

# Possibility of preparing a molecular state for generation of electromagnetic field with high angular momentum

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Grounds are given for the approach to preparation of an anisotropic diamagnetic medium from a set of "molecule + field" systems in a dynamic (multiplet) state.

1. Radiation carries the fluxes of energy, momentum, and angular momentum away from a source. The character of angular momentum (orbital, spin) and its value are closely related to the spatial peculiarities of the radiation source and the wave process, and the law of conservation of the angular momentum is connected with the spatial isotropy and holds in the "molecule + field" systems (Ref. 1). The spatial structure of a laser wave is such that even intense radiation carries the angular momentum  $M_x = \pm \hbar$ ;  $h = 2\pi\hbar$  is the Planck's constant or quantum of action.

Quantum electrodynamics allows the existence of a field with  $M_x \gg \hbar$ , and some peculiarities of such a radiation have been investigated theoretically.<sup>2</sup> It was noted, in particular, that such a field (even of a low intensity) must induce high magnetic moment in a unit volume of the medium and change the medium reflectivity. Therefore, the study of the field with  $M_x \gg \hbar$  is interesting not only from pure scientific, but also from a practical point of view, in particular, for solution of the problems of optical wave propagation in the atmosphere.

Laser physics and molecular spectroscopy suggest no methods for generation of a field with  $M_x \gg \hbar$ . This is caused by a very low probability of the transition of a molecule, as a radiation source, to a high rotational level, for example,  $|J' \sim 100, M' \rangle$  followed by the radiative transition to the lowest level  $|0 \rangle$ ;  $J'$  and  $M'$  are the quantum numbers of the total angular momentum of a molecule and its projection on the direction of the exciting field polarization. The situation does not change even when the strength of the exciting field  $E_0$  is sufficient to rearrange the rotational motion of the molecule and to change the probability of molecular transitions.<sup>3,4</sup> That means that, the frequency resonance between molecules and the field and the statistical approach to formation of molecular transitions do not lead to accumulation of qualitatively new contributions to the intramolecular motion and, correspondingly, to distortion of the spatial distribution of the potential energy of molecular rotation. As a

consequence, there is no real possibility of generating the field with  $M_x \gg \hbar$  by a molecular medium.

The conclusion suggests itself that a significant change of the probability of rotational transitions and distortion of the electron–nucleus configuration of the potential energy of rotation with the corresponding accumulation of high angular momentum by the molecule can be obtained only at a certain spatiotemporal (amplitude-frequency) configuration of the perturbing field, and this configuration should emphasize the vector nature of light and the law of conservation of angular momentum. So, the problem is to find and consider, within the framework of quantum mechanics and Maxwell equations, such peculiarities of molecules, pump field, and conditions of their interaction at the critical period of the intramolecular motion  $T_{cr} \approx 1/B_e$ , which were earlier ignored because of their low individual significance. Here  $B_e$  is the rotational constant of a diatomic molecule.

The aim of this paper is to find a combination of the peculiarities and conditions of the energy exchange between the field of optical biharmonic ( $\omega_1$  and  $\omega_2$ ) and a medium of diatomic molecules, which would allow the molecules and the field to prepare an anisotropic diamagnetic medium from a set of "molecule + field" systems in the dynamic (multiplet) state for the period  $T_{cr} \approx 1/B_e$ , on average, and then to generate radiation with  $M_x \gg \hbar$  at the cooperative frequency  $\omega_p$ .

