

# Control of elementary chemical reactions by femtosecond light pulses\*

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**Abstract.** Essentially new approaches are proposed for controlling the yield of products of a chemical reaction by femtosecond laser pulses. The main attention is paid to the method based on the change in phase characteristics of femtosecond pulses. The quantum yield of the product  $\text{NH}(c^1I)$  upon multiphoton photodissociation of ammonium in gas is shown to increase by a factor of twelve with chirp variation from  $-1250 \text{ fs}^2$  to  $2000 \text{ fs}^2$ . It is demonstrated by the example of two-proton transfer in [2,2'-bipyridyl],3,3'-diol in solution that the approach based on the variation in phase characteristics of a pump femtosecond pulse can be also used for controlling reactions in solutions.

**Keywords:** elementary chemical reactions, femtosecond pulses, phase characteristics, wave-packet dynamics.

Over the entire history of the development of chemistry the researchers have made attempts to answer the question of how the yield of products of a chemical reaction can be controlled. At the macroscopic level, reactions were controlled by varying pressure, temperature, and other parameters determining the conditions of their proceeding. The control of elementary reactions by varying the type of interaction of light with reagents has long been attempted. The review of such attempts is presented in Ref. [1].

Our paper is devoted to the development of the essentially new possibilities for controlling the course of chemical reactions with the help of a novel tool in chemistry – femtosecond light pulses. We review the known examples of such a control, propose new approaches based on the specific properties of femtosecond pulses and present new experimental confirmations of the efficiency of using phase characteristics of femtosecond pulses for controlling elementary reactions in gases and solutions.

Before proceeding to the discussion of the methods for controlling chemical reactions, we consider the features of

the new tool and the experimental possibilities offered by them.

The electric field strength  $E(t)$  of a femtosecond light pulse at a given point in space in the coordinate system whose  $z$  axis coincides with the direction of propagation of the laser pulse can be written in the form

$$\begin{aligned} E_x(t) &= E_{x0}f(t) \cos[\omega_0 t + \delta_x + \alpha(t)], \\ E_y(t) &= E_{y0}f(t) \cos[\omega_0 t + \delta_y + \alpha(t)], \\ E_z(t) &= 0, \end{aligned} \quad (1)$$

where  $f(t)$  is the function describing the time dependence of the pulse envelope;  $\alpha(t)$  is the frequency modulation during the pulse duration;  $\omega_0$  is the pulse carrier frequency. The light pulse polarisation is described by the ratio  $E_{x0}/E_{y0}$  and phases  $\delta_x$  and  $\delta_y$ . The pulse envelope  $f(t) \sim [I(t)]^{1/2}$  (where  $I(t)$  is the pulse intensity) can have various shapes.

Usually, Gaussian pulses with a linear modulation are considered, for which

$$f(t) = \exp(-t^2/2\tau^2), \quad (2)$$

$$\alpha(t) = \frac{1}{2}\gamma t^2, \quad (3)$$

where  $\tau$  is the pulse duration;  $\gamma$  is the phase modulation rate (the time chirp).

The light pulse field can be also defined in terms of spectral characteristics

$$E_j(\omega) = \int_{-\infty}^{\infty} E_j(t) \exp(i\omega t) dt, \quad (4)$$

where  $j = x, y$ . Because the functions  $f(t)$  and  $\exp(i\alpha t)$  usually vary slowly compared to the function  $\exp(i\omega_0 t)$ , we have

$$E_j(\omega) = \frac{1}{2} \exp(-i\delta_j) F_j(\omega - \omega_0) = \frac{1}{2} \exp(-i\delta_j) E_{j0}. \quad (5)$$

Note that  $|F_j(\omega - \omega_0)| \sim S(\omega)^{1/2}$ , where  $S(\omega)$  is the experimentally measured spectrum of the pulse power. The spectral phase  $\phi(\omega - \omega_0)$  is related to the functions  $f(t)$  and  $\alpha(t)$  in a rather complicated way. In particular, for Gaussian pulses defined by expressions (2) and (3), we have

$$\phi(\omega - \omega_0) = \frac{1}{2} \beta (\omega - \omega_0)^2. \quad (6)$$

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The modulation of the carrier frequency is determined by the spectral chirp

$$\beta = \frac{4\gamma\tau^4}{16(\ln 2)^2 + \gamma^2\tau^4}. \quad (7)$$

The spectral chirp of a pulse with the Gaussian envelope is related to the pulse duration by the expression

$$\beta = 0.36\tau_0^2 \left[ \left( \frac{\tau}{\tau_0} \right)^2 - 1 \right]^{1/2}, \quad (8)$$

where  $\tau$  and  $\tau_0$  are the FWHM durations of chirped and bandwidth-limited pulses, respectively.

