

Radiative processes in a bichromatic laser field with multiple frequencies

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Contents

1. Introduction	1131
2. Atoms and ions in a bichromatic laser field	1132
2.1. Coherent phase control at bound-bound transitions	
2.2. Photoionisation of atoms and photodestruction of negative ions by bichromatic radiation	
2.3. Phase control of elementary processes involving a laser field	
3. Molecular processes in a bichromatic laser field	1139
3.1. Photodissociation and photoionisation of molecules by bichromatic radiation	
3.2. Coherent phase control of the vibrational-rotational excitation of molecules	
4. Coherent control of photoeffect in a bichromatic field	1141
4.1. External photoeffect in a bichromatic laser field	
4.2. Internal photoeffect upon bichromatic excitation of solids	
4.3. Bichromatic control of the spin of charge carriers in semiconductors	
5. Conclusions	1145
6. References	1145

Abstract. A review of experimental and theoretical studies on radiative processes induced by a bichromatic (two-frequency) laser radiation in atoms, molecules, and solids is presented. The optical excitation of matter is considered at transitions of different types: bound–bound, bound–free, and free–free. A special attention is devoted to the possibility of the coherent control of the basic characteristics of radiative phenomena by varying the relative phase of monochromatic components of radiation. It is shown that this method opens up new possibilities for controlling photoinduced processes.

Keywords: bichromatic radiation, coherent phase control, quantum interference.

1. Introduction

This review is devoted to coherent processes induced in matter by coherent radiation representing the superposition of a laser field at the fundamental harmonic ω and its n th harmonic. The expression for the electric field vector of this radiation has the form

$$F(t) = e_1 F_{01} \cos(\omega t + \varphi_1) + e_n F_{0n} \cos(\omega_n t + \varphi_n), \quad (1)$$

where $\omega_n = n\omega$ is the frequency of the n th harmonic of laser radiation (n is a natural number greater than unity); $F_{01,0n}$ are the amplitudes of monochromatic components; $e_{1,n}$ are the unit polarisation vectors of the electric field; and $\varphi_{1,n}$ are the initial phases.

Photoinduced processes considered here are based on the quantum interference of the amplitudes of transitions between the states of a system induced by bichromatic laser field (1). The nature of this interference can be simply explained by the example of interaction of a quantum system with electromagnetic radiation of a moderate intensity, when the process can be described by using the perturbation theory. Let us assume that the quantum system can undergo transitions from the initial state $|i\rangle$ to the final state $|f\rangle$ via two channels related to the action of the monochromatic components of field (1) on the system. To these channels, two quantum-mechanical amplitudes A_1 and A_2 correspond (Fig. 1) which are related to absorption (or emission) of $m_{1,2}^{(1)}$ field photons at the fundamental harmonic and $m_{1,2}^{(n)}$ photons at the n th harmonic.

In this case, the law of conservation of energy should be fulfilled, which is expressed by the equalities

$$m_1^{(1)} \hbar\omega + m_1^{(n)} \hbar\omega_n = E_f - E_i \quad (2)$$

for the first channel and

$$m_2^{(1)} \hbar\omega + m_2^{(n)} \hbar\omega_n = E_f - E_i \quad (3)$$

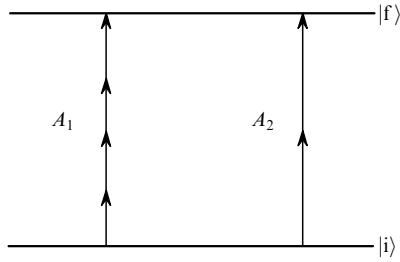


Figure 1. Two transition channels of a system from the $|i\rangle$ state to the $|f\rangle$ state with amplitudes A_1 and A_2 excited by bichromatic radiation with multiple frequencies.

for the second channel, where $m_1^{(1)} \neq m_2^{(1)}$, $m_1^{(n)} \neq m_2^{(n)}$ [$m_{1,2}^{(1),(n)}$ are integers, which can be also zero and negative]; $E_{i,f}$ are the energies of the initial and final states. According to the general principles of quantum mechanics, the total probability of the transition is proportional to the square of the modulus of the total amplitude of the process

$$W_{\text{tot}} \propto |A_1 + A_2|^2 = |A_1|^2 + |A_2|^2 + 2|A_1||A_2|\cos(\varphi_n - n\varphi_1 + \Delta\varphi_i), \quad (4)$$

where $\Delta\varphi_i$ is the internal phase shift. The first term in the right-hand side of equality (4) describes the transition probability in the first channel, the second one – the transition probability in the second channel, and the third one describes their quantum interference. It is important that the interference term in expression (4) for the total probability of the process depends on the combination of the initial phases of monochromatic components of field (1): $\Delta\varphi = \varphi_n - n\varphi_1$ (which we call below the bichromatic or relative phase), so that by varying $\Delta\varphi$, we can change the probability of the $|i\rangle \rightarrow |f\rangle$ transition. Such a control of the photoprocess probability is called the coherent phase control in the bichromatic radiation field.

In recent years coherent processes in bichromatic fields of another type are widely used when the frequencies of monochromatic components are not multiples but, on the contrary, close to each other. These phenomena include, for example, the coherent population trapping and related electromagnetically induced transparency. Effects of this type and the coherent control in a field with multiple frequencies considered here have a common nature and their own specific features.

Let us consider briefly the first studies on radiative processes in a bichromatic laser field with multiple frequencies performed in our country at the stage of investigations, when the relation between different processes of this type was not clear.

The idea of coherent control was first proposed by Russian scientists Afanas'ev and Manykin in paper [1] in which the possibility of 'bleaching' of materials irradiated by a superposition of electromagnetic waves under the conditions of multiquantum resonance was studied. The authors of paper [1] considered this effect by studying the third harmonic generation of laser radiation in the case of a resonance at the doubled frequency: $2\omega = \omega_0$, where ω_0 is the eigenfrequency of the medium in which radiation propagates. It was pointed out in [1] that the

medium is 'bleached' due to the mutual compensation of the quantum-mechanical amplitudes of channels rather than due to the equating of populations of the upper and lower levels, as in the case of saturation.

One of the forms of coherent phase control is the coherent photogalvanic effect predicted in [2]. This effect was calculated by Baskin and Entin [3, 4] in the case of absorption of radiation by free carriers and in the region of impurity-band transitions in semiconductors. The effect consists in the appearance of a stationary current induced by a superposition of coherent electromagnetic waves with multiple frequencies. Unlike the volume photogalvanic effect, the coherent effect can be observed both in a centrally symmetric crystal and an isotropic medium. The characteristic feature of this effect is the dependence of the photocurrent on the bichromatic phase $\Delta\varphi$.

The coherent photogalvanic effect was considered in [5] as the mechanism of photoinduced second harmonic generation in optical fibres, in which, as is known, the second-order nonlinearity is absent before optical excitation. It was shown [6] that a prolonged exposure of an optical fibre to laser radiation induces the quadratic optical susceptibility $\chi^{(2)}$ in the fibre and thus second harmonic generation becomes possible. It was assumed in [5] that $\chi^{(2)}$ is induced by an electric field appearing due to the coherent photogalvanic effect. The proposed mechanism of photoinduced second harmonic generation adequately explained experimental data, which seemed contradictory when other models were used.

One of the first experimental papers, which initiated the systematic study of photoprocesses in a bichromatic field with multiple frequencies, was the work of Zel'dovich and Chudinov [7], where a photocurrent from a photomultiplier cathode exposed to radiation at the fundamental and second harmonic was measured. In this case, the quantum interference of the amplitudes of one- and two-quantum photoeffects took place. The bichromatic phase difference $\Delta\varphi = \varphi_2 - 2\varphi_1$ was changed by rotating a plane-parallel glass plate. The phase modulation depth of the photocurrent was 10 %, which convincingly demonstrated the presence of quantum interference upon external photoeffect. This question is discussed in detail in paper [8].

Studies on the coherent control of radiative processes in a bichromatic laser field can be divided into three groups: the phase control of atomic processes, bichromatic control of molecular dynamics, and coherent control of photoeffect from the surface and inside solids. The analysis of experimental and theoretical papers in these three directions of investigations is the subject of this review.

2. Atoms and ions in a bichromatic laser field

2.1 Coherent phase control at bound–bound transitions

Upon excitation of bound–bound transitions by bichromatic radiation with multiple frequencies, the quantum interference of the channel amplitudes A_1 and A_2 is considerably restricted by the selection rules. Indeed, the parity of the number of photons in each channel should be the same, otherwise one of the amplitudes of the process is zero due to the parity selection rule. Therefore, interference is absent, for example, upon excitation of a ground-state atom by bichromatic radiation with the frequency ratio 1:2. In the case of a bichromatic field consisting of

radiation at the fundamental frequency and its third harmonic, the coherent phase control of atomic excitation in the discrete spectrum is possible. Thus, if $3\hbar\omega = E_f - E_i$, the quantum interference of the one-photon absorption of the third harmonic and three-photon absorption at the fundamental frequency takes place. Such a situation appears, for example, upon multiphoton ionisation of atoms in the presence of an intermediate resonance state [9], when the induced third harmonic and radiation at the fundamental frequency cause the destructive interference during excitation of the resonance level [10]. As a result, the excitation of the resonance level is suppressed. In this case, the third harmonic generation and photoinduced process occur in the same volume, so that the phase difference between monochromatic components of radiation cannot be externally controlled. On the contrary, the phase shift of the generated third harmonic with respect to the fundamental radiation is fixed and equal to 180° , which causes the destructive interference upon excitation of the resonance level.

The coherent control of bichromatic excitation of an atom at a bound–bound transition upon external variation of the relative phase was first demonstrated in [11]. The $6s\ ^1S_0 \rightarrow 6p\ ^1P_1$ transition in the mercury atom was excited by the fundamental radiation at 554 nm and its third harmonic. In this case, the quantum interference of the amplitude of one-photon absorption at the third-harmonic frequency and the amplitude of three-photon absorption at the fundamental frequency took place, i.e. the ‘1 + 3’ phase control scheme was realised. The expression for the total probability of bichromatic excitation of an atom taking into account two channels of the process, obtained within the framework of the perturbation theory, has the form

$$W_{\text{tot}} = \frac{2\pi}{\hbar^2} \left| \mu F_{\text{uv}} e^{i\varphi_3} + \mu^{(3)} (F_{\text{vis}} e^{i\varphi_1})^3 \right|^2 g(\omega_0 - 3\omega), \quad (5)$$

where μ and $\mu^{(3)}$ are the electric dipole moments for one- and three-photon transitions; F_{vis} and F_{uv} are the electric field strengths of fundamental and third-harmonic radiation, respectively; and $g(\omega_0 - 3\omega)$ is the spectral line shape. One can see from (5) that the square of the modulus in the right-hand side of the equality gives the interference term containing the factor $\cos(\varphi_3 - 3\varphi_1)$, which describes the phase dependence of the probability of the process.

