

INTERACTION OF LASER RADIATION WITH MATTER. LASER PLASMA

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Phase-sensitive electric modulation of photoluminescence upon bichromatic excitation of atoms

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***Abstract*. A new type of modulation of the photoluminescence intensity of atoms excited by a bichromatic laser radiation with the frequency ratio 1 : 2 is proposed and analysed. The modulation is produced by alternating electric field acting on atoms and occurs due to the quantum interference of the amplitudes of two excitation channels of an atom, which proves to be possible because the applied electric field removes the parity selection rule for one of the excitation channels. An important feature of this process is the dependence of photoluminescence on the phase difference of monochromatic components of exciting radiation. The calculation was performed for an alkali metal atom excited at the s – s transition taking the saturation effect into account.**

Keywords: *photoluminescence, bichromatic radiation, quantum interference.*

1. Introduction

The control of the rate of a photoinduced process by varying the phase difference between the monochromatic components of radiation initiating the process is called coherent (phase) control in a bichromatic field. This phenomenon is based on the interference between the quantum-mechanical amplitudes of two channels of the system transition from the initial to final state excited by the monochromatic components of the bichromatic field representing a coherent mixture of the fundamental and second-harmonic radiation.

Various types of coherent control in a bichromatic laser field were studied theoretically and experimentally in many papers (see reviews [1, 2]). The coherent control of photoionisation of atoms [3 – 5], photodecomposition of negative ions [6], photoionisation [7] and photodissociation [8] of molecules, external [9, 10] and internal [11] photoeffect was considered, as well as the control of a number of 'photo-assisted' processes in a bichromatic laser field was studied: scattering of electrons [12] and X-rays [13] by atoms, the generation of X-rays and radiative recombination [14], etc.

The author proposed in paper [15] a new method for the coherent control of photoexcitation of atoms in a discrete spectrum by a bichromatic radiation field with the 1 : 2 frequency ratio, when the dipole transition in a centrally symmetric system is forbidden in one of the channels by parity selection rules. To overcome this, it was proposed [15] to use an electrostatic field removing the parity selection rules, thereby allowing the coherent control of excitation of an atom in a discrete spectrum at different parities of the number of photons in each of the channels of the process. A substantial advantage of the coherent control of this type is the presence of a new parameter – the electrostatic field strength F_0.

It was shown in [15], in particular, that in the case of destructive interference of the amplitudes of excitation channels, the field strength value F_0 exists at which the total probability of the process proves to be zero, so that the excited transition is 'bleached'. It was proposed in [15] to detect the coherent control by means of a photoluminescence signal at the adjacent electronic transition. Note that the luminescent phase control method was first proposed in [16] to analyse the excitation of an atom in a discrete spectrum by bichromatic radiation with the 1 : 3 frequency ratio.

This paper is devoted to the development and generalisation of the method for coherent control of excitation of atoms. The generalisation consists in the replacement of an electrostatic field removing the parity selection rule by an alternating electric field. In this case, photoluminescence appearing upon such excitation is modulated in time, the type of modulation being substantially dependent on the phase difference of the bichromatic field. Thus, the phase-sensitive electric modulation of luminescence is produced, which represents a new type of coherent control in a bichromatic laser field.

2. Calculation of the photoluminescence signal intensity

To reveal the basic qualitative characteristics of the proposed coherent control, we consider, as in [15], the simplest case of excitation of an alkali metal atom from the ground to the first excited s state (with the zero orbital momentum) by bichromatic radiation of the type

$$\boldsymbol{F}(t) = \boldsymbol{F}_1 \cos(\omega t + \varphi_1) + \boldsymbol{F}_2 \cos(2\omega t + \varphi_2), \qquad (1)$$

where $\boldsymbol{F}_{1,2}$ are the amplitudes of the electric field of monochromatic components of radiation; $\varphi_{1,2}$ are the initial radiation phases at the fundamental and second-harmonic

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frequencies. The fundamental frequency ω is determined from the energy conservation law

$$2\hbar\omega = \Delta E_{\mathrm{fi}} \equiv \hbar\omega_{\mathrm{fi}}, \tag{2}$$

where ΔE_{fi} is the excitation energy; ω_{fi} is the frequency of the excited transition in the atom. Note that the characteristic energy of the transitions of this type in alkali atoms lies in the range from 2.2 to 3.4 eV [17], so that the corresponding fundamental frequency lies in the easily accessible near-IR region.