2. The first peculiarity is connected with the participation of not only two biharmonic waves ( $\omega_1$  and  $\omega_2$ ) linearly polarized along  $Y$  axis and propagating along the  $X$  axis, but also the waves of square electric and magnetic polarization induced in the molecular medium at the difference  $\Delta\omega = (\omega_1 - \omega_2)$  and sum  $2\omega_s = (\omega_1 + \omega_2)$  frequencies.<sup>5</sup> Usually, the amplitude of square-polarization waves is so low that they remain unnoticed. An exception can be only the situation at propagation of a certain superposition of intense biharmonic waves and weak square-polarization waves in the molecular medium is connected with their spatiotemporal dispersion at a marked change of the electronic polarizability  $\epsilon_j(\omega_1, \omega_2)$  and diamagnetic

susceptibility  $\chi_{dJ}(\omega_1, \omega_2)$  of molecules. In this case, the amplitude  $\gamma$  of the change of electro- and magneto-optical characteristics of the medium at the carrier frequency  $\omega_s = (\omega_1 + \omega_2)/2$  of the resulting strength of the bichromatic field can be even amplified up to rather a high value. For example, due to the feedback that may occur between the precession-nutation motion of electrons and the orbital revolution of the nuclei of the medium molecules during the period  $T_{cr} \approx 1/B_e$ .

To reveal prerequisites for the appearance of the feedback between the motion of electrons and nuclei in the molecule, let us express the resulting strength of the bichromatic field in terms of the balance amplitude-phase modulation<sup>6</sup>

$$E(t) = E_1 \exp -i(\omega_1 t + \psi_1) + E_2 \exp -i(\omega_2 t + \psi_2) + c.c. \equiv A(t) \exp i\Phi_s(t) + c.c. \quad (1)$$

It is the difference  $\Omega(\Delta t'_{K,K+1}) = \Delta\omega/2 \cdot \beta/f(t'_K)$  in the carrier frequency  $\omega_s$  of the resulting field at the neighboring moments in time  $t'_K$  that is of interest. Here  $f(t) = [1 + m \cos 2\Delta\phi(t)]$  is the function that modulates the amplitude  $A(t) = E_0 f(t)^{1/2}$  of the resulting field with the frequency modulation  $\sim \Delta\omega/2$ ;  $m = 2E_1 E_2/E_0^2$  and  $\beta = (E_1^2 - E_2^2)/E_0^2$  are the depth of amplitude modulation and the index of phase modulation;  $E_0 = (E_1^2 + E_2^2)^{1/2}$ ;  $2\Delta\phi(t) = (\Delta\omega t + \Delta\psi)$ ;  $\Delta\psi = (\psi_1 - \psi_2)$  is the difference between the initial phases of the biharmonic. We use  $K = 0, 1, 2, \dots, J'$  to denote the number of a rotational level and half-cycle

$$T_{s,K}/2 = \pi/\omega''_{s,K} = \pi/\{\omega_s + \beta\Delta\omega/2/[1 + m \cos 2\Delta\phi(t''_K)]\} \approx 10^{-16} \text{ s}$$

of the carrier frequency  $\omega_s$  during the period  $T_{cr} \approx 1/B_e$ , having the meaning of the duration of the wave packet of the resulting field. Here  $\omega''_{s,K} = \omega_s + \Omega(t''_K)$  is the instantaneous value of the carrier frequency  $\omega(t) = d\Phi_s(t)/dt = \omega_s + \Omega(t)$  at the time  $t''_K$ ;  $\Phi_s(t) = [\omega_s t + \psi(t) + \psi_s]$ . The time moments  $t'_K = (K - 1/2)T_{s,K}/2$  and  $t''_K = K T_{s,K}/2$  correspond to the maximum and minimum (alternation of signs) values  $\pm \mathbf{E}(r, t, \omega_1, \omega_2)$ ,  $\pm \mathbf{H}(r, t, \omega_1, \omega_2)$  of the vectors of the resulting field strength.