The dependence of the total excitation rate of the $6s\ ^1S_0 \rightarrow 6p\ ^1P_1$ transition on the bichromatic phase $\Delta\varphi = \varphi_3 - 3\varphi_1$ was detected in experiments [11] by the photocurrent appearing due to two-photon ionisation of the $6p\ ^1P_1$ state of the mercury atom by fundamental radiation. The fundamental pulse energy was 4.5 mJ and its duration was 15 ns. The third harmonic was generated by a focused fundamental radiation in chamber (1) (Fig. 2) containing mercury vapour at a relatively high pressure of 100 mTorr. The bichromatic phase was varied by transmitting fundamental radiation and its third harmonic through cell (2) with argon, whose pressure was varied from 0 to 38 Torr, according to the expression

$$\Delta\varphi = \frac{6\pi l \Delta\rho}{\lambda_1} [n(\lambda_1) - n(\lambda_3)], \quad (6)$$

where l is the laser beam path in a cell filled with argon; $\Delta\rho$ is the change in the argon density; and $n(\lambda_j)$ is the refractive

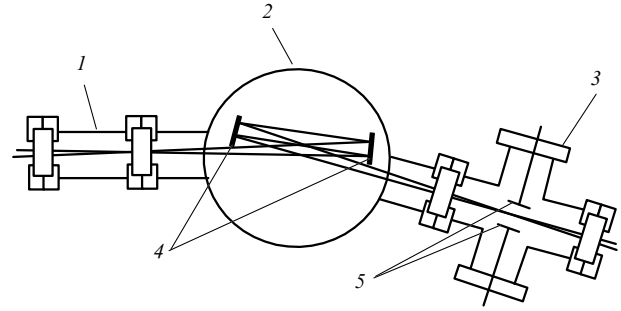


Figure 2. Scheme of the experimental setup [11]: (1) high-pressure chamber with mercury vapour; (2) variable-pressure chamber with argon; (3) low-pressure detection chamber with mercury vapour; (4) focusing spherical aluminium-coated mirrors; (5) collector platinum plates.

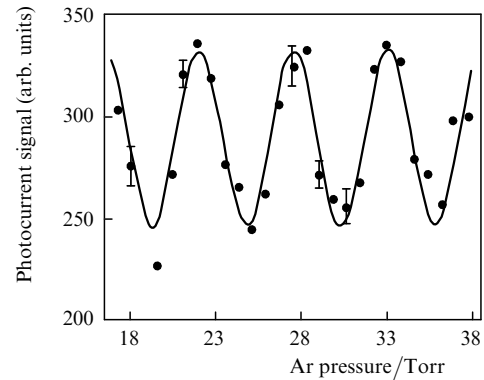


Figure 3. Photocurrent signal recorded in experiments [11] upon bichromatic excitation of mercury vapour as a function of argon pressure. Circles are experimental data; the solid curve is the best approximation.

index of argon at the wavelength λ_j . The photocurrent signal appeared after focusing bichromatic radiation with the controllable phase difference $\Delta\varphi = \varphi_3 - 3\varphi_1$ by mirrors (4) in second chamber (3) with mercury vapour at a low pressure of 2–3 mTorr. A low pressure in the detection chamber was required to reduce the contribution of the third harmonic generated in it. The photocurrent was detected with a collector consisting of a pair of platinum plates (5) across which a bias voltage was applied. The phase modulation depth of the photocurrent detected in experiments [11], which characterises the efficiency of coherent phase control, was 15%, while the maximum possible calculated values was 30% (Fig. 3).

In [12], the coherent control was achieved by using another scheme for exciting atoms. The quantum interference between different channels of the process was observed by exciting krypton and xenon atoms from the ground to the excited state ($5p[5/2]_2$ for krypton and $4f[3/2]_2$ for xenon) by fundamental and third-harmonic radiation. The excitation probability was detected by the photocurrent caused by the one-photon ionisation of the excited state of the atom by fundamental radiation. The phase dependence of the process appeared due to the quantum interference of two excitation channels: four-photon absorption at the fundamental frequency and absorption of one photon at the fundamental frequency and one photon at the

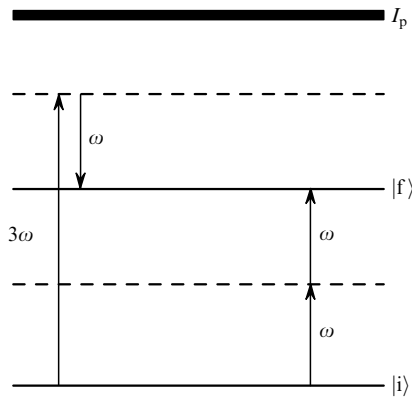


Figure 4. Scheme of the coherent control of excitation of atomic states due to the quantum interference of two-atomic processes (I_p is the ionisation potential of an atom).

third-harmonic frequency, i.e. the ‘4 + 2’ coherent control scheme was realised. In this case, due to a high nonlinearity of the process, a higher value of the modulation coefficient was obtained than in [11].

The coherent ‘2 + 2’ control of bichromatic excitation of atoms was considered in pioneering paper [1] under the condition of two-photon resonance upon third harmonic generation. In this case, the Raman scattering amplitude of the third harmonic induced by fundamental radiation interferes with the amplitude of two-photon absorption of fundamental radiation (Fig. 4). Thus, the quantum interference of processes involving the same number of photons takes place.

The probability of bichromatic photoexcitation of alkali atoms due to quantum interference of two-photon processes of type considered in [1] was calculated in [13] by using the perturbation theory. The excitation of the $ns \rightarrow (n+1)s$ transition was considered, where ns is the ground state of the alkali atom (n is the principal quantum number). It was shown that the optimal intensity ratio $\eta = I_3/I_1$ of the third and first harmonics at which the phase modulation depth of excitation is maximal is determined by the expression

$$\eta_{\text{opt}} = \left[\frac{c_{\text{if}}(\omega, -\omega)}{c_{\text{if}}(3\omega, \omega)} \right]^2, \quad (7)$$

where c_{if} is the scalar part of the tensor of photon scattering by an atom accompanied by excitation of the latter. The specific feature of this phase control scheme is the independence of optimal ratio (7) on the intensity of monochromatic components of radiation.

In most experimental papers on the phase control of atomic excitation, the effect was detected by the photocurrent, which requires the ionisation of the excited level in the case of bound–bound transitions. In [14], another, luminescence method was proposed for detecting the phase control upon bichromatic excitation of atoms in the discrete spectrum by using the ‘2 + 2’ scheme (Fig. 4). The phase dependence of the excitation probability of the $ns \rightarrow (n+1)s$ transition in an alkali atom is determined in this method by the photoluminescence signal at the $(n+1)s \rightarrow np$ or $np \rightarrow ns$ transition.

We considered above the bichromatic excitation of atoms in the discrete spectrum. It is interesting to study

the features of coherent control upon the transition of an atomic electron from the bound to autoionisation state. Such an experiment was performed in [15]. The phase control of excitation of a calcium atom to the $4p7s[1/2]_1^0$ autoionisation state was observed due to interference of two channels: three-photon absorption of fundamental radiation and one-photon absorption of the third harmonic (the ‘3 + 1’ phase control scheme). Pumping was performed by 435-nm, 5-mJ, 10-ns pulses from a tunable dye laser with the spectral linewidth of 0.1 cm^{-1} . The phase modulation depth was $\sim 43\%$. The authors of [15] pointed out that their paper was the first step on the way to achieve the control of the spectral shape of the autoionisation resonance by varying the bichromatic phase. Another important result of their experiments is the demonstration of the phase control without focusing laser radiation, which is important for applications, for example, for the coherent control of photochemical reactions.

In [16], a new method of the coherent phase control of excitation of atoms in the discrete spectrum by bichromatic radiation with the frequency ratio 1:2 was analysed when the amplitudes of two-photon absorption at the fundamental frequency and one-photon absorption at the second-harmonic frequency interfere. As mentioned above, in the case of a spherically symmetric system, one of the above-mentioned channels is forbidden by the dipole selection rules. To overcome this difficulty, it was proposed in [16] to use a stationary homogeneous electric field mixing states of different parities, so that the phase control of excitation proves to be possible. The calculation was performed under conditions of the applicability of the perturbation theory for the interaction of an electromagnetic field with a valence electron. The excitation of the $s \rightarrow s$ transition in an alkali metal atom was considered, when the one-photon process is forbidden by the selection rules in the absence of a stationary electric field. It was shown that the condition of the equality of amplitudes of one- and two-photon absorption, when the phase control efficiency is maximal, is given by the expression

$$\left(|\cos \chi| \sqrt{\eta} r_{01} \right)_{\text{opt}} = \left| \frac{c_{\text{if}}(\omega, -\omega)}{2c_{\text{if}}(2\omega, 0)} \right|, \quad (8)$$

where $r_{01} = F_0/F_1$ is the ratio of the stationary electric field strength to the amplitude of the electric field of fundamental radiation; $\eta = I_2/I_1$ is the intensity ratio of monochromatic radiation components; and χ is the angle between the vectors of the stationary electric field and electric field of the second harmonic. It was also shown in [16] that for the phase difference $\Delta\varphi = \pi$ corresponding to the destructive interference between channels there exists the electric field strength at which the excited atomic transition is ‘bleached’ (Fig. 5). One can see from Fig. 5 that the ‘bleaching’ electric field strength in the case under study is rather high (10^4 V cm^{-1}). This figure also presents the dependence of the process probability on the strength F_0 in the case of constructive interference for $\Delta\varphi = 2\pi$ [curve (2)] and in the absence of interference between channels for $\Delta\varphi = \pi/2$ [curve (3)].

The method for coherent control of bichromatic excitation of atoms proposed in [16] was generalised in [17] to the case when the parity selection rule for one-photon absorption is removed in an alternating electric field. In this case, the basic features of the phenomenon are

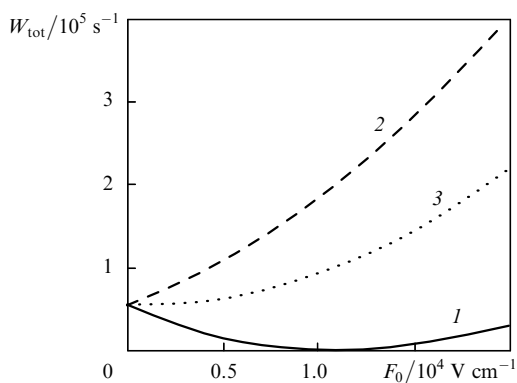


Figure 5. Probability of excitation W_{tot} of the $3s \rightarrow 4s$ transition in a sodium atom by bichromatic radiation with the frequency ratio 1:2 as a function of the stationary electric field strength F_0 for the bichromatic phase $\Delta\varphi = \varphi_2 - 2\varphi_1 = \pi$ (1), 2π (2), and $\pi/2$ (3) and $I_1 = 10^4 \text{ W cm}^{-2}$, $\eta = I_2/I_1 = 100$, and $\varkappa = 0$ [16].

preserved if the electric field frequency Ω satisfies the inequality $\Omega \ll A_f$, where A_f is the probability (per unit time) of the excited-state decay. Luminescence appearing upon such bichromatic excitation, whose modulation in time is determined by the electric field strength and the relative phase of bichromatic excitation, was analysed in [17].

