigma_\pm$-components of magnetic sublevels $M_J$ of the level $|J, M_J\rangle$ occupy the position corresponding to the bonded superposition of electron orbital perturbations of the ensemble of "molecule + field" systems at the frequencies $\Omega^*_{\kappa,\kappa+1}$ and $\omega^*_{s\kappa}$.

The state $|K(t''_{\kappa=J})M_J\rangle$ includes the level $|0\rangle$ and $(2J + 1)$ sublevels of the rotational level $|J, M_J\rangle$ with the aligned projections (spins) of the total moment of momentum onto the axis $X'$: $M_x = J_p = 0, \pm \hbar, \pm 2\hbar$, ..., $\pm J\hbar$, and is the bonded superposition of orbital perturbations of the ensemble of "diamagnetic molecule + field." The ensemble of "molecule + field" systems is a potentially magnetic medium with the anisotropic distribution of projections of the angular momenta $\mathbf{K}(t''_{\kappa=J})$ onto the $X'$ axis (spatial inversion[11]).

The prepared ensemble of "molecule + field" systems *self-excites* within $T \approx 1/B_e \approx 10^{-11}$ s and makes the radiative transition between the states having the spatial coordinates: $R \to -R$ ($x \to -x$, $y \to -y$, $z \to -z$) (Ref. 11), i.e., the ensemble of "molecule + field" systems generates the field with MM $\mathbf{K}_p \approx \hbar 2(J + 1)$ at the cooperative frequency (23) and is simultaneously filled by energy and MM for the period $T_p \approx 2\pi/\omega_p \approx 10^{-15}$ s by the closed absorption-emission scheme.

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