The deviation  $\Omega(t'_{K,K+1})$  connects the square-polarization waves at the time interval

$$10^{-16} \text{ s} \approx 2\pi/\omega_s \ll \Delta t \ll 4\pi/\Delta\omega \approx 10^{-12} \text{ s} \quad (2)$$

and the lags of the responses of the electric and magnetic polarization of the medium to the applied field of the optical biharmonic. In this case, at the moment  $t''_K$  the behavior of any molecule that happened to be within the coherent volume

$$V \approx \pi\Delta R^2 \Delta L \quad (3)$$

depends on the action of the vectors  $\pm \mathbf{E}(r, t, \omega_1, \omega_2)$  and  $\pm \mathbf{H}(r, t, \omega_1, \omega_2)$ , at the same moment, on other molecules in the spatial interval

$$3 \cdot 10^{-6} \text{ cm} \approx 2\pi v_{ph}/\omega_s \ll v_{ph}\Delta t \ll 4\pi v_{ph}/\Delta\omega \approx 3 \cdot 10^{-2} \text{ cm}. \quad (4)$$

That means that, locality of the response of the molecules of a medium to the radiation is violated within the spatial interval (4) and, as a consequence, the strengths of the effective electric  $E'$  and magnetic  $H'$  fields differ from that of the mean Maxwell field.<sup>5</sup> Here  $\Delta R \approx v_{ph}\pi/\Delta\omega$  is the size of the medium layer at the entrance  $x = 0$  in the plane  $YZ$  covered by the pump field;  $\Delta L \approx v_{ph}2\pi/\delta\omega$  is the length of the time coherence of the field. The parameter  $\Delta L$  shows that between the points  $x = 0$  and  $x = L$  there is some fixed difference (and sum) between the phases of two traveling waves of the frequencies  $\omega_1$  and  $\omega_2$ , and the phase coincidence is not obligatory;  $\delta\omega \approx \delta\omega_1 \approx \delta\omega_2 \ll |\omega_1 - \omega_2|$  is the width of spectral components of the pump field;  $v_{ph} = c/n$  is the phase velocity of the wave in the medium;  $n = \sqrt{\epsilon\chi}$ ,  $\epsilon$ ,  $\chi = \chi_p + \chi_d$  are the refractive index, electric permeability, and sum of paramagnetic and diamagnetic susceptibilities of the medium at the frequency  $\omega(t)$  making cyclic loops  $T_{s,K}$  about its mean value  $\omega_s$ .

Since the value and the sign of the deviation  $\Omega(\Delta t'_{K,K+1})$  depend on the frequency  $(\omega_1/\omega_2) >< 1$  and intensity  $(E_1/E_2)^2 >< 1$  ratios of the biharmonic components, at the moments  $t'_K$ , the deviation can determine the direction and the rate of energy exchange between the biharmonic waves and molecules via the induced waves of square polarization. In this case the control parameter  $(E_1/E_2)^2 \neq 1$  determines:

– the integer number of cyclic loops  $\sum K T_{s,K}/2 = J'(J' + 1)T_{s,K}$  on the interval (2);

– the difference (uncompensated step)  $\delta\Omega(\Delta t'_{K,K+1})$  of the deviation  $\Omega(\Delta t'_{K,K+1})$  on “odd” and “even” half-cycles  $T_{s,K}/2$  of the instantaneous frequency  $\omega(t)$  with respect to the time moments  $t''_K$ .

The step  $\delta\Omega(\Delta t'_{K,K+1})$  determines the difference of the areas  $\sigma$  described by the end of the vector  $\mathbf{E}$  (pseudovector  $\mathbf{H}$ ) in the plane  $YX$  ( $ZX$ ) during “odd” and “even” half-cycles  $T_{s,K}/2$  with respect to the axis  $X$  and, correspondingly, the step  $\mathbf{q}$  of the change of the electron polarizability

$$\epsilon_f(\omega_1, \omega_2) = 1 + \gamma(\omega_1, \omega_2) \cos(\mathbf{q}\mathbf{r}) \quad (5)$$

(diamagnetic susceptibility  $\chi_{dJ}(\omega_1, \omega_2)$  of the medium) for  $T_{s,K}$ . The coefficient  $\gamma$  is the small amplitude of the change in  $\epsilon$ , and  $\mathbf{q}$  indicates the direction and the step of this change. The change of the electro- and magneto-optical characteristics of the medium is accompanied by the change of the intramolecular current and charge distribution in molecules within the coherent volume (3).