2.2 Photoionisation of atoms and photodestruction of negative ions by bichromatic radiation

The coherent control of atomic processes upon bichromatic excitation of bound-bound transitions has its specific features caused by the degeneracy of the final state of the system over the propagation direction of the momentum of the ionised electron. This degeneracy permits the phase control of the angular distribution of photocurrent. In this case, the symmetry of the radiation pattern is determined by the number of photons in each channel involved in the process. If these numbers of photons have different parities, the angular distribution of photoelectrons is, generally speaking, asymmetric, the asymmetry degree being determined by the bichromatic phase of radiation. If the parity of the numbers of photons is the same, the angular photoionisation diagram is symmetric. Thus, unlike the situation considered in section 2.1, the coherent control of photoionisation of atoms is possible under the action of bichromatic radiation with the frequency ratio determined by the numbers of different parities.

The first studies on bichromatic photoionisation of atoms were performed in strong laser fields of intensity $I \geq 10^{13} \text{ W cm}^{-2}$. At such intensities, photoionisation has the above-threshold nature, i.e. the most probable number of absorbed photons exceeds the threshold value. As a result, the additional photosensitive parameter appears – the shape of the photoelectron spectrum. Coherent control of this type was observed in [18] upon ionisation of krypton atoms by a coherent mixture of the fundamental radiation of a neodymium laser at 1064 nm and its second harmonic. The spectrum of photoelectrons was recorded in the above-threshold ionisation regime for different values of the bichromatic phase. It was shown that the intensity ratio of the second photoelectron peak at 2 eV and the first peak at 1 eV depends periodically on the bichromatic phase with the modulation depth $\sim 45\%$, the energy of the peaks being invariable. It was pointed out in [18] that the ratio of

photoelectron maxima weakly depends on fluctuations of the laser radiation, so that the modulation is mainly determined by the phase ratio of monochromatic components of radiation.

The experimental demonstration [18] of the coherent control of bichromatic ionisation of an atom in a strong laser field stimulated the theoretical study of this problem. The corresponding analysis was performed in [19] for a hydrogen atom in the case of monochromatic components of the same intensity with the frequency ratio 1:2. The ionisation rate, the angular distribution of photoelectrons and above-threshold ionisation spectra were calculated for different bichromatic phases and different radiation intensities by solving numerically the temporal Schrödinger equation. In addition, variations in the phase dependence of these parameters on passing from fundamental radiation at 1064 nm to shorter wavelengths 532 and 355 nm were investigated. It was shown that in the long-wavelength case (at 1064 nm), tunnel photoionisation takes place when the photoionisation probability is determined by the peak value of the electric field strength of bichromatic radiation. In turn, this peak value depends on the relative phase determining the time dependence of the total electric field.

Thus, phase effects in the tunnel ionisation of atoms by bichromatic radiation can be qualitatively explained mainly by the interference of the monochromatic components of the radiation field rather than by the interference of quantum-mechanical amplitudes. The calculation [19] showed that the top-bottom asymmetry (in the case of the vertical orientation of the field polarisation vector) in the angular distribution of photoelectrons depends considerably on the bichromatic phase and increases with increasing the radiation intensity. The results of calculations also confirmed the phase dependence of the shape of the above-threshold photoelectron spectrum observed in experiments [18]. The complicated physical picture of bichromatic ionisation of an atom analysed in [19] is related to a high intensity of laser radiation, when ionisation cannot be described by the perturbation theory. For laser fields with a relatively low intensity ($I < 10^{10} \text{ W cm}^{-2}$), when the perturbation theory can be applied, the situation is simplified, and coherent control can be described in terms of the interference of quantum-mechanical amplitudes (see Introduction).

The photoionisation of sodium atoms by the bichromatic field of a neodymium laser with the frequency ratio 1:2 was performed by Russian researchers in [20]. Sodium atoms were preliminarily excited by radiation from a dye laser at 589 nm and a LiF:F_2^- colour centre laser at 1140 nm via the intermediate $3p$ state to the $4s$ level, which then was ionised by the bichromatic field of the neodymium laser. The phase dependence of bichromatic ionisation appeared due to the quantum interference of one-photon absorption of the second harmonic and two-photon absorption of the fundamental radiation. The phase modulation depth of the probability of atom ionisation in the given direction measured in [20] was 15%. The qualitative explanation of the polar asymmetry proposed in [20] was based on the analysis of the angular dependence of the wave function of an ionised electron, which is determined by the coherent superposition of the s , p , and d waves. This superposition appears due to excitation of the initial s state during one- and two-photon processes and describes the symmetric angular distribution of the atomic electron escape probability.

The asymmetry of the angular distribution of photoelectrons appearing due to the quantum interference of free-free transitions was also measured in [21] where rubidium atoms were ionised by bichromatic radiation of a moderate intensity. Ionisation was performed by 560-nm, 10-ns pulses from a dye laser and its second harmonic. Both laser beams with the same polarisation intersected the beam of ionised rubidium atoms at an angle of 108° . A photoelectron current signal was taken from four detectors detecting electrons with momentum vectors making angles $\Theta = 0, 45^\circ, 90^\circ$, and 180° with the radiation polarisation vector. The maximum modulation depth (about 50%) was observed for the observation angles $\Theta = 0$ and 45° , the corresponding phase dependences being shifted with respect to each other by π . The value of this shift is determined by the cross sections for excitation of the initial state of rubidium atoms to the 2S -, $^2D_{5/2}$ - и $^2D_{3/2}$ states of the continuous spectrum and by the relative phases of their wave functions. This relation can be used for determining parameters of the wave functions of the continuous spectrum from the phase dependences of the angular distribution of photocurrent upon bichromatic ionisation of atoms.

A simple theoretical consideration of this problem based on the perturbation theory is presented in [21]. An important result obtained in this paper is that upon ionisation of an atom by a bichromatic field with the frequency ratio 1:2, the phase modulation of the ionisation probability takes place only for a fixed escape angle of a photoelectron. The integrated (over the electron escape angle) photoionisation probability is independent of the bichromatic phase. The latter circumstance follows from the orthogonality of the wave functions of the continuous-spectrum states to which an atomic electron is excited due to one- or two-photon absorption.

The situation is different if the parity of the number of photons in each of the channels of the process is the same. In this case, the coherent control of the angle-integrated escape of photoelectrons is possible, but at the same time the asymmetry of the angular distribution of photoelectrons is absent. This case was studied experimentally in [22] where sodium atoms were ionised by bichromatic radiation with the frequency ratio 1:3 when the phase control is caused by the interference of the amplitudes of one- and three-photon absorption. The ionising radiation was a coherent mixture of the fundamental radiation of a neodymium laser at 1064 nm and its third harmonic. Each point of the phase dependence of the photoionisation signal was obtained by averaging the data over 1000 pulses of ionising radiation. The phase modulation depth achieved in experiments [22] by optimising the interaction and detection conditions was 84% (the highest value obtained in such experiments).

Coherent phase effects appearing upon bichromatic '1 + 3' ionisation of the ground state of the hydrogen atom are described in detail in [23]. The calculation was performed within the framework of the perturbation theory for laser wavelengths in the range from 190 to 260 nm. The quantum interference of three-photon absorption at the fundamental frequency and one-photon absorption at the third-harmonic frequency was taken into account. The expression

$$\Gamma = \Gamma_{3\text{ph}} + \Gamma_{1\text{ph}} + \frac{6}{k^2} \frac{I_1}{I_a} \left(\frac{I_1}{I_3} \right)^{1/2} \Gamma_{1\text{ph}} \left| \frac{S}{\mathcal{F}} \right| \times \cos(\Delta\varphi + \arg S - \arg \mathcal{F}) \quad (9)$$

was obtained for the total ionisation rate, where $\Gamma_{1\text{ph},3\text{ph}}$ are the rates of one- and three-photon processes; $k \equiv \hbar\omega/|E_{1s}|$; E_{1s} is the electron energy in the 1s state; $I_a \simeq 10^{16} \text{ W cm}^{-2}$ is the atomic intensity; $\Delta\varphi = \varphi_3 - 3\varphi_1$; \mathcal{F} is the scalar amplitude of one-photon ionisation; $S = A + 2B + 3C/5$; A , B , and C are functions of frequency ω determining the amplitude of three-photon ionisation of the hydrogen atom. The explicit dependences of A , B , and C on frequency ω in terms of hypergeometric and Appel functions are obtained in [24]. The phase dependences of the angular distribution of photoelectrons were analysed for different radiation intensities by using expression (9). It is important that the angular distribution of photoelectrons remains symmetric for all the values of parameters. The symmetry of the angular distribution is explained by the fact that, unlike the '1 + 2' phase control scheme, in the '1 + 3' scheme only odd angular harmonics of the continuous spectrum are excited.

In the case of the circular polarisation of one of the components of bichromatic radiation, the atom photoionisation probability will depend on the direction of rotation of the field vector (circular dichroism), the dichroism value being determined by the relative phase of radiation. Circular dichroism upon two-photon bichromatic ionisation of the hydrogen atom using the '1 + 3' scheme, as in [23], was studied theoretically in [25] as the function of the laser frequency, intensity, and relative phase. The calculation was performed within the framework of the perturbation theory for circularly polarised fundamental radiation and linearly polarised third-harmonic radiation. It was assumed that the polarisation vectors of monochromatic components lie in the same plane. The fundamental radiation wavelengths were considered near two-photon resonances at 194, 203.5, and 240 nm, where the amplitudes of one- and three-photon processes are close to each other in modulus. The calculated dependences of circular dichroism on the azimuthal escape angle of photoelectrons show that it strongly depends on the bichromatic phase $\Delta\varphi = \varphi_3 - 3\varphi_1$ and intensities of the monochromatic components of radiation.