The important parameter of a femtosecond pulse is its spectral FWHM  $\Delta\omega$ , which is determined by the uncertainty relation  $\tau\Delta\omega = \text{const}$ . This constant depends on the shape of the pulse envelope and its chirp. For a Gaussian pulse with  $\gamma = 0$  (which is called a bandwidth-limited pulse), this constant is 2.773.

Let us list the main experimental possibilities related to the properties of femtosecond pulses.

(i) The short duration of femtosecond pulses allows one to study the time evolution of chemical processes with the time resolution that is even less than the light pulse duration ( $10^{-14}$  s).

(ii) The coherence and large spectral width of a femtosecond pulse provide coherent excitation of several vibrational or rotational states. This new type of the excited states is called a nonstationary quantum state or a coherent nuclear wave packet. The specific feature of such a state is the presence in a molecule of several simultaneously excited stationary energy states with certain relative phases.

(iii) The high intensity (peak power) of femtosecond pulses along with the low pulse power allows one to easily realise multiphoton absorption processes, to use a variety of methods of nonlinear spectroscopy without sample heating and to obtain the light field strengths exceeding those of intramolecular fields.

These properties of femtosecond pulses suggest the following approaches to controlling chemical reactions.

(1) *Optical distortions of the statistical properties of monomolecular reactions.* This approach is based on the preparation with the help of a femtosecond pulse of vibrationally excited molecules with the intramolecular distribution of the vibrational energy at which a monomolecular reaction does not obey statistical laws. To control the dissociation of the molecule into the required products, the following two conditions should be fulfilled. The excitation energy should be localised in a certain part of the molecule and the intramolecular energy distribution should not occur during the time of the reaction. The nonstatistical decays of radicals vibrationally excited in the ground electronic state by femtosecond pulses have been observed in Refs [2, 3].

(2) *Optical action on the potential-energy surface (PES).* In this method, the PES is changed at the required instant of time with the help of femtosecond pulses, which allow one to control the dynamics of a transient state. A simplest example of such a control is photodissociation of OCIO [4].

(3) *The use of dynamics of nonadiabatic transitions.* The method for controlling the time evolution of a molecule using nonadiabatic transitions (both upward and downward) was proposed in Ref. [5]. It involves the preparation of a wave packet by a femtosecond pulse followed by its excitation by a second pulse at the required instant of time,

resulting in the system transfer to a new state. The preparation of molecules in various excited electronic states [6] and the production of  $\text{Na}^+$  and  $\text{Na}^{2+}$  ions [7] using a pair of femtosecond pulses with various delay times serve as the experimental demonstration of this method.

(4) *The use of the orientation dynamics.* The implementation of this approach is expected in the future. The reaction efficiency is known to depend substantially on the relative orientation of colliding partners both in gases and a condensed phase. Femtosecond pulses have already been applied for the preparation of coherent rotational packets in the ground electronic state [8, 9], which can be used in reactions in Van der Waals complexes and condensed media.

(5) *The use of the coherent dynamics.* The possibility of such an approach was analysed in detail in Ref. [10]. Here, we consider the application of chirped pulses for the preparation of a nuclear wave packet with certain phase and amplitude parameters [11]. Obviously, the control of this type is possible if the reaction proceeds faster than the dephasing of a molecular system. Because the characteristic coherence times of nuclear wave packets are, as a rule, 100–1000 fs, the reactions of excited molecules should proceed for even shorter times. This requires considerable excitation energies, and probably for this reason the multiphoton absorption is commonly used which can provide the excitation energy up to 10–15 eV.

The influence of a chirp on the intramolecular dynamics and photophysical and photochemical processes has been studied at the beginning of the development of femtochemistry (see Table 1).