The second peculiarity of the energy exchange between the waves is associated with the opposite  $J$  dependence of the electron polarizability  $\epsilon_f(\omega)$  and diamagnetic susceptibility  $\chi_{dJ}(\omega)$  of molecules for optical frequencies  $\omega$  (Refs. 4 and 7). As a consequence, the shift of the lowest molecular energy

level  $|0\rangle$  is mostly determined by the action of the electric component of the resulting field strength (quadratic Stark effect) in the plane  $YX$ , whereas the shift of the high level  $|J', M'\rangle$  is determined by the magnetic component (quadratic Zeeman effect) in the plane  $ZX$ . Polarization of the quadrupole and magnetic dipole rotational transitions

$$|1\rangle \leftrightarrow |0\rangle \text{ and } |0\rangle \leftrightarrow |J', M'\rangle \quad (6)$$

at the frequencies  $\Delta\omega$  and  $2\omega_s$  of the secondary waves acquires certain dependence on the difference between the polarizability of molecules at the initial  $|1\rangle$  and final  $|J', M'\rangle$  rotational levels. This spatial inhomogeneity of perturbation of electron motion in a molecule at the frequencies  $\Delta\omega$  and  $2\omega_s$  and uncompensated step of the deviation  $\delta\Omega(\Delta t'_{K,K+1})$  lead to appearance of the feedback between the perturbing and the intramolecular fields and, correspondingly, to the feedback between the precession-nutation motion of electrons and the orbital revolution of nuclei of molecules during the period  $T_{cr} \approx 1/B_e$ .

The third peculiarity of the energy exchange is that the lowest rotational level  $|0\rangle$  is involved in it and the minimum potential energy of the molecule-rotator is equal to the difference between the potential energy of rotation at the equilibrium point  $r \approx r_e$  and at the position of the level  $|0\rangle$ .

This energy occupies the infinitely small volume  $\delta V \approx \delta r \pi(r_e/2)^2$ , which can play a principal part in the change of the regime of intramolecular motion, the structure of rotational energy levels, and the probability of transitions between them for the following reason. Near the level  $|0\rangle$  the natural angular momentum  $\mathbf{J} = 0$  of the molecule-rotator and its properties manifest themselves in the quasi-classical limit. It is just the scale  $kr_e \ll 1$ , where  $E_r \sim 1/r_e^2$  and  $H \sim 1/r_e$  (for magnetic multipole  $E \sim 1/r_e$  and  $H \sim 1/r_e^2$ ), near the point  $r \approx r_e$  that determines the finite contribution of the perturbing field to the angular momentum of the incipient spherical wave (Gaitler statement<sup>8</sup>). That is, on the scale  $kr_e \ll 1$  a link can arise between the spatial and spin subsystems of the molecule via the spin moment  $s'$  of photons of the perturbing field. Here  $k = 2\pi/\lambda$  is the wave number;  $\delta r \approx (r - r_e)$  is the shift of nuclei from the equilibrium position.

3. The presented peculiarities of the energy exchange between the waves and the molecular medium allow three conditions to be used for preparing an ensemble of "molecule + field" systems in the dynamic (multiplet) state.