The first photoexcitation channel of an atom is two-photon absorption of radiation at the fundamental frequency. In the case of the s–s transition under study, this process is allowed by selections rules for a spherically symmetric system. The second photoexcitation channel – one-photon absorption of the second harmonic, is forbidden by parity selection rules [18]. These rules are removed when an electric field, which is assumed spatially homogeneous, is applied. Unlike [15], we will consider below an alternating electric field with a sufficiently low frequency. For simplicity, we assume that this field is harmonic:

$$\boldsymbol{F}_{\mathrm{lf}}(t) = \boldsymbol{F}_0 \cos(\Omega t). \tag{3}$$

The frequency Ω of this alternating field should be low enough in order that the excited system could follow adiabatically variations in the electric field strength. In this case, the calculation of the probability of atom excitation from the initial state $|\mathrm{i}\rangle = |n\mathrm{s}\rangle$ to the final state $|\mathrm{f}\rangle = |(n+1)\mathrm{s}\rangle$ by radiation (1) in the presence of electric field (3) is completely analogous to calculations performed in [15]. Therefore, by using the calculation method described in [15], we can obtain the expression

$$w_{\mathrm{fi}}^{\mathrm{tot}}(\varphi, t) = \frac{4\pi^3}{\hbar^2 c^2} G_{\mathrm{fi}}(\omega) I_1^2 |c_1 + 2r_{01}\cos(\Omega t)\sqrt{\eta}\exp(\mathrm{i}\varphi)c_2|^2 \tag{4}$$

for the $|\mathrm{i}\rangle \rightarrow |\mathrm{f}\rangle$ transition excitation probability per unit time, where $G_{\mathrm{fi}}(\omega)$ is the line shape function of the excited atomic transition; $r_{01} = F_0/F_1$ is the ratio of the amplitudes of low-frequency electric field (3) and the electric field of fundamental radiation; $I_1 = cF_1^2/8\pi$ is the fundamental radiation intensity; $\eta = I_2/I_1$ is the ratio of the intensities of monochromatic components of the field; $\varphi = 2\varphi_1 - \varphi_2$ is the controllable phase shift between monochromatic components of radiation (1) – 'bichromatic phase'. The angle between the vectors $\boldsymbol{F}_0$ and $\boldsymbol{F}_2$ is assumed zero. In (4), $c_1 = c_{\mathrm{fi}}(\omega, -\omega)$ and $c_2 = c_{\mathrm{fi}}(2\omega, 0)$, where $c_{\mathrm{fi}}(\omega, \omega')$ is the scalar part of the tensor of electromagnetic radiation scattering by an atom followed by excitation of the transition under study, which also describes Raman scattering of light by an atom [19]. The parameters $c_{1,2}$ together with the line shape function $G_{\mathrm{fi}}(\omega)$ describe the dynamics of an optical electron during excitation of an atom via the first and second channels.

The above-mentioned adiabaticity condition for the action of an alternating electric field on an atom, which was used to derive expression (4), can be written in the form

$$\Omega \ll A_{\mathrm{f}}, \tag{5}$$

where A_{f} is the excited-state decay probability (per unit time). Because $A_{\mathrm{f}} \approx 10^8\ \mathrm{s}^{-1}$ in the case under study, inequality (5) restricts a rather broad frequency range of the electric field.

Expression (4) was derived assuming that the amplitudes of electric fields involved in the process are much lower than the characteristic atomic field strength ($F_{\mathrm{a}} = 5.14 \times 10^9$ V cm^{-1}), i.e., the interaction of external fields with atoms can be described with the help of the perturbation theory. The perturbation theory can be applied in this case also due to the absence of one-photon resonance at the allowed dipole transition, in the presence of which even relatively weak fields should be considered outside the framework of the perturbation theory [20].