It is well known [26] that the angular distribution of photoelectrons upon monochromatic ionisation of an atom depends on the radiation polarisation. In the case of the atomic effect in a bichromatic field, the polarisation dependence of the radiation pattern should be modified taking the relative phase $\Delta\varphi$ into account. This problem was analysed theoretically for photoionisation of calcium atoms in the '1 + 2' coherent control scheme [27]. The calculation was performed within the framework of the perturbation theory for elliptically polarised fundamental radiation at 400 nm and linearly polarised second-harmonic radiation. It was shown that the type of polarisation and azimuthal symmetry in the photocurrent directivity diagram depends considerably on the polarisation characteristics of the bichromatic field and the relative phase difference $\Delta\varphi$, so that a new type of the coherent control of photoprocesses is possible – the polarisation – phase control.

The directivity diagram of the atomic photoeffect in the above-threshold regime, when the process occurs with absorption of different numbers N of photons, depends, generally speaking, on N . This dependence upon photoionisation of xenon atoms by the intense bichromatic radiation ($I > 10^{14} \text{ W cm}^{-2}$) with the frequency ratio 1:2 was recently investigated in [28]. It was shown that the

angular distribution of photoelectrons considerably changed on passing from one electron peak to another. While if the angular distribution for the first peak (at the minimal energy) was almost symmetric, it had strong polar asymmetry in all other cases.

Bichromatic field (1) can be considered as the simplest realisation of the periodic sequence of laser pulses of a complicated shape. For a certain choice of parameters in expression (1), namely, when

$$F(t) = F_{\text{amp}}[2 \sin(\omega t - \phi) - \sin(2\omega t - 2\phi)]/4, \quad (10)$$

the time dependence of the total field can be written in the form

$$F(t) = F_{\text{amp}} \sin^2\left(\frac{\pi t}{T}\right) \sin(\omega t + \phi), \quad (11)$$

where $T = 2\pi/\omega$. It is interesting to note that the relative phase $\Delta\phi = \phi_2 - 2\phi_1$ is zero in this case. Variations in the parameter ϕ change the type of field oscillations inside the envelope, thereby changing the photoionisation rate of an atom. The above-threshold ionisation of xenon atoms in bichromatic field (10), (11) was calculated in [29]. By using the non-perturbation approach developed in [30], the general expression was obtained for the frequency-angular distribution of the above-threshold photoionisation rate taking into account contributions from different channels to the formation of a photoelectron peak with the specified energy $q\hbar\omega$ (q is an integer). The calculation showed the rate of ionisation by a linearly polarised bichromatic field is much higher than that by a circularly polarised field (at the same radiation intensity). By discussing possible applications of the bichromatic ionisation scheme, the authors of [29] pointed out that the phase ϕ of a wave packet can be stabilised within $\pm 0.1\pi$ with the help of a special interferometric technique [31].

The bichromatic photodestruction of atomic negative ions has been studied so far only theoretically, as far as we know. From the point of view of the theory of multiphoton processes, negative ions are a very convenient object for studies for which relatively simple and reliable analytic expressions can be obtained. This circumstance is explained by a weak interaction of the outer-shell electron with an atom. As a result, the excited states of the negative atom can be described in the plane-wave approximation, which is valid for free particles, which substantially simplifies the derivation of basic relations.

For example, the photodestruction of the negative ion of the atomic hydrogen caused by a bichromatic field with the frequency ratio 1:2 was calculated in [32] by a simple analytic method proposed in [33]. This method, which is the modification of the known Keldysh method [34], makes it possible to obtain reliable quantitative results for negative ions without time-consuming calculations. A linearly polarised bichromatic field of a moderate intensity ($I = 10^9 - 10^{11} \text{ W cm}^{-2}$) with a photon energy of 0.1 eV at the fundamental frequency was considered. The angular dependences of photoelectrons were calculated for different relative phases $\Delta\phi$, different intensity ratios $\eta = I_2/I_1$, different numbers of absorbed photons, and two photodetachment regimes (multiphoton and tunnelling).

In all the cases, the angular distribution strongly depended on the bichromatic phase. As the radiation

intensity was increased (passage to the tunnelling regime), the process probability oscillated stronger upon changing the photoelectron escape angle. At the same time, the angle-integrated probability of the process proved to be weakly dependent on the relative phase $\Delta\phi$ for the specified number of absorbed photons. After summation over this number, the phase dependence of the photodetachment probability virtually disappeared. The authors of [33] proposed to use the asymmetry of the photocurrent directivity diagram for obtaining a directed beam of neutral hydrogen atoms.

The same method was used in [35] to calculate the probability of electron photodetachment from the negative hydrogen ion in the bichromatic field with the frequency ratio 1:3. As mentioned above, the phase dependence of photoionisation depends considerably on the parity of numbers representing the frequency ratio of the monochromatic components of radiation. If these numbers have different parities, the photocurrent has the angular asymmetry, but the phase dependence of the probability integral is vanishingly small. In the opposite case, the directivity diagram is symmetric, but the angle-integrated probability of the photoprocess strongly depends on the bichromatic phase. This is confirmed by calculations performed in [35], where monochromatic components of radiation were assumed linearly polarised and collinear. The calculated angular distribution of photoelectrons proved to be polar-symmetric (with respect to the polarisation vector of radiation) but depended on the bichromatic phase. The phase dependence of the angular distribution is determined by the parameters of the problem such as the intensity ratio $\eta = I_3/I_1$ and the number of absorbed photons (the electron peak number in the energy spectrum). A comparison of results obtained in [35] with the results of complicated calculations [36] has demonstrated a high accuracy of the method used in [35].

As was first shown in [27], in a bichromatic field with the noncollinear polarisation geometry, along with the phase control, the polarisation-phase control is also possible at which the direction of photoelectron escape can be controlled by varying the polarisation of the bichromatic field and the relative phase. This question was later discussed in detail in [37], where the photodestruction of the negative hydrogen ion was studied in the bichromatic field with the frequency ratio 1:2 and linearly polarised monochromatic components. The angle α between their unit polarisation vectors e_1 and e_2 was treated as a variable parameter. The coherent control scheme of a special type was considered in which photons at the fundamental frequency were absorbed in one photodetachment channel, while second-harmonic photons were absorbed and photons at the fundamental frequency were simultaneously emitted in another channel. The calculation was performed by using the perturbation theory and model of the zero-radius potential for the electric field of the atomic core [38]. The dependences of the photocurrent direction and value on the angle α between the polarisation vectors of monochromatic components were analysed for different values of the relative phase $\Delta\phi$, and the phase dependences of the photocurrent were studied for the fixed angle α and different fundamental frequencies. As a result, it was shown that the value and direction of the photocurrent produced upon bichromatic destruction of negative ions can be controlled by varying the polarisation and relative phase of monochromatic components.

The action of the bichromatic field on a weakly bound electron via the perturbation of the atomic core [polarisation channel of the process (see review [39]) was neglected in the above-cited papers on the photodestruction of negative ions. This action can be quite significant for negative ions of alkali metals in which the polarisability of the neutral atom is high. In this case, the laser field induces a significant alternate dipole moment and related alternate polarisation potential in the electron shell of the ion core, which gives rise to a new photodetachment channel for the outer-shell electron. The influence of the polarisation channel on the photodestruction probability of the negative ions of alkali metals in the bichromatic field with the frequency ratio 1 : 2 was taken into account in [40]. The general expression describing the spectrally angular distribution of the electron photodetachment probability was obtained taking into account its dependence on the bichromatic phase. This expression predicted the polar asymmetry of the photocurrent directivity diagram, whose value was determined by the laser frequency and the relative phase $\Delta\varphi$. It was shown in [40] that interference between the direct and polarisation channels of the process reduces the photodetachment probability and changes the angular directivity of the photocurrent.

2.3 Phase control of elementary processes involving a laser field

A laser field can change the nature of elementary processes (such as scattering of electrons and photons by atoms, photorecombination of electrons by ions, etc.), which occur in the absence of electromagnetic radiation as well. Such laser-assisted processes in a monochromatic electromagnetic field were extensively studied beginning from the middle of the past century in connection with the problem of the development of a free-electron laser [41]. The scattering of fast electrons by a statistical potential in the presence of a bichromatic field was analysed in papers [42–45], where it was shown that the scattering probability can be efficiently controlled by varying the relative phase. The excitation of a hydrogen atom by an electron impact in the presence of a bichromatic laser field was calculated in paper [46] by using the Born approximation taking into account the perturbation of atomic states by the laser field. In this paper, the influence of the bichromatic phase on the angular dependences of the cross section and its symmetry was studied.

Let us discuss in more detail one of the recent papers [47] devoted to the phase control of scattering of electrons by a hydrogen atom in a bichromatic laser field with the frequency ratio 1 : 3. The authors of this paper considered the quantum interference of two free–free transitions of the scattering electron: two-photon absorption at the fundamental frequency (transition 1) and absorption of a third-harmonic photon and simultaneous simulated emission of a photon at the fundamental frequency (transition 2). The differential cross section σ of scattering after which the electron energy changes by $2\hbar\omega$ (ω is the fundamental radiation frequency) was obtained in the form

$$\frac{1}{I_1^2} \frac{d\sigma}{d\Omega_f} = (2\pi)^4 \frac{p_f}{p_i} \left[\mathfrak{F}_1^2 + \frac{I_3}{I_1} \mathfrak{F}_2^2 + 2 \left(\frac{I_3}{I_1} \right)^{1/2} \mathfrak{F}_1 \mathfrak{F}_2 \cos \Delta\varphi \right], \quad (12)$$

where $I_{1,3}$ are the intensities of the monochromatic components of radiation; Ω_f is the solid angle of electron scattering; $p_{i,f}$ are its initial and final momenta; and $\mathfrak{F}_{1,2}$ are the normalised quantum amplitudes of the two channels of

the process depending on the scattering angle. Each of these amplitudes is the sum of three terms describing the electron (static), polarisation, and mixed scattering mechanisms, respectively. The expressions for amplitudes $\mathfrak{F}_{1,2}$ in terms of hypergeometric functions were obtained in [48, 49]. The phase–angular dependences of the scattering cross section were calculated from (12) for three fundamental photon energies ($\hbar\omega = 1.17, 3,$ and 4 eV), the collinear polarisation of monochromatic components of radiation and $p_i \parallel e_{1,3}$. The analysis performed in [47] has demonstrated a strong dependence of the coherent phase control of electron scattering by an atom on the polarisation of the atomic core by the laser field. It was shown that the role of the atomic polarisation increases with increasing laser radiation frequency and decreasing the scattering angle of the electron.

The influence of resonance mono- and bichromatic fields on elastic and inelastic scattering of electrons by the hydrogen atom was studied in [50] by using the non-perturbation Born–Floquet theory. Bichromatic radiation with the frequency ratio 1 : 3 was considered, the third harmonic being in resonance with the $2s \rightarrow 3p$ transition in the hydrogen atom. The calculation was performed for monochromatic radiation components of the same intensity. This probably explains a weak dependence of the electron scattering cross section on the relative phase of bichromatic radiation obtained in this paper.