First, the biharmonic waves must induce two volume dynamic grids of square polarization in the medium at such frequency difference  $\Delta\omega$  and the sum  $2\omega_s$  that the phases of the grids fall within the area of "lock-in" by the phases of oscillation of the difference between the energy levels of the quadrupole  $\Delta\phi_{01}(t)$  and magnetic dipole  $\Delta\phi_{0J'}(t)$  rotational transitions (6). In this case, each of the amplitudes  $Q^\Delta$  and  $Q^{2s}$  of

polarization oscillations determines the rate and direction of the energy exchange between the waves and the medium on the time (2) and spatial (4) scales of its grid. Detuning between  $\omega_1$  and  $\omega_2$  becomes dependent on the dispersion of the refractive index  $n(\omega_1, \omega_2)$  of the medium formed by the Stark (Zeeman) shift, among other factors, of the difference between the levels of rotational transitions (6) and on the diffraction of the interacting waves.<sup>5,9</sup> If the contribution of the shift of the molecular energy levels (6) to  $n(\omega_1, \omega_2)$  [see Eq. (3)] exceeds all other contributions at the initial moment  $t''_{K=0}$  of the interaction, then the direction of the energy exchange between the waves and the medium is determined by the "lock-in" of the phase of the square-polarization waves by the phases of oscillation of the difference between levels of the adjacent molecular transitions (6), rather than the wave detuning.

Second, the deviation  $\Omega(\Delta t'_{K,K+1})$  must link the square-polarization waves at the frequencies  $\Delta\omega$  and  $2\omega_s$  via the rotational levels of the quadrupole and magnetic dipole transitions (6) within the cyclic loops  $T_{s,K}$  so that the instantaneous value of the carrier frequency  $\omega(t)$  be in resonance with the transitions (6) at the moments  $t'_K$  and  $t''_K$ , respectively.

In this case, further behavior of each molecule-rotator from the volume (3) under the action of  $\delta$ -accelerations from  $\pm\mathbf{E}(r, t, \omega_1, \omega_2)$  and  $\pm\mathbf{H}(r, t, \omega_1, \omega_2)$  vectors of the pump field at the moments  $t'_K$  agrees not only by the time interval (2), but by the space interval (4) as well. The agreement manifests itself in the fact that the electron orbits are deformed in the plane  $YX$  with respect to the center of gravity at the moments  $t'_K$  and the orbital revolution of electrons about the artificial angular momentum  $\mathbf{J}'$  accelerates in the plane  $ZX$  and removes the spatial degeneration of the levels (6) with the rate  $\omega''_{s,K} \approx \omega_s + \Omega(t''_K)$ .

Since the molecule-rotator is near the level  $|0\rangle$ , the above-mentioned manifestations of the Stark and Zeeman effects superimpose in the planes  $YX \perp ZX$  and give rise to the forced nutation and precession of the electron orbits about the vector  $\mathbf{J}'$  at the initial moment  $t''_{K=0}$  of the interaction. If the nutation period is assumed to be equal to  $T_{nut} \approx T_{s,K} \approx 2\pi/\omega_{0J'}$  i.e., to be inverse to the frequency of the magnetic dipole transition  $|0\rangle \leftrightarrow |J', M'\rangle$ , then nutation and precession of electrons are "locked-in" by the half-cycles  $\Delta t'_{K,K+1}$  of the neighboring alternations of the sign of the vector  $\pm\mathbf{E}(y, t, \omega_1, \omega_2)$  and the direction of rotation of the pseudovector  $\pm\mathbf{H}(z, t, \omega_1, \omega_2)$  of the pump field. The sum of  $K$  periods of nutation of electron orbits is equal to the period of their precession  $\sum T_{nut} \approx \sum T_{s,K} \approx 2\pi/\Omega(\Delta t''_{K=0, K=J'})$ , and the nutation phase falls within the area of "lock-in" by the phase of oscillation of the difference between the levels  $\Delta\phi_{0J'}(t)$  of the magnetic dipole transition. Simultaneously, the phase of precession of electron orbits falls within the area of "lock-in" by the phase of oscillation of the

difference between the levels  $\Delta\phi_{01}(t)$  of the quadrupole transition (6), and the frequency of precession and orbital revolution of molecular nuclei  $\Omega_{pr} \approx \Omega_{nucl}$  falls in resonance with the amplitude modulation at the frequency  $\Delta\omega/2 \approx 2B_e$  of the resulting strength of the bichromatic field.