**Table 1.** Examples of the chirp effect on some physicochemical processes

Process	Parameters or processes depending on chirp	References
$\text{Na}_2 + nh\nu \rightarrow \text{Na}^+$ $\text{Na}_2 + nh\nu \rightarrow \text{Na}_2^+$	Quantum yield of ions	[12]
$\text{NaI} + hv_1 \rightarrow \text{NaI}$ $\text{NaI}^* \rightarrow \text{Na} + \text{I}$ $\text{NaI}^* + hv_2 \rightarrow \text{Na}^* + \text{I}$	$\text{Na}^*/\text{Na}$ ratio	[13]
Vibrational dynamics of $\text{I}_2$ in the excited electronic state	Vibrational dynamics	[14, 15]
Rotational dynamics of $\text{D}_2$ and $\text{N}_2$ in the ground electronic state	Rotational dynamics	[8]
Fluorescence of LD690 molecules	Quantum yield of fluorescence	[16]

One can see from Table 1 that the quantum yields of ionisation of the  $\text{Na}_2$  molecule and formation of excited sodium atoms, the dynamics of vibrational wave packets of the  $\text{I}_2$  molecule in the excited electronic state, and the dynamics of rotational wave packets of  $\text{D}_2$  and  $\text{N}_2$  molecules in the ground electronic state depend on the exciting pulse chirp. It was shown in paper [16] that the quantum yield of fluorescence of dye solutions depends on the chirp sign in the case of high intensities of the exciting pulse.

The above examples demonstrate that the phase parameters of pulses can be used for controlling the dynamics of atoms in molecules. It also follows from these data that, by changing excitation of atoms in a transient state with the

help of femtosecond pulses, we can control their dynamics in the transient state, thereby controlling the yield of products in an elementary chemical reaction.

Recently, experimental papers were published that demonstrated that the phase characteristics of an exciting pulse could substantially change the channels of the product yield upon photodissociation of polyatomic molecules.

The study of multiphoton dissociation of  $\text{CH}_2\text{I}_2$  showed that, provided the excitation energy is sufficient, the reaction can proceed via several channels. In Ref. [17], the dependence of multiphoton photodissociation of  $\text{CH}_2\text{I}_2$  on the sign and value of the spectral chirp  $\beta$  was considered. Excitation was performed both by 624-nm pulses and by 312-nm frequency-doubled pulses. The spectral chirp  $\beta$  was varied from  $-2000$  to  $2400 \text{ fs}^2$ .

Upon multiphoton absorption of 624-nm laser pulses, the well-known fluorescence of molecular iodine was observed at 342 nm. The maximum yield of molecular iodine in the electronic state  $D$  was observed for  $\beta = -500 \text{ fs}^2$ , while the minimum yield was observed for  $\beta = -2400 \text{ fs}^2$ , the yield of molecular iodine decreasing by a factor of 2.9. The dependence of the product yield on the chirp value was also observed upon excitation of  $\text{CH}_2\text{I}_2$  molecules by 312-nm pulses, being, however, of the opposite type: the maximum and minimum yields were obtained for  $\beta = -2400 \text{ fs}^2$  and  $-500 \text{ fs}^2$ , respectively. The absence of the symmetry of the dependence of the product yield on the chirp relative to the zero chirp means that the changes in the product yield observed in experiments are related to the phase characteristics of the exciting pulse rather than to its duration.

Because the parameters of electronic PESs at such energies are unknown, it is necessary to find femtosecond pulses with adequate properties that can be used to modify complicated molecular systems, which is very difficult to calculate quantum-chemically. For this reason, a number of researchers are developing another approach. Thus, it was shown in Ref. [18] that, within the optimal control theory, the scheme involving the feedback over the yield of reaction products could be used. This scheme allows one to determine the pulse shape and its spectrum, which maximise the yield of a certain product.

This approach was experimentally realised in Ref. [19] where the reaction channels were automatically optimised for a metalloorganic compound. Due to multiphoton photodissociation produced by femtosecond pulses, two channels of the yield of reaction products are formed, which are detected with a mass spectrometer. The mass ratio being measured is processed with a computer using a special algorithm, which generates a signal to change the phase parameters of a femtosecond pulse in order to increase the ratio.