Figure 1 shows the scheme of bichromatic excitation of luminescence from the ground state of a sodium atom. The pump wavelength at the fundamental frequency is $\lambda_1 =$ 777 nm and luminescence wavelengths λ_{fj} and λ_{fi} are 1140 and 590 nm, respectively. We neglect the fine splitting of the intermediate 3p level. Below, we will consider for definiteness luminescence at the 4s $\rightarrow$ 3p transition ($|\mathrm{f}\rangle \rightarrow |\mathrm{j}\rangle$). The luminescence power density emitted at this transition from the unit volume to the unit solid angle is

$$q_{\mathrm{fj}} = N_{\mathrm{f}} A_{\mathrm{fj}} \frac{\hbar\omega_{\mathrm{fj}}}{4\pi}, \tag{6}$$

where N_{f} is the concentration of atoms excited by bichromatic radiation in the $|\mathrm{f}\rangle$ state; A_{fj} is the Einstein coefficient for spontaneous radiation; and $\hbar\omega_{\mathrm{fj}}$ is the transition energy.

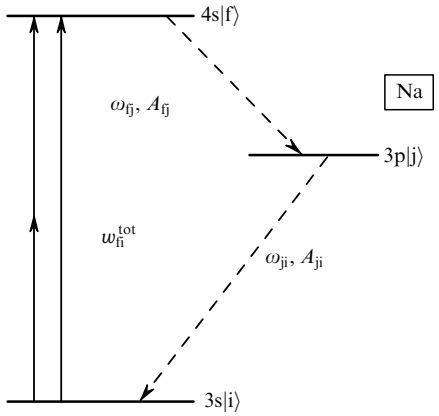

Figure 1. Scheme of bichromatic excitation of luminescence in a sodium atom.

To calculate N_{f}, we assume that luminescence is excited in a quasi-stationary regime. This condition is satisfied if the duration of a bichromatic radiation pulse exceeds the characteristic relaxation times of the excited $|\mathrm{f}\rangle$ and $|\mathrm{j}\rangle$ levels, which are of the order of 10^{-8} s in our case. By using this assumption, we can easily obtain the equality (see details in [16])

$$q_{\mathrm{fj}} = q_{\mathrm{fj}}^{(\mathrm{s})} \frac{T_1 w_{\mathrm{fi}}^{\mathrm{tot}}}{1 + T_1 w_{\mathrm{fi}}^{\mathrm{tot}}}, \tag{7}$$

where

$$q_{\mathrm{fj}}^{(\mathrm{s})} = \frac{\hbar\omega_{\mathrm{fj}}}{4\pi}\frac{N_0}{T_1} \tag{8}$$

is the luminescence power density in the saturation regime;

$$T_1 = A_{\mathrm{fj}}^{-1} + A_{\mathrm{ji}}^{-1} \tag{9}$$

is the relaxation time of the three-level system shown in Fig. 1; $w_{\mathrm{fi}}^{\mathrm{tot}}$ is the bichromatic excitation probability (4); and N_0 is the total concentration of sodium atoms.

Expressions (7)–(9) were obtained by neglecting relaxation caused by resonance energy transfer between sodium atoms. This neglect imposes the restriction $N_0 < N_0^*$ on the atomic concentration. The estimate shows [16] that $N_0^* \approx 10^{13}$ cm^{-3} in our case.

3. Phase-sensitive modulation of luminescence

Let us use the expressions obtained in the previous section to analyse the modulation of photoluminescence excited by bichromatic field (1), which is caused by low-frequency electric field (1) acting on the atomic transition.

One can see from expression (4) that the dependence of the total excitation probability on the relative phase difference of monochromatic components is most distinct if the terms under the modulus sign describing the amplitudes of two channels have equal moduli. This condition determined the optimal value of the parameter η (the intensity ratio of the second harmonic and fundamental radiation). In the case of electrostatic field (3) ($\Omega = 0$), the optimal parameter η is described by the expression

$$\tilde{\eta}_{\mathrm{opt}} = \left|\frac{c_1}{2c_2 r_{01}}\right|^2. \tag{10}$$

Here, the tilde means that this equality is obtained for a constant electric field. For excitation of a sodium atom from the ground state to the first excited s state, when $c_1 \simeq 715$ and $c_2 = 8.4$, expression (10) can be rewritten in the form

$$\tilde{\eta}_{\mathrm{opt}} = \frac{1800}{r_{01}^2}. \tag{11}$$

This gives the dependence of the optimal intensity ratio of monochromatic radiation components (1) on the parameter $r_{01} = F_0/F_1$.