The theory of another laser-induced process, the ionisation of atoms by an electron impact in the presence of a bichromatic field, was developed in [51]. The scattering of electrons was described by using Volkov wave functions. The distortion of the bound state of the atomic electron was taken into account in the first order of the perturbation theory in the laser field. The wave functions of the continuous spectrum of the ionised electron were calculated approximately taking into account the influence of bichromatic radiation in all the orders of its strength. The latter circumstance proved to be quite critical for the adequate description of the effect. It was shown that the differential cross section of the process can be changed by an order of magnitude by varying the bichromatic phase.

The approach developed in [51] was used in [52] to calculate the cross section for scattering of X-rays by an atom in a bichromatic field with the frequency ratios 1 : 2 and 1 : 3. According to [52], an electron produced due to X-ray ionisation in this process exchanges the energy with the laser field and then recombines, by transferring the energy excess to an X-ray photon. As a result, the scattered X-ray photon has the energy that differs from its initial value by an integer number of laser field photons. Therefore, this process can be used to increase the X-ray photon energy. The numerical results obtained in the paper show that the cross section of the process in the bichromatic field increases by a few orders of magnitude compared to the scattering cross section of X-rays in a monochromatic laser field [53]. In addition, the scattering probability proved to be strongly dependent on the bichromatic phase, so that the efficient coherent control of this process is possible.

In [54], the radiative recombination and generation of X-rays were studied in a bichromatic field representing a coherent mixture of fundamental radiation with its third harmonic. This process is of interest from the point of view of its application for generating high-order harmonics upon laser heating of a plasma. The probability of electron

recombination by an atomic ion accompanied by the absorption (emission) of the specified number of laser photons was calculated as a function of the bichromatic phase by using quantum-mechanical and classical approaches. A special variant of the Keldysh–Faisal–Reiss model was used taking into account the influence of the Coulomb field of an ion on a recombining electron. The results of calculations show that the radiative recombination probability strongly depends on the relative phase of radiation and the coherent phase control of this process is possible.

Analysis of elementary processes in a bichromatic field showed that variations in the relative phase not only change the probability of the process but also permit the control of its basic parameters such as the scattered photon energy, the number of emitted (absorbed) photons, etc. The theoretical results obtained in [54] require experimental verification and the elucidation of the possibility of their practical applications.

3. Molecular processes in a bichromatic laser field

3.1 Photodissociation and photoionisation of molecules by bichromatic radiation

The idea of coherent control of laser-induced processes in molecules by using bichromatic radiation with multiple frequencies was first proposed in [55]. In this paper, the possibility to control the yield of dissociation products of a molecule in a bichromatic field with the frequency ratio 1 : 3 was analysed. Within the framework of the perturbation theory, the general expression was obtained for the probability $P(E, q)$ of producing the specified set (q) of dissociation products of a molecule excited to the state with energy E as a function of the relative phase of monochromatic components of radiation. This expression is similar to the formula for one-photon dissociation of the superposition state of a molecule [56]; however it describes a simpler experimental situation. In [55], the principle experimental scheme was proposed to observe the coherent radiative control of chemical reactions, which was later used in many studies in this field.

One of such experiments on the coherent laser control of bound–bound transitions in HCl and CO molecules was performed in [57]. The basic features of the process studied in this paper were the same as in paper [11] on bichromatic excitation of mercury atoms. The quantum interference of the three- and one-photon excitation of the bound–bound transition was detected by the ion current produced upon photoionisation of the upper term. Molecules were excited and ionised by radiation from a dye laser and its third harmonic ($\lambda_3 = 110 - 115$ nm) generated in a chamber with krypton. The laser pulse energy at the fundamental frequency was varied from 0.8 to 1.6 mJ. The third-harmonic intensity was varied by changing the krypton pressure. The optimal intensity ratio $\eta = I_3/I_1$ of monochromatic components, at which the control efficiency was maximal, was approximately 10^{-6} . The relative phase was varied by transmitting bichromatic radiation through a variable-pressure cell with argon or hydrogen. The laser beam obtained in this way was focused to a molecular beam in a reaction chamber, and photoexcitation products were recorded with a mass spectrometer.

Unlike paper [11], as the intermediate resonance level, different electronic and rotational states were used in [57]. In addition, along with a simple photoionisation detection system, dissociative ionisation products were detected. The maximum efficiency of the coherent control was 40 % for HCl molecules and 20 % for carbon monoxide, the optimal ratio of the probabilities of one- and three-photon excitation of the intermediated state of the molecule being equal to unity.

A new type of the phase control upon exposure of molecules to bichromatic radiation, which is absent in the atomic case, is the coherent control of photodissociation upon excitation of a molecule from the ground state to the repulsive electronic state. This process was studied theoretically in [58] for the photodissociation of molecular H_2^+ ions in an intense bichromatic field with the frequency ratios 1 : 2 and 1 : 3. The irradiation of ions by 780-nm, 150-fs pulses of intensity $I_1 = 10^{14}$ W cm $^{-2}$ was considered. The intensities of second and third harmonics were $I_2 = 3 \times 10^{13}$ W cm $^{-2}$ and $I_3 = 5 \times 10^{14}$ W cm $^{-2}$, respectively. In this case, the above-threshold photoionisation regime was realised, when the number of absorbed photons exceeded the minimal possible number determined by the energy threshold of the process.

Because the perturbation theory is inapplicable in the radiation intensity range under study, photodissociation was described by solving numerically the temporal Schrödinger equation by a special method developed for short pulses [59]. As in the case of the above-threshold photoionisation of atoms, the probability of the process studied in [58] is mainly determined by the peak value of the radiation intensity. Therefore, the calculated dependence of the photodissociation rate on the bichromatic phase is related most likely to the interference of monochromatic components of radiation rather than to the quantum interference of the transition channels of the system from the initial to final state. The authors of paper [58] calculated the probability of the bichromatic photodissociation of the H_2^+ ion excited from the $1s\sigma_g$ ground state to the repulsive $2p\sigma_u$ term for different relative phases $\Delta\varphi$. It was shown that even small coherent admixture of the second or third harmonic resulted in a considerable increase in the photodissociation rate. In addition, when the second harmonic is admixed, the phase dependence of the asymmetry of the proton escape forward or backward with respect to the radiation polarisation vector takes place.

This asymmetry of the dissociation-product escape was experimentally observed in [60], where the phase control of the photodissociation of molecular HD^+ ions excited by the fundamental radiation at 1053 nm and its second harmonic was studied. The ground and lower repulsive terms of this ion between which the radiative transition occurs are shown in Fig. 6.

The duration of the fundamental radiation pulse emitted by a mode-locked Nd:YLF laser was 50 ps and the pulse energy was 2 mJ (after amplification). The relative phase difference was varied by turning a glass plate through which bichromatic radiation was transmitted. The maximum power density of laser radiation focused in a reaction chamber was 2×10^{13} W cm $^{-2}$. Monochromatic components of the laser field in the interaction region had the same linear polarisation directed along the axis of a time-of-flight spectrometer. Along this axis a stationary electric field was applied to collect charged fragments on a detector. The

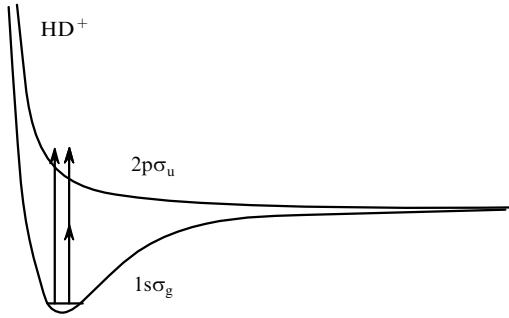


Figure 6. Potential curves for the $1\sigma_g$ and $2p\sigma_u$ states of the molecular HD^+ ion [60].

asymmetry of the photodissociation-product escape was measured by time-of-flight spectra characterising the directional distributions (to the detector and from it) of protons H^+ and deuterons D^+ (Fig. 7). Figure 7 also presents the time dependences of the electric field of bichromatic radiation for the phase difference corresponding to the given time-of-flight spectrum.

The time-of-flight spectra of protons (solid curves) exhibit two maxima characterising the asymmetry of their angular distribution. The first maximum (at a smaller time) corresponds to protons with the initial velocity directed to the detector, while the second one – from the detector. One can see from Fig. 7 that the relative intensity of these maxima is determined by the time dependence of the electric field strength, which in turn depends on the relative phase of

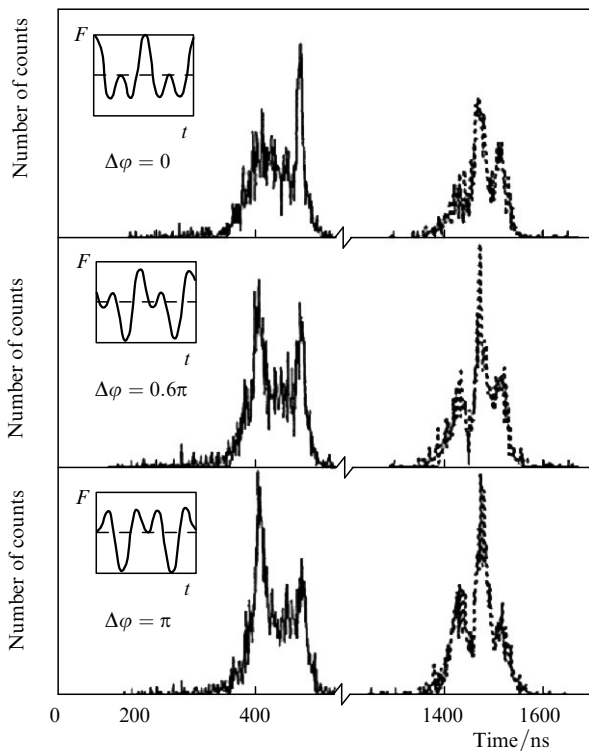


Figure 7. Time-of-flight spectra of photodissociation products of H^+ (solid curves) and D^+ (dashed curves) of the molecular HD^+ ion for different bichromatic phases [60]. The inset shows the time dependences of the electric field of bichromatic radiation for the given phase value.

monochromatic components. A similar situation is observed for deuterons (dashed curves) with the exception that their time-of-flight spectra have the central peak related to the presence of residual hydrogen and, hence, H_2^+ ions in the reaction chamber.