Another example is our study of ammonia photodissociation upon multiphoton absorption [20]. 300- $\mu\text{J}$ , 50-fs laser pulses at 616 nm with a pulse repetition rate of 25 Hz were focused inside a chamber with gaseous ammonia at room temperature at a pressure of 3 Torr. The pulse chirp was changed by varying the optical path in a prism compressor. The pulse duration was measured at each position of the compressor with the help of an autocorrelator; the chirp value was changed from  $2000 \text{ fs}^2$  to  $-2000 \text{ fs}^2$ . The dependence of the pulse duration on the chirp  $\beta$  is shown in Fig. 1. Along with the pulse duration, the energy of each pulse and the integrated intensity of fluorescence in the range from 300 nm to 400 nm were detected for all chirp values.

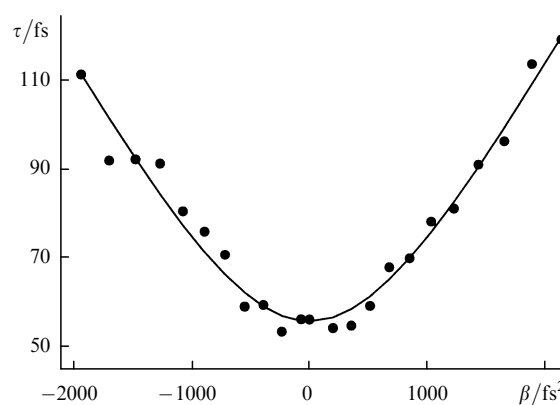
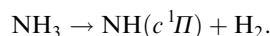


Figure 1. Dependence of the pulse duration  $\tau$  on the pump-pulse chirp  $\beta$ .

It is known that when the energy of one-photon excitation exceeds 6 eV, ammonia dissociates into the  $\text{NH}_2$  radical in the ground  $X^2B_1$  state and a hydrogen atom. When the excitation energy of ammonia exceeds 9.4 eV, another channel of the reaction is realised [21]:

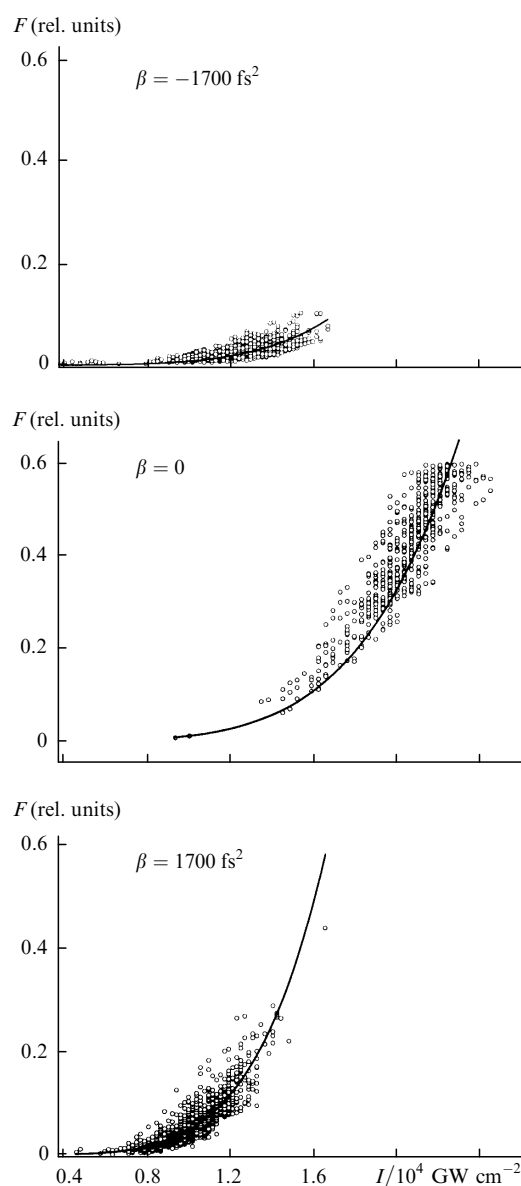


We studied multiphoton dissociation of ammonia irradiated by 615-nm (2-eV) femtosecond pulses. In this case, the excitation energy of ammonia will exceed 9.4 eV if five photons are absorbed. By absorbing the energy of 10 eV, the ammonia molecule undergoes the transition from the ground state to an excited electronic state. Because the excitation energy is still insufficient for ionisation, the ammonia molecule dissociates via two channels. The NH radical is formed upon photodissociation in the  $c^1\Pi$  excited electronic state, from which fluorescence is observed upon the  $c^1\Pi \rightarrow a^1A$  transition at 324 nm [21].