The uncompensated step  $\delta\Omega(\Delta t'_{K,K+1})$  of deviation of the carrier frequency  $\omega_s$  leads to appearance of the feedback (phase memory) between the precession-nutation motion of electrons and the orbital revolution of nuclei, so:

- the half-cycle  $\Delta t'_{K,K+1} = (t'_K - t'_{K+1})$  between two neighboring maximum values of the vector  $\pm\mathbf{E}(r, t, \omega_1, \omega_2)$  of the resulting field strength becomes dependent on the electric energy  $W_{S,K}$ , the molecules received from the field at the previous half-cycle  $\Delta t'_{K-1,K}$  due to quadratic Stark effect with respect to the level  $|0\rangle$ ;

- the half-cycle  $\Delta t''_{K,K+1} = (t''_K - t''_{K+1})$  between two neighboring alterations of the direction of rotation of the pseudovector  $\pm\mathbf{H}(r, t, \omega_1, \omega_2)$  becomes dependent on the diamagnetic energy  $\delta W_{Z,K}$ , the molecules received from the field at the previous half-cycle  $\Delta t''_{K-1,K}$  due to quadratic Zeeman effect with respect to the level  $|J', M'\rangle$ ;

- the cyclic loops  $T_{s,K}$  are transformed into hysteresis spiral loops  $T'_{s,K}$  of the phase memory, within which the energy exchange is directed toward the transfer of the energy  $\delta W_{S,K}$  and  $\delta W_{Z,K}$  and the spin moment  $s'$  of photons of the pump field to electrons of the molecules.

The size of the hysteresis loop  $T'_{s,K}$  depends on the initial density of the energy flux  $\mathbf{S}$ , as well as on the rate of its depletion and depletion of the control parameter  $(\mathbf{E}_1/\mathbf{E}_2)^2$  of the pump field to the extent, at which the period  $T_{cr} \approx 1/B_e$  and the time interval (2) self-organize in the sum of the integer number of loops  $\Sigma K T'_{s,K}/2$ . Within the hysteresis loop  $T'_{s,K}$ , the result of intensification of the precession of electron orbits (and deceleration of nucleus rotation around the vector  $\mathbf{J} = \hbar\sqrt{2} \rightarrow 0$  via the feedback) by  $\delta$ -accelerations from the vectors  $\pm\mathbf{E}(y, t, \omega_1, \omega_2)$  and  $\pm\mathbf{H}(z, t, \omega_1, \omega_2)$  of the pump field at the moments  $t'_K$  is accompanied by the change of the orbital current of the linking electron, and the result of the forced nutation is accompanied by the change of the magnitude and direction of the artificial angular momentum  $\mathbf{J}'(t''_{K=0}) = 0 \rightarrow \mathbf{J}'(t''_{K=1,2,\dots,J'})$ . The forced intramolecular motion intensifies (shift-decay of the levels (6)) at the moments  $t'_K$ , and the corresponding phase space of the molecule is filled by a dosed amount of uncompensated energies  $\delta W_{S,K}$  and  $\delta W_{Z,K}$  at the moments  $t''_K$ .

Third, the initial density of the energy flux  $\mathbf{S}$  of the pump field must create such values of  $\delta W_{S,K}$  and  $\delta W_{Z,K}$  that the "lock-in" of the phase of the secondary waves at the frequencies  $\Delta\omega$  and  $2\omega_s$  by the phases of oscillation of the difference between the levels of the transitions (6) is accompanied by autophasing of the

forced intramolecular motion with the tunnel transition of molecules via the channel

$$|1\rangle \rightarrow |0\rangle \leftrightarrow |J', M'\rangle \quad (7)$$

and accumulation of the level  $|J', M'\rangle$  splitting.