It follows from expression (7) for the photoluminescence power density that far from saturation, when the inequality

$$w_{\mathrm{fi}}^{\mathrm{tot}} \ll T_1^{-1} \tag{12}$$

is fulfilled, the phase dependence of photoluminescence coincides with that of the excitation probability (4). The estimated fundamental radiation intensity at which the first channel is saturated upon two-photon excitation of the sodium atom (see Fig. 1) is

$$I_1^{\mathrm{sat}}[\mathrm{Na}] \approx 1.6 \times 10^5 \text{ W cm}^{-2}. \tag{13}$$

If $I_1 \ll I_1^{\mathrm{sat}}$ (unsaturated regime), expression (7) takes the form

$$q_{\mathrm{fj}} \simeq \frac{\hbar\omega_{\mathrm{fj}}}{4\pi} N_0 w_{\mathrm{fi}}^{\mathrm{tot}}. \tag{14}$$

By substituting expression (14) for the excitation probability into (14) and separating explicitly the dependence on the bichromatic phase $\varphi = 2\varphi_1 - \varphi_2$ and the phase $\chi = \Omega t$ of the low-frequency electric field, we obtain the luminescence power density in the unsaturated regime

$$q_{\mathrm{fj}}^{\mathrm{nsat}}(\chi, \varphi, p) = q_{\mathrm{fj}}^{(0)}\left(1 + p^2\cos^2\chi + 2p\cos\chi\cos\varphi\right), \tag{15}$$

where

$$p = \frac{2c_2\sqrt{\eta}r_{01}}{c_1} = \left(\frac{\eta}{\tilde{\eta}_{\mathrm{opt}}}\right)^{1/2} \tag{16}$$

is the low-frequency modulation parameter, which is proportional to the amplitude ratio of one-photon absorption of the second harmonic (for $\Omega = 0$) to two-photon absorption of fundamental radiation. The dimensional factor $q_{\mathrm{fj}}^{(0)}$ in the right-hand side of (15) is the luminescence power density in the absence of the second harmonic. From (15), we obtain the expression for the phase modulation depth (with respect to the bichromatic phase $\varphi = 2\varphi_1 - \varphi_2$):

$$\zeta(p, \chi) = \frac{4p\cos\chi}{1 + p^2\cos^2\chi}. \tag{17}$$

This parameter determines the efficiency of coherent control of the process under study [15]. The maximum efficiency is achieved for $p = 1$ and $\chi = \pi n$, when $|\zeta| = 2$.

Let us introduce the coefficient of luminescence modulation by a low-frequency electric field (3) (phase $\chi = \Omega t$)

$$k_{\mathrm{mod}} = 2\,\frac{(q_{\mathrm{fj}})_{\max} - (q_{\mathrm{fj}})_{\min}}{(q_{\mathrm{fj}})_{\max} + (q_{\mathrm{fj}})_{\min}}. \tag{18}$$

This coefficient is a function of the bichromatic phase $\varphi = 2\varphi_1 - \varphi_2$ and parameter p [see expression (16)]. Far from saturation, when inequality (12) is fulfilled, this function can be written in the form

$$k_{\mathrm{mod}}^{\mathrm{nsat}}(\varphi, p) = 2\,\frac{(|p| + |\cos\varphi|)^2}{2\sin^2\varphi + (|p| + |\cos\varphi|)^2}. \tag{19}$$

It follows from this equality that the maximum modulation coefficient is achieved for $\varphi = \pi n$. In this case, $k_{\mathrm{mod}}^{\mathrm{nsat}} = 2$ for all values of the parameter p. The coefficient has the minimal value $k_{\min}^{\mathrm{nsat}} = 2p^2/(2 + p^2)$ at $\varphi = n\pi/2$, when the interference term in the expression for the excitation probability is zero.