Therefore, by monitoring the fluorescence signal, which is proportional to the population of the NH radical in the  $c^1\Pi$  state, we can estimate the quantum yield of the ammonia dissociation via this channel. We detected the dependence of the integrated intensity  $F$  of fluorescence in the spectral region from 300 nm to 340 nm on the intensity  $I$  of exciting pulses in a focus for the spectral chirp varied from  $-2000 \text{ fs}^2$  to  $2000 \text{ fs}^2$ . Fig. 2 shows the dependences  $F(I)$  for some values of the spectral chirp. The solid curves correspond to  $F = A(\beta)I^5$ , where  $A(\beta)$  is a coefficient depending on the spectral chirp. One can see that the experimental data and calculations are in good agreement. This means that we deal with five-photon absorption.

The coefficient  $A(\beta)$ , which characterises the influence of the chirp on the ammonia photodissociation, can be determined from the dependences presented in Fig. 2. The normalised dependence  $A(\beta)$  is shown in Fig. 3. Because the intensity of fluorescence of the  $\text{NH}(c^1\Pi)$  radical is proportional to its population in the  $c^1\Pi$  state, the dependence shown in Fig. 3 gives the dependence of the quantum yield of the  $\text{NH}(c^1\Pi)$  radical (in relative units) on the spectral chirp. The ratio of the maximum quantum yield to the minimum yield in Fig. 3 is equal to 12. Note also that the dependence of the quantum yield is asymmetric relative to the zero chirp, which means that the variations observed in experiments are related to the phase parameters of the pulse rather than to its duration.

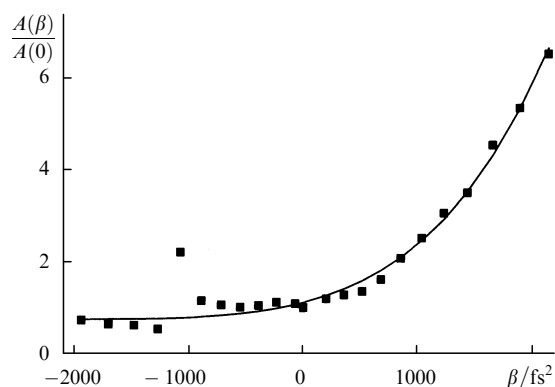
Effects caused by the chirp determine the PES parameters and the dynamics of wave packets. It was mentioned



**Figure 2.** Dependence of the integrated intensity  $F$  of fluorescence in the 300–340-nm range on the pump-pulse intensity  $I$  for different  $\beta$ .

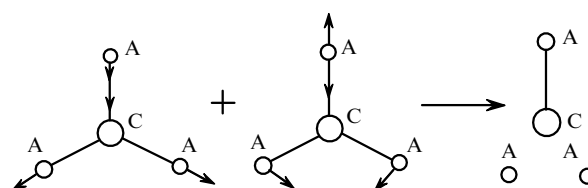
above that the parameters of highly excited electronic PESs are unknown and it is difficult to calculate them quantum-chemically. This severely complicates the interpretation of experimental data, which can be explained only qualitatively. A change in the chirp of the exciting pulse results in the different distributions of the initial phases of stationary states whose superposition forms a wave packet. This causes, as was noted above, a change in the dynamics of atoms in a transient state.