The time-of-flight spectra were used in [60] to obtain the dependences of the asymmetry of the dissociation-product escape, the ratios $\beta(\text{H}^+) = N_{\text{for}}(\text{H}^+)/N_{\text{back}}(\text{H}^+)$ and $\beta(\text{D}^+) = N_{\text{for}}(\text{D}^+)/N_{\text{back}}(\text{H}^+)$ [where $N_{\text{for}}(\text{H}^+)$, $N_{\text{back}}(\text{H}^+)$ and $N_{\text{for}}(\text{D}^+)$, $N_{\text{back}}(\text{D}^+)$ are the numbers of protons and deuterons escaped forward and backward, respectively] on the bichromatic phase and the phase dependence of the ratio of the dissociation-product yield $\varkappa = N(\text{H}^+)/N(\text{D}^+)$ (Fig. 8). The relative phase was calibrated by using the dependence of the electron current appearing upon photoionisation of krypton atoms by bichromatic radiation of the rotation angle of a phase-rotating plate (Fig. 8c).

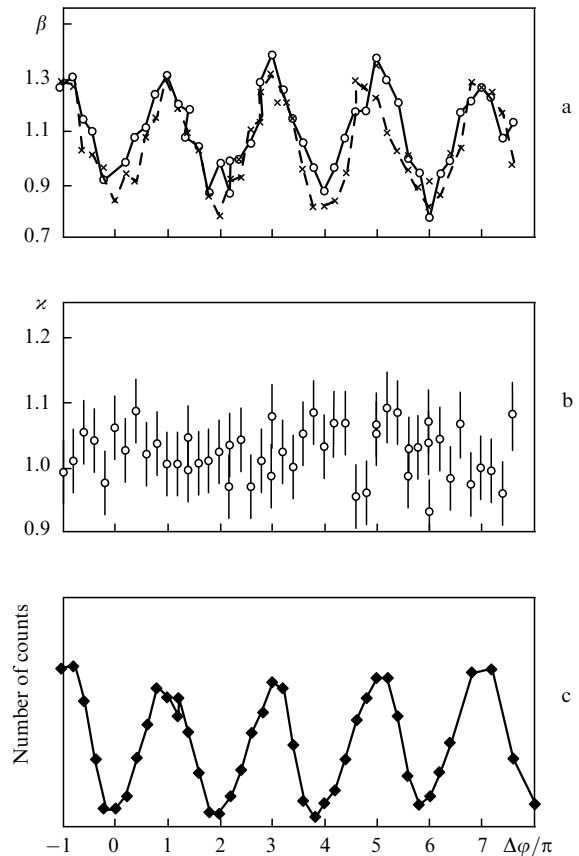


Figure 8. Phase dependences of the ratio of photodissociation products of the molecular HD^+ ion with initial velocities directed to a detector and from it, $\beta(\text{H}^+)$ (\circ) and $\beta(\text{D}^+)$ ($+$) (a), the phase dependence of the isotope separation parameter \varkappa (b), and the photoelectron current upon bichromatic ionisation of krypton (c) [60].

In [60], the dependence of the phase modulation depth of parameters $\beta(\text{H}^+)$ and $\beta(\text{D}^+)$ on the intensity ratio of monochromatic radiation components was also studied. It was shown that the maximum phase modulation depth of the asymmetry of dissociation-product escape ($\sim 70\%$) was achieved when the intensities of fundamental radiation and second harmonic were identical, i.e. when the phase

modulation of the bichromatic field strength was maximal. The latter is the result of photodissociation in a strong laser field, when the photodissociation rate is determined by the peak radiation intensity.

The coincidence of the phase dependences of the parameter β for protons and deuterons (Fig. 8a) shows that no isotope separation occurs for isotopes moving in the given direction during photodissociation.

Thus, the authors of [60] have demonstrated the coherent phase control of the angular distribution of products of the same type upon bichromatic photodissociation of a molecular ion. At the same time, the phase dependence of the ratio α for dissociation products ejected in the same direction was not observed (within the experimental error 9%).

3.2 Coherent phase control of the vibrational–rotational excitation of molecules

It is interesting to consider bichromatic vibrational–rotational excitation of molecules optically active in the IR region. This process was analysed theoretically in [61] for the interaction of CO molecules with bichromatic radiation with the frequency ratio 1:3. The two branches of the vibrational–rotational ($v_i = 0, J_i = J \rightarrow v_f = 3, J_f = J \pm 1$) transition (P and R branches) (where $v_{i,f}$ and $J_{i,f}$ are the vibrational and rotational quantum numbers, respectively) without changing the electronic state of the molecule were considered. The photon energy at the fundamental frequency in this case is ~ 0.26 eV. The calculation was performed by using the perturbation theory taking into account the anharmonicity of molecular vibrations by assuming that the ground electronic state is the $^1\Sigma$ state. Based on the Morse model for the interatomic potential, the three- and one-photon matrix elements of the transition dipole moment and the corresponding quantum-mechanical amplitudes were calculated.

By using these amplitudes, the expression was obtained for the total photoexcitation probability $W_{\text{tot}}(\Delta\varphi, J)$ of the molecule averaged over the projections of the angular momentum with the specified quantum number J , which takes into account the interference of the one- and three-photon channels of the process. The substitution of the numerical values of molecular parameters to the expression for $W_{\text{tot}}(\Delta\varphi, J)$, in which the most probable value obtained from the Boltzmann distribution is used as the rotational quantum number, gives the following expression for the optimal intensity ratio $\eta = I_3/I_1$ of monochromatic components in the R branch at temperature 300 K: $[\eta_{\text{opt}}(I_1)]^{1/2} = 4.24 \times 10^{-10} I_1$ (where I_1 is measured in W cm^{-2}). Analysis of the dependence of the parameter η_{opt} on the temperature of the molecular gas and rotational excitation branch showed that at relatively low temperatures the ratio η_{opt} is approximately the same for the P and R branches. As the temperature is increased, the ratio η_{opt} decreases, the decrease being stronger for the P branch of rotational excitation.

A new aspect in the coherent control of bichromatic excitation of a vibrational–rotational transition found in [61] is the dependence of the phase modulation of the excitation probability on the vibrational quantum number J of the initial state. In particular, there exist such values of $J_0^{\text{P,R}}$ near which the phase modulation coefficient changes its sign. It follows from the law of conservation of energy that the resonance laser frequency depends on J . Therefore, the

characteristics of the coherent control of the process under study can be changed by varying the laser frequency. This also can be done by changing the temperature of a molecular gas.

There also exist other methods of laser control of radiative molecular processes. For example, the photodissociation of Na_2 dimers in a two-frequency laser field with aliquant frequencies allowing the control of the yield of one of the sodium atoms in the specified excited state (3p or 3d) was performed in [62]. By varying one of the laser frequencies, the yield of $\text{Na}(3d)$ was increased by 25% at the expense of the corresponding decrease in the yield of $\text{Na}(3p)$. Thus, the control of the product yield in the specified channel of the photoreaction was experimentally demonstrated for the first time. A specific feature of photodissociation studied in [62] is its independence of the relative phase of monochromatic components of laser radiation, although this process is based on quantum interference. This circumstance is related to the specific dependence of the amplitudes of channels on the strengths of monochromatic components of radiation.

A detailed review of studies on the coherent control of molecular dynamics, including excitation from a superposition state, laser control of collision processes, control of the chaos dynamics, etc. is presented in [63].

4. Coherent control of photoeffect in a bichromatic field

An important new aspect appearing in the study of the bichromatic photoeffect from the surface and volume of solids is a different symmetry of the problem compared to that in the case of atoms. Indeed, dipole selection rules for atoms, determining the properties of bichromatic photoionisation, depend on the spherical symmetry of the system. The symmetry of the problem decreases in the case of solids, the selection rules are modified, and bichromatic photoeffect has its own specific properties in each particular case.

4.1 External photoeffect in a bichromatic laser field

The type of external photoeffect is determined by the nature of interaction of photoelectrons with a sample being irradiated. If the excess of the photon energy and momentum is transferred during photon absorption to ionised centres inside a solid, we are dealing with the volume photoeffect. In another limiting case, when interaction with the surface potential plays a key role, the surface photoeffect takes place. These two mechanisms can be distinguished by the polarisation dependence of the photocurrent: for the surface photoeffect, the projection of the field vector on the normal to the surface should be nonzero. The first experimental paper on the phase control of photocurrent from the surface of a solid under the action of bichromatic radiation was performed by Russian researchers [7] in the case of a dominating volume photoeffect.

Experiments on the external photoeffect in a bichromatic field with the frequency ratio 1:2 and the theoretical interpretation of this effect are presented in [8]. In this paper, the electron current from the cesium–antimonide photocathode of a FEU-127 photomultiplier was recorded. The red boundary of the photoeffect in this case was at the wavelength $\lambda_{\text{max}} = 600$ nm. The fundamental radiation of a

neodymium laser at $\lambda_1 = 1064$ nm and its second harmonic were used. The relative phase difference $\Delta\varphi$ between monochromatic components was varied by transmitting radiation through a variable-pressure cell with air. The phase dependence of the photocurrent appeared due to the quantum interference of two-photon absorption of fundamental radiation and one-photon absorption of the second harmonic, the single-photon photoeffect at the wavelength λ_1 being absent ($\lambda_1 > \lambda_{\max}$). To achieve the maximum efficiency of the phase modulation, the intensity of monochromatic radiation components was selected so that the single-photon and two-photon currents were approximately equal. The phase modulation depth of the photocurrent was a few tens of percent.

The theoretical treatment performed in [8] was based on the model of the zero-radius potential for ionised centres in a solid. This model gives simple analytic expressions for the amplitudes of one- and two-photon absorption. According to [8], the probability of photoionisation accompanied by the escape of an electron within the solid angle $d\Omega_n$ has the form

$$\frac{dW}{d\Omega_n} = B|f_1(\mathbf{nF}_2) + f_0F_1^2 + f_2[(\mathbf{nF}_1)^2 - F_1^2/3]|^2, \quad (13)$$

where the frequency-dependent coefficients $f_{0,1,2}$ describe the excitation of an ionised electron from the ground s state to the s , p , and d states of the continuous spectrum. As follows from the dipole selection rules, excitation to a state with an odd quantum number ($l=1$) of the angular momentum occurs due to one-photon absorption of the second harmonic radiation. Two-photon absorption of fundamental radiation leads to the excitation of a photoelectron to states with even quantum numbers $l=0, 2$. After squaring the modulus in the right-hand side of equality (13), the interference terms containing the product of strengths $F_2F_1^2$ appear, which determine the dependence of the photoionisation probability on the bichromatic phase. These terms also describe the polar asymmetry of the escape of photoelectrons. This asymmetry, in the opinion of authors [8], can be the basis of the mechanism for recording the hologram of the second-order dielectric susceptibility resulting in the photoinduced second harmonic generation in an optical fibre [5, 6].