Autophasing of the forced intramolecular motion can occur in the case, when rotational SRS and energy exchange between the fluxes  $\mathbf{E}_1$  and  $\mathbf{E}_2$  due to oscillations at the Stark shift frequency are less efficient than formation of square-polarization waves at the frequencies  $\Delta\omega$  and  $2\omega_s$ . This is possible if the frequency detuning of the field is comparable with the frequency of rotation of molecular axis and significantly exceeds the relaxation rate of polarization  $|\Delta\omega| \approx 4B_e \gg \gamma_{01}$ , and the energy density flux  $\mathbf{S}$  of the field provides for such an excess of the strength  $E_0$  ( $H_0$ ) above the threshold value  $\mathbf{E}_0 \geq \mathbf{E}_{thr}$  ( $\mathbf{H}_0 \geq \mathbf{H}_{thr}$ ), at which the sum of the shifts of the electron orbit for  $K$  cycles  $T'_{s,K}$  exceeds its rotational constant

$$\Sigma K |\epsilon_{01}(0, \omega_1, \omega_2, E_0^2) E_0^2|/4 \geq B_e, \quad (8a)$$

$$\Sigma K |\chi_{dJ'}(\mathbf{J}', \omega_1, \omega_2, H_0^2) H_0^2|/4 \geq B_e. \quad (8b)$$

In this case, the regime of natural intramolecular motion can transform into the regime of autophasing of the precession-nutation motion of electrons and arrangement of the molecular axes along the artificial angular momentum  $\mathbf{J}'$ , on average, during the period  $T_{cr} \approx 1/B_e$ .

This electromagnetic effect we predict for an ensemble of molecules is an optical analog of the Veksler effect of autophasing.<sup>10</sup> Veksler proposed the mechanism of autophasing for accelerators of the cyclotron type as a way to conserve, for unlimited time, the resonance between the stable motion of relativistic particles in the accelerator space and a high-frequency field: the electric one as accelerating and the magnetic field as a control one. Oraevskii found the optical analog of this mechanism for a molecule-oscillator and used it for preparation of a dynamic (superexcited) vibrational state.<sup>11</sup>

4. These three conditions allow the following model to be proposed for preparation of the dynamic state of an electro-dipole diatomic molecule in the field of optical biharmonic.

Let the molecule be in the ground electron-vibrational state  $X^1\Sigma^+$  and walk over rotational levels due to collisions with neighboring particles. This walk lasts until at some collision the molecule takes the level  $|1\rangle$  at the moment  $t''_{K=0} = 0$  of the beginning of interaction with the field of biharmonic (1). The state of the molecule  $|1, M\rangle = |H_{z0} M\rangle$  becomes nonstationary, as it can be denoted as  $|J'(t''_{K=0}) M'\rangle$ . The absolute value of the natural angular momentum of the molecule is  $\mathbf{J}(t''_{K=0}) = \hbar\sqrt{2}$ , and that of the artificial momentum (oriented along the field  $\mathbf{J}'(t''_{K=0}) \parallel H_{z0}(t''_{K=0})$ ) is  $\mathbf{J}'(t''_{K=0}) \approx 0$ .

The electric and magnetic components of the resulting field strength induce dipole moments in the molecule at the frequencies  $\Delta\omega$  and  $2\omega_s$  and perturb the orbital revolution of its electrons at each hysteresis loop  $T'_{s,K}$  of the period  $T_{cr} \approx 1/B_e$ . The resonance (in phase) feedback or phase memory arises between the perturbing and the molecular fields because of the uncompensated step of the deviation  $\delta\Omega(\Delta t'_{K,K+1})$ . This feedback leads to periodical intensification of the precession-nutation motion of electrons about the vector  $\mathbf{J}'(t''_{K=1,2,\dots,J'})$  and to termination of the orbital revolution of nuclei about the vector  $\mathbf{J}$  with a certain step  $\mathbf{q}$ .