Fig. 4 explains schematically the control of the product yield with the help of the phase characteristics of a femtosecond pulse by the example of a four-atomic molecule. The usual photodissociation of molecules of this type involves the dissociation of the C–A bond, when the vibrational energy is released on the valence vibration, which determines the reaction coordinate. Let us assume that the components of the wave packet are antisymmetric and deformation vibrations, which determine the reaction coordinate according to the concerted mechanism. One can see from



**Figure 3.** Dependence of the coefficient  $A(\beta)$  on the spectral chirp  $\beta$ .

Fig. 4 that if the motion of atoms A is synchronised so that two bonds are synchronously stretched and their deformation vibrations, which occur simultaneously with the C–A bond stretch, are involved in the formation of the new A–A bond, then the concerted mechanism will be realised more efficiently. It seems that the concerted mechanism can be efficiently realised in a rather large class of reactions by synchronising the motion of atoms.

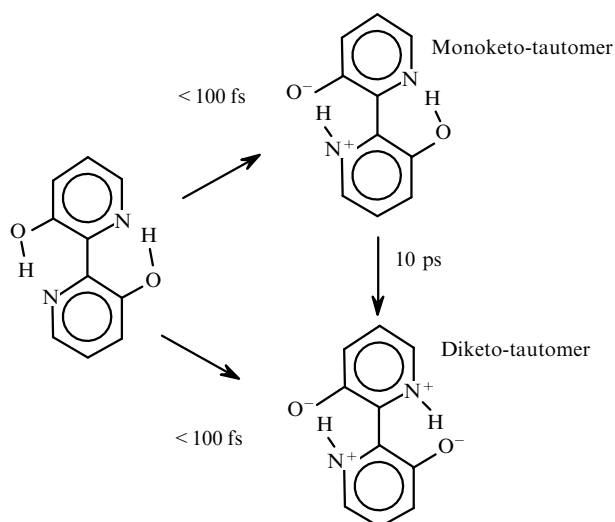


**Figure 4.** Qualitative interpretation of the reaction control using phase characteristics of femtosecond pulses.

We considered above the examples of controlling reactions using multiphoton excitation of molecules in gases. The question arises of the possibility of the reaction control in condensed media. We studied [2,2'-bipyridyl],3,3'-diol in hexane solution. In the ground electronic state, this molecule exists only in a dienol form. Upon excitation of the molecule by femtosecond pulses, the transfer of protons takes place via two channels [22, 23], as shown in Fig. 5. In one of the channels, both protons undergo transfer simultaneously for the time less than 100 fs, resulting in the formation of the so-called diketo-tautomer. In another channel, one proton is transferred for the same time, and monoketo-tautomer is formed. The second proton in this tautomer is transferred for the time about of 10 ps, resulting in the formation of diketo-tautomer as well. This tautomer decays for a few nanoseconds to the ground electronic state.

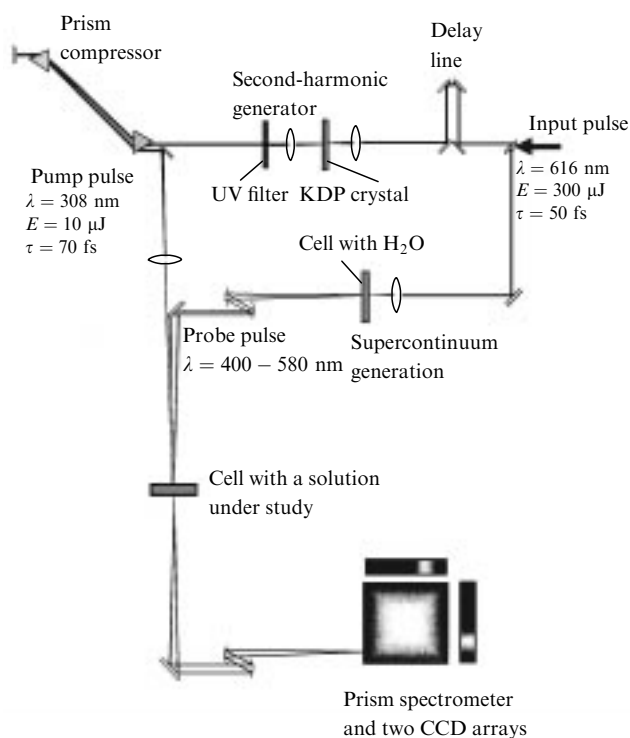
Because these processes occur faster than for 100 fs, it is reasonable to assume that the two-proton transfer is a concerted process. Then, we can expect that the relation between the channels of one- and two-proton transfer can be varied by changing the chirp of the exciting pulse. We studied the dependence of the ratio of initial concentrations of monoketo- and diketo-tautomers on the value and sign of the spectral chirp [24].