In the unsaturated regime, the ratio of the maximum and minimum modulation coefficients is $k_{\max}^{\mathrm{nsat}}/k_{\min}^{\mathrm{nsat}} = 1 + 2/p^2$. This ratio is large for small values of p and tends to unity at large values of p.

The modulation of the photoluminescence power density calculated from (15) for three values of the bichromatic phase ($\varphi = \pi/3,\ \pi/2,\ \pi$) and the parameters $\eta = 1800$, $I_1 = 10^4$ Wcm$^{-2} \ll I_1^{\mathrm{sat}}$, $N_0 = 10^{12}$ cm^{-3}, and $r_{01} = 1$ ($F_0 \approx 10^3$ V cm^{-1}) is shown in Fig. 2. As expected [see expression (19)], the minimal modulation $k_{\mathrm{mod}} = 2/3$ takes place when the bichromatic phase is $\varphi = \pi/2$, and the maximum modulation $k_{\mathrm{mod}} = 2$ is obtained when $\varphi = \pi$. When $\varphi = \pi/3$ ($k_{\mathrm{mod}} = 6/5$), additional maxima and minima appear on the photoluminescence modulation curve. Figure 2 demonstrates the 'top–bottom' asymmetry of

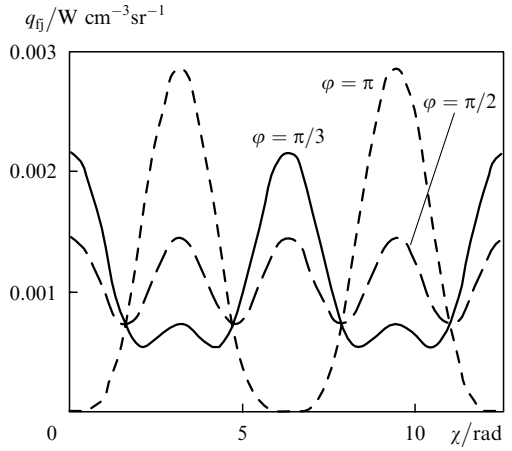

Figure 2. Modulation of the photoluminescence power density upon monochromatic excitation of a sodium atom in a low-frequency electric field in the unsaturated regime ($I_1 = 10^4$ W cm$^{-2} \ll I_1^{sat}$) for different values of the bichromatic phase φ and $p = 1$.

modulation curves when the interference between channels is not zero: $\varphi \neq \pi/2$.

The dependence of the type of low-frequency modulation of photoluminescence in the unsaturated regime [see condition (12)] on the parameter $p = (\eta/\tilde{\eta}_{opt})^{1/2}$ for a fixed bichromatic phase $\varphi = \pi/3$ is shown in Fig. 3. One can see that, when the parameter p is small, the modulation curve has 'flat' parts and the modulation coefficient is low. Our calculations show that, as the parameter p further decreases, the modulation curves becomes symmetric and the modulation depth decreases down to $k_{mod} = 2/7$. As the parameter p increases, the smaller maximum increases for $\chi = \pi$, 3π, and the curve becomes symmetric in the limit $p \to \infty$.

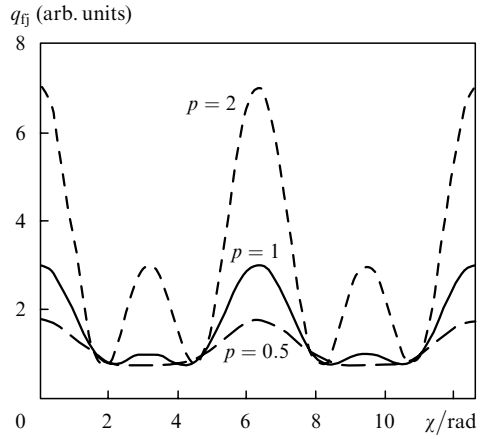

Figure 3. Dependences of the type of low-frequency modulation of photoluminescence on the modulation parameter p in the unsaturated regime for the fixed bichromatic phase $\varphi = \pi/3$.