As a consequence, the amplitude  $\gamma$  of nonlinear corrections of magnetic optical characteristics of the molecular medium at the frequencies  $\Delta\omega$  and  $2\omega_s$  and the forced intramolecular motion near the point of bifurcation of  $|0\rangle$  increases at each loop  $T'_{s,K}$  simultaneously with the dosed growth of the multiplet splitting of the levels (6) and the sum of uncompensated energies  $\delta W_{S,K}$  and  $\delta W_{Z,K}$ . That is, molecules memorize the phases of the pump waves and use them along with the amplitude to accumulate the orbital moment  $l$  of linking electrons and the spin moment  $s'$  of photons of the biharmonic, as well as to prepare the dynamic state in the "molecule + field" systems and to generate a spherical wave with the angular momentum  $M_x = M' \hbar$ .

As this takes place, the regime of the natural intramolecular motion transforms into the regime of the stable precession-nutation motion of electrons with the arrangement of molecular axes with respect to the vector  $\mathbf{J}'(t''_{K=0,1,2,\dots,J'})$  at the rate  $\omega''_{s,K} \approx \omega_s + \Omega(t''_K)$ . The molecules receive the electric  $\delta W_{S,K}$  and diamagnetic  $\delta W_{Z,K}$  energies and the spin moment  $s'$  of the absorbed photons and distribute the spin moment among the moments ( $j = l + s$ ) of an electron and the orbital moment of nuclei  $R$  during the period  $T_{cr} \approx 1/B_e$ . Thus, the molecules undergo the tunnel transition via the channel (7) similarly to the atomic transition due to intensification of the Larmor precession in an external alternating magnetic field.<sup>7</sup>

The tunnel transition of molecules via the channel (7) to the prepared dynamic (multiplet) state  $|\mathbf{J}'(t''_{K=J'}) M'\rangle$  leads to formation of the anisotropic diamagnetic medium consisting of "molecule + field" systems, on the average, during the period of the intramolecular motion  $T_{cr} \approx 1/B_e$ . The dynamic state  $|\mathbf{J}'(t''_{K=J'}) M'\rangle$  includes the lowest level  $|0\rangle$  and  $M' = (2J' + 1)$  of the magnetic sublevels of the high level  $|J', M'\rangle$ .

The medium consisting of the "molecule + field" systems is capable of generating the spherical wave with the angular momentum  $M_x = M' \hbar$  in the self-excitation mode at the cooperative frequency<sup>12</sup>:

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

on the time interval equal to the coherence time of the biharmonic field

$$T_{cr} \approx 1/B_e \leq \Delta t_{gen} \ll T_{coh} \approx 2\pi/\delta\omega. \quad (9)$$

Here  $A(J', j) \Big|_{t''_{K=J'}} \approx B_e$  is the multiplet splitting parameter depending on  $J' = s' + j$  and  $j = l + s$  and determining the value of the multiplet splitting of the rotational levels in the prepared state  $|\mathbf{J}'(t''_{K=J'}) M'\rangle$ .

The interaction of the field of biharmonic  $\omega_1$  and  $\omega_2$  with the systems "molecule + field" at the interval (9) differs drastically from the interaction with molecules during the period  $T_{cr} \approx 1/B_e$ . Now the interaction proceeds in agreement with the principle of removal of the prepared quanta of energy and quanta of action  $M' \hbar$  at the frequency  $\omega_p$  from the multiplet state of each of the "molecule + field" systems and preparation of new quanta of action  $M' \hbar$  in it at the frequency  $\omega_p$  for each closed "absorption-emission" cycle  $T_p = 2\pi/\omega_p \approx T'_{s,K}$ .

If no additional interaction arises between individual "molecule + field" systems of the anisotropic diamagnetic medium, then in the time interval  $T_{coh}$  it decays into the "electromagnetic background" and free molecules in some rotational-vibrational-electronic state and becomes isotropic again.

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