It is important that the external photoeffect observed in [8] appeared upon the normal incidence of bichromatic radiation on a photocathode, which suggests that this process has the volume nature. The volume photoeffect is typical for materials with a low conductivity. In the case of metals, we are dealing with the external photoeffect which appears due to the action of the surface potential. The theory of the external surface photoeffect in a bichromatic field was developed in papers [64–66]. The simplest approach to the description of this phenomenon is based on the Sommerfeld step model of the surface potential $U(x)$, which is approximated by the expression $U(x) = U_0\theta(x)$, where $\theta(x)$ is the Heaviside function (the x axis is directed along the normal to the surface).

Such an approach was realised in paper [65], where the electron current produced upon exposure of a metal surface to bichromatic radiation with the frequency ratio 1:2 was calculated. In this case, the quantum interference of one-photon absorption of the second harmonic and two-photon absorption of fundamental radiation takes place. According

to [65], the normal projection of the photocurrent as a function of the bichromatic phase $\Delta\varphi = \varphi_2 - 2\varphi_1$ has the form

$$j_x = j_x^\Sigma [1 + (\zeta/2) \cos(\Delta\varphi + \Delta\varphi_i)], \quad (14)$$

where the photocurrent density j_x^Σ by neglecting quantum interference, the phased modulation coefficient ζ , and the internal phase shift $\Delta\varphi_i$ are expressed in terms of the amplitudes A_1 and A_2 of one- and two-photon absorption, respectively. The calculation was performed for the external photoeffect from the surface of metal sodium (the work function $A_{\text{out}} = 2.3$ eV). Figure 9 presents the dependences of the coefficient ζ , characterising the efficiency of the coherent control of photocurrent, on the fundamental radiation photon energy calculated for different intensity ratios $\eta = I_2/I_1$ and $I_1 = 3.5 \times 10^{10}$ W cm $^{-2}$. One can see that for the specified value of I_1 , the optimal intensities of monochromatic components are close to each other. The breaks in the curves in Fig. 9 correspond to the beginning of the photoeffect at the fundamental frequency. Note that the theory developed in [65] is valid for sufficiently short laser pulses, when the heating and damage of the irradiated surface can be neglected.

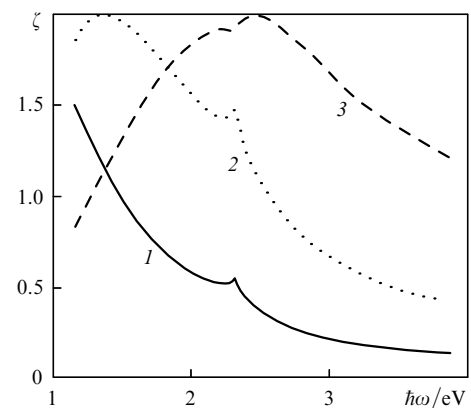


Figure 9. Phase modulation coefficient ζ of the electron photocurrent from the surface of metal sodium induced by a bichromatic laser field ($I_1 = 3.5 \times 10^{10}$ W cm $^{-2}$) as a function of the fundamental radiation energy for the parameter $\eta = I_2/I_1 = 10\%$ (1), 1% (2), and 0.1% (3) [65].

The Sommerfeld step model used in [64] to calculate the external photoeffect in a bichromatic laser field assumes the zero spatial width of the region of variation of the surface potential. It is interesting to find the region of applicability of this approximation. This question was studied in [65], where the function $U(x)$ was approximated by the expression

$$U(x) = \frac{U_0}{2} \left(1 + \tanh \frac{x}{2a} \right),$$

where a is the surface barrier width. It was shown that the amplitudes of one- and two-photon absorption near the two-photon photoeffect threshold weakly depend on the barrier width, so that the Sommerfeld step model well describes the process. Away from the threshold the

dependence of the absorption amplitudes on the parameter a becomes stronger; however, the modulus of their ratio $|A_2/A_1|$ changes weakly. Because the phase modulation coefficient is represented in the form $\zeta = 2 \sin[\arctan |A_2/A_1|]$, we can conclude that the efficiency of the coherent control of photocurrent weakly depends on the surface barrier width. At the same time, according to [67], the internal phase shift $\Delta\varphi_i = \arg(A_2/A_1)$ is more sensitive to the value of a , so that the phase dependence of photocurrent (14) will shift with increasing the surface barrier width.

The possibility of using the phase dependence of the current density (14) of bichromatic surface photoeffect for measuring variations in the work function A_{out} along a sample surface was discussed in [66]. The nonlinear optical diagnostics of surfaces of this type is based on the fact that the internal phase shift $\Delta\varphi_i$ is a function of the parameters of a sample and radiation frequency. In the Sommerfeld step model for electrons ejected from the Fermi surface, this function has the form

$$\Delta\varphi_i = \arctan \left[\frac{2(E_F + \hbar\omega)^{1/2} - (E_F + 2\hbar\omega)^{1/2}}{2(A_{\text{out}} - \hbar\omega)^{1/2} - A_{\text{out}}^{1/2}} \right],$$

$$A_{\text{out}} > \hbar\omega > A_{\text{out}}/2, \quad (15)$$

where E_F is the Fermi energy. As follows from (14) and (15), a change in the work function causes the shift of the phase dependence of the photocurrent. Compensating this shift by varying the bichromatic phase $\Delta\varphi$, we can determine the variation of the work function within the framework of model (15). For practical applications of this method, more realistic models of the surface potential should be used, because, as mentioned above, the internal phase shift $\Delta\varphi_i$ is quite sensitive to the potential barrier width. This dependence can be weakened by using laser radiation at frequencies near the threshold of two-photon photoeffect.

4.2 Internal photoeffect upon bichromatic excitation of solids

Internal photoeffect appears due to photoinduced transitions of electric current carriers inside solids. These transitions can be interband, intraband, and impurity–band. It is well known [68] that in the case of a centrally symmetric sample in the absence of an external field, the monochromatic excitation of a stationary electric current is impossible, i.e. the volume photogalvanic effect is absent. At the same time, upon excitation of charge carriers in a centrally symmetric solid by a bichromatic field of type (1), the so-called coherent photogalvanic effect is possible in the absence of an external voltage. The theory of this effect for the case of intraband and impurity–band transitions was developed in [2–4], as mentioned in Introduction.

The first experimental paper on the internal photoeffect upon bichromatic excitation [69] was devoted to the phase control of current in a layered AlGaAs/GaAs structure. A standard photodetector based on this structure has the considerable second-order dielectric susceptibility $\chi^{(2)}$ in the IR region, which is responsible for two-photon absorption. Therefore, the quantum interference of the amplitudes of one- and two-photon absorption is possible upon excitation of transitions between the energy subbands of

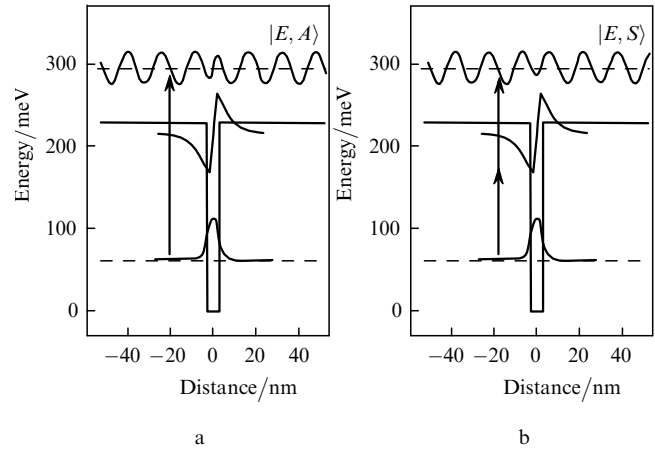


Figure 10. Energy level diagram for the system of GaAs/Ga_{0.74}Al_{0.26}As quantum wells and wave functions for the states between which one-photon absorption at $\lambda_2 = 5.3 \mu\text{m}$ (a) and two-photon absorption at $\lambda_1 = 10.6 \mu\text{m}$ (b) occur [69].

quantum wells in the AlGaAs/GaAs sample. This circumstance was used in [69] to obtain the phase control of photocurrent. The photocurrent was produced upon excitation of electrons by bichromatic radiation at $\lambda_1 = 10.6 \mu\text{m}$ and $\lambda_2 = 5.3 \mu\text{m}$ from the ground (bound) state of the quantum system to a state corresponding to the infinite motion (Fig. 10).

Radiation at the fundamental frequency was generated by a single-mode CO₂ laser emitting 100-ns pulses with a pulse repetition rate of 2 Hz. Frequency doubling was performed in an AgGaSe₂ crystal with the efficiency of 2×10^{-4} . The amplitudes of one- and two-photon absorption were equalised with the help of a set of optical filters to obtain the maximum phase control efficiency. The photocurrent in a periodic GaAs/Ga_{0.74}Al_{0.26}As structure containing 2.5 quantum wells of thickness 55 Å separated by 325 Å was measured at 82 K. The value and direction of the recorded photocurrent were changed in the interval $\pm 5 \mu\text{A}$ by varying the relative phase $\Delta\varphi = \varphi_2 - 2\varphi_1$. Thus, the authors of [69] have demonstrated experimentally the coherent control of photoprocesses in semiconductors with the help of bichromatic radiation with multiple frequencies.

The coherent control of photocurrent due to excitation of interband transitions in bulk semiconductors by bichromatic radiation with the frequency ratio 1 : 2 was considered theoretically in [70]. The densities of photoinduced electron and hole currents were determined from kinetic equations taking into account the charge and current injection rates upon excitation of electrons from the valence band to the conduction band. A specific feature of the internal photoeffect upon bichromatic excitation of a centrosymmetric material is that the current density injection rate is proportional to the ‘cross’ term containing the product of the electric-field strengths of fundamental and second-harmonic radiations [the last term in the second equality in (4)]. Calculations performed in [70] for the case of bichromatic excitation on the internal photoeffect in GaAs by neglecting the noncentrosymmetric part of the dielectric susceptibility showed that the photocurrent produced under real experimental conditions can be easily measured.