In the saturation regime, when the inequality opposite to condition (12) is fulfilled, the dependence of the luminescence power density on the atom excitation probability, as follows from (7), becomes weaker, except the values of the phase $\chi = \Omega t$ at which the excitation probability of the atomic transition is low. Figure 4 shows the dependences of the luminescence signal on the phase of a low-frequency electric field for the same values of the bichromatic phase and parameter p as in Fig. 2, but for higher fundamental radiation intensity ($I_1 = 5 \times 10^5$ W cm$^{-2} > I_1^{sat}$). It follows from Fig. 4 that the high modulation coefficient $k_{mod} = 2$ is preserved for $\varphi = \pi$, when interference between channels in the excitation probability is maximal, and the values of the phase χ exist at which this probability is zero. In this case, the hole width in the corresponding curve decreases and the phase dependence is weak far from values $\chi = 2\pi n$. For $\varphi = \pi/3$ and $\pi/2$, when $w_{fi}^{tot} \neq 0$ for all phases χ of the low-frequency field, the modulation coefficient drastically decreases with increasing the laser radiation intensity due to saturation. In the limit $I_1 \gg I_1^{sat}$, the modulation coefficient tends to zero for $\varphi \neq \pi n$, while for$\varphi = \pi n$ a hole in the corresponding curve becomes infinitely narrow. Thus, in the saturation regime the type of modulation of the luminescence signal as a function of the phase of a low-frequency electric field (3) substantially changes compared to the case of a relatively low intensity of the laser field [when inequality (12) is fulfilled].

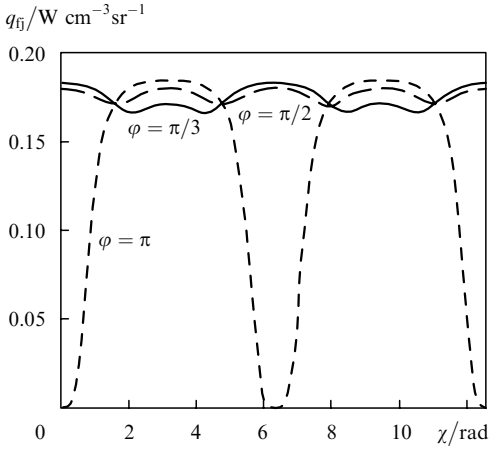

Figure 4. Dependences of the photoluminescence density power on the phase φ of a low-frequency electric field in the saturation regime for $I_1 = 5 \times 10^5$ W cm$^{-2} > I_1^{sat}$.

4. Conclusions

The modulation of photoluminescence excited in an atom by bichromatic radiation with the frequency ratio 1 : 2 has been calculated and analysed. The modulation was produced by an alternating low-frequency field removing the parity selection rule for one of the excitation channels.

The calculation was performed for an alkali metal atom excited from the ground to the first s state, when the two-photon transition is allowed and one-photon transition is forbidden for a spherically symmetric system. The expression was obtained for the power density of luminescence excited by bichromatic laser radiation, which takes into account the saturation of the excited transition. The atomic dynamics is described in this expression by the scalar part of the tensor of light scattering with simultaneous excitation of an atom, which also determines the Raman scattering cross section.

It was shown that the type of luminescence modulation caused by the alternating electric field substantially depends on the bichromatic phase $\varphi = 2\varphi_1 - \varphi_2$, the modulation

parameter p (16), and the intensity of monochromatic components of pump radiation. The highest modulation coefficient is obtained at moderate intensities of the laser field [without saturation of the excited atomic transition, when condition (12) is fulfilled] and the values of the bichromatic phase and parameter p at which interference between the channels upon excitation of an atom is maximal. In the saturation regime, the modulation coefficient drastically decreases, except narrow regions of variation in the low-frequency phase $\chi = \Omega t$, where the probability of the process is close to zero due to the destructive interference between excitation channels.

It was found that the parameters of the problem required for observing luminescence modulation such as the frequencies and intensities of monochromatic components of radiation and the strength of a low-frequency electric field lie in the easily accessible region of values.

The process studied in the paper can be considered as a new type of the phase coherent control of excitation of atoms in a discrete energy spectrum, whose practical application is facilitated by the luminescent detection method.

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