The corresponding experiment was performed in [71] by using picosecond and femtosecond laser pulses at

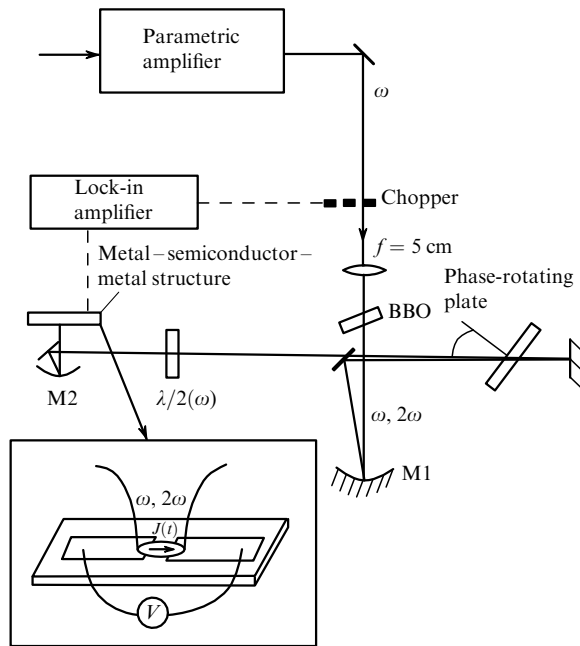


Figure 11. Scheme of the experimental setup [71] for observation of the coherent control of photocurrent in a bulk GaAs sample appearing under the action of bichromatic excitation in the absence of an external voltage (M1 and M2 are spherical mirrors).

$\lambda_1 = 1550$ nm and the second harmonic of this radiation (Fig. 11). To excite the internal photoeffect in a bulk GaAs sample, short laser pulses are required because the phase relaxation time of charge carriers is very short (of the order of 10^{-13} s). Pulses of duration 175 fs with a repetition rate of 250 kHz and the average power up to 30 mW were generated by an optical parametric oscillator tunable in a broad wavelength range from 1100 to 2900 nm. Picosecond pulses at 1550 nm with a repetition rate of 82 MHz and an average power of 150 mW were obtained from a parametric amplifier. The second harmonic was generated in both cases in a barium β -borate crystal.

The coherent control of photocurrent was observed at room temperature in samples of different thickness of two types: in normal gallium arsenide (obtained under normal conditions) and gallium arsenide grown at low temperature (LT-GaAs). The electron-hole recombination time in samples of the first type exceeds 10 ns (at the carrier concentration lower than 10^{18} cm $^{-3}$), and the phase control of photocurrent is possible only upon excitation by laser pulses with a relatively low repetition rate due to the charge accumulation effect. Samples of the second type have a short carrier trapping time (of the order of a picosecond), which prevents the charge accumulation. Because of this, the coherent control of the internal photoeffect in them was observed upon excitation by both bichromatic radiation sources. The maximum current density in a sample of normal GaAs upon excitation of the internal photoeffect by a parametric oscillator was 500 nA μm^{-2} . When a parametric amplifier and an LT-GaAs sample were used, the maximum current density was 3 nA μm^{-2} . The sinusoidal dependences of the photovoltage on the relative phase $\Delta\varphi = \varphi_2 - 2\varphi_1$ (Fig. 12) obtained in [71] demonstrate the coherent control of the internal photoeffect in bulk semi-

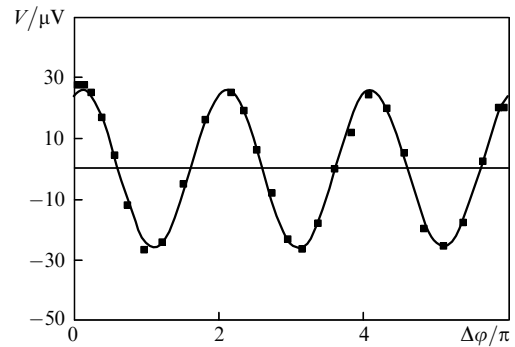


Figure 12. Phase dependence of the photovoltage V produced due to bichromatic generation of photocurrent in a bulk GaAs sample [71]. Solid curve is the best sinusoidal fit of the experimental data.

conductors by using bichromatic radiation with multiple frequencies without applying an external voltage.

In [72], the quantum interference of one- and two-photon absorption was studied upon bichromatic generation of charge carriers in noncentrosymmetric semiconductors. It was shown experimentally that this interference produces the phase modulation of the charge carrier concentration and the transmission coefficient of monochromatic components of radiation by changing their relative phase. A bulk gallium arsenide sample at room temperature was exposed in experiments [72] to radiation at $\lambda_1 = 1550$ nm and its second harmonic. In this case, two-photon absorption is possible because the sample possesses the second-order dielectric susceptibility $\chi^{(2)}$ (which was neglected in the theory [70]). The duration of the fundamental radiation pulse was 120 fs, the pulse repetition rate and average pulse power were 250 kHz and 8 mW, respectively. Radiation at a wavelength of 1650 nm, whose second harmonic was used to probe the charge carrier density, had similar parameters. Radiation frequency doubling was performed in a barium β -borate crystal, and the bichromatic phase $\Delta\varphi$ was varied with the help of a Michelson interferometer. The experimental dependences of the charge carrier concentrations and transmission coefficient at 775 nm on the relative phase of monochromatic components were obtained. These phase dependences proved to be shifted with respect to each other by $\Delta\varphi_\chi = (0.35 \pm 0.05)\pi$. Analysis performed in [72] showed that the phase shift $\Delta\varphi_\chi$ is related to the argument of the complex quantity $\chi^{(2)}$. The ratio $\text{Im}\chi^{(2)}/\text{Re}\chi^{(2)} = 0.32 \pm 0.06$, determined from this relation, is close to the theoretical value 0.37 obtained in [73]. Thus, the phase control of the internal photoeffect opens up new possibilities for experimental measurements of the optical constants of materials.

4.3 Bichromatic control of the spin of current carriers in semiconductors

Apart from the use of bichromatic excitation for the coherent control of electric current and carrier concentration, it is interesting to apply it for controlling the spin of charge carriers in semiconductors. This direction is promising both for the fundamental study of the spin dynamics in multiparticle systems and for applications as the possible mechanism for optical data processing and storage [74]. Such a control in a bichromatic field with the frequency ratio 1 : 2 was first proposed in [75]. In this paper,

the idea of quantum interference was combined with the idea of optical generation of spin-polarised carriers by exposing a semiconductor to circularly polarised radiation [76]. It was shown that, for a certain polarisation of monochromatic components of radiation, the quantum interference of one- and two-photon absorption produces the spin current in the semiconductor, which depends on the relative phase and appears in the absence of an external field. Note that the spin current can be produced without the accompanying electric current, which allows one to study the spin dynamics in a pure form. Thus, in the case of the same circular polarisation of monochromatic components, the spin current consists of two components, one of which is related to the electric current, whereas another is not related to it. If the linear mutually orthogonal polarisation of bichromatic radiation takes place, the spin current is not accompanied by the electric current but is a superposition of four antiparallel currents in pairs. In this case, the spins of carriers moving in the opposite directions in each of the two pairs are antiparallel to each other. As a result, the total electric current is absent. This method of spin control is promising, in particular, because it permits the manipulation of carrier spins at the femtosecond scale by using ultrashort laser pulses.

The scheme [75] for the coherent control of the spins of current carriers in semiconductors was realised in [77]. The spin-polarised electric current was observed in a gallium arsenide sample at room temperature excited by 150-fs laser pulses at wavelengths 1550 and 775 nm with the same circular polarisation. The electric-current component perpendicular to the laser beam was measured as a function of the relative phase of monochromatic components. The results obtained in [77] confirmed theoretical conclusions reported in [75].

The coherent generation of spin-polarised carriers upon bichromatic excitation of semiconductors was first directly demonstrated in papers [78, 79]. In [78], the coherent generation and control of the purely spin current was observed by exposing a layered GaAs/AlGaAs structure to 110-fs, 1420-nm pulses of fundamental radiation and its second harmonic. In the case of a linear mutually orthogonal orientation of monochromatic components of radiation, the excited current carriers with antiparallel spins move in the opposite directions [75]. As a result, the spatial separation of the 'upward' and 'downward' spin-localisation regions occurs during the time of the order of the pulse relaxation time. This spatial separation was recorded in experiments [78] with the help of circularly polarised probe radiation, which selectively excited carriers with the specified spin projection. The dependences of the sample transmission τ on the bichromatic phase were obtained on each side of the excitation region (in the propagation direction of current carriers) and in the focal region. In accordance with theoretical predictions [75], the phase dependences of τ at points away from the excitation region proved to be modulated with the phase shift 180° , and a change in transmission in the excitation region was independent of the bichromatic phase. Thus, the coherent control of electrons with the specified spin orientation excited by bichromatic radiation in a semiconductor was demonstrated without the appearance of the electric current.

The purely spin current was directly observed upon bichromatic excitation of a bulk ZnSe sample in [79]. This semiconductor has a long spin relaxation time even at

relatively high temperatures. In addition, its energy gap width (2.72 eV at 100 K) permits the use of radiation from a Ti:sapphire laser at a wavelength of 400 nm and its second harmonic for pumping. The phase-dependent effect was recorded by the spatial displacement of the circularly polarised photoluminescence signal, which was a measure of the concentration of charge carriers with the specified spin orientation. This measuring method is based on the selection rules for dipole radiation, which relate the polarisation of photoluminescence with the direction of spins of electrons undergoing the optical transition. In [79], the spatial separation of regions with the right and left circular polarisations of photoluminescence was measured as a function of the relative phase of monochromatic components. The sinusoidal form of this dependence demonstrates the coherent control of the value and direction of antiparallel currents of carriers with oppositely oriented spins.

Experiments [69, 71, 78, 79] on the coherent control of bichromatic excitation of semiconductors have demonstrated wide possibilities for controlling the dynamics of current carriers by using the quantum interference of the amplitudes of optical transitions. A detailed theory of these phenomena accounting for the electron-hole interaction is presented in recent paper [80]. In particular, it was shown that exciton effects increase the probability of one- and two-photon absorption and introduce the phase shift to the excitation amplitudes of electric and spin currents.

5. Conclusions

The papers analysed in the review lay a theoretical foundation and verify experimentally the basic principles of the coherent control of radiative processes by bichromatic radiation with multiple frequencies. The quantum interference of optical excitation amplitudes on which this control is based can be used to change different characteristics of radiative processes by varying the relative phase and polarisation of monochromatic components of radiation. Experiments performed to date have demonstrated the possibility of the phase control of excitation of atoms in discrete and continuous spectra, the angular and polarisation control of photocurrent from gases and surface of solids, the phase control of photodissociation and photoionisation of molecules, and the coherent control of current carriers in semiconductors, including spin control.

Note that in recent papers the principles of the coherent control of photoprocesses by bichromatic radiation with multiple frequencies were extended from the optical range to other ranges. Thus, theoretical and experimental studies on the coherent control of the ionisation of the Rydberg state of the hydrogen atom ($n = 51$) under the action of a two-frequency electric field (6 and 18 GHz) by varying the relative phase were performed in [81]. The phase dependence of the probability of excitation of the helium atom from the Rydberg 28^3S state by a bichromatic field of the same type as in [81] was experimentally obtained in [82].

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