The experiments were performed using a setup shown in Fig. 6. A pulse train was divided with a beamsplitter into



**Figure 5.** Scheme of the two-proton transfer reaction in [2,2'-bipyridyl],3,3'-diol.

two trains, one of which was used to prepare a pump pulse and another, to prepare a probe pulse. As a pump pulse, the second harmonic was used at 308 nm. The probing was performed by a supercontinuum pulse, which was generated upon focusing a femtosecond pulse in a cell with distilled water.



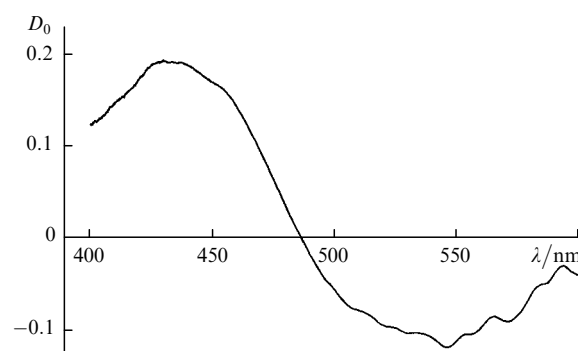
**Figure 6.** Scheme of the experimental setup.

The time delay between the pump and probe pulses was produced with the help of an optical delay line, which provided the variation in the time delay around 50 ps with a step of 1.5 fs. The solution of a substance under study in hexane circulated through a quartz cell of thickness 1 mm.

The pump and probe pulses were focused in a spot inside the cell. A part of the probe pulse was used as a reference pulse, which also passed through the cell, but in the region that was not excited by the pump pulse. After the cell, the reference and probe pulses were directed into a prism spectrometer, where their spectra were detected simultaneously using two CCD arrays.

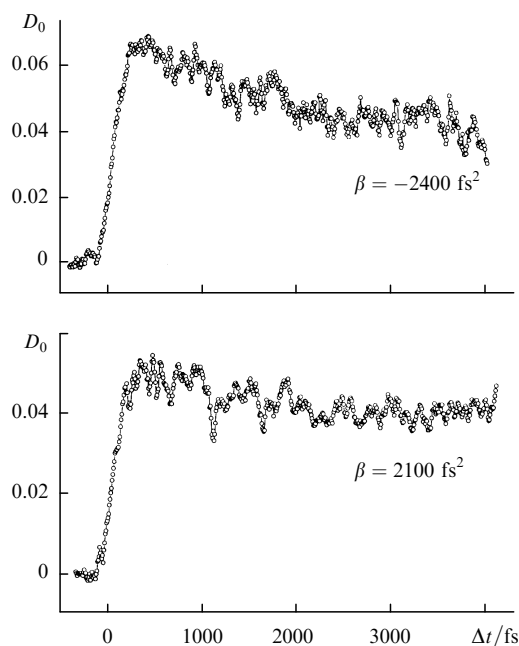
The spectrum of the probe pulse propagating through the excited region of the solution was normalised to the spectrum of the reference pulse and was averaged over ten realisations. A similar procedure was repeated in the absence of excitation. The logarithm of the ratio of the spectra detected upon excitation and without excitation gives the absorption spectrum of intermediate products emerging upon irradiation by the pump pulse for a certain time delay. The chirp of supercontinuum is taken into account by processing the experimental data using the procedure described in Ref. [25].

Fig. 7 shows the spectrum of photoinduced absorption in [2,2'-bipyridyl],3,3'-diol for the 1.5-ps time delay. One can see that absorption is observed in the region from 400 to 480 nm (the optical density  $D_0$  is positive), while in the region from 480 to 600 nm, bleaching is observed (the optical density is negative). The bleaching is caused by stimulated emission of tautomers formed in excited electronic states. The absorption is related to the  $S_1 \rightarrow S_n$  transitions in both tautomers. Note here that as the chirp value is changed, the dynamics of the bleaching and absorption spectra in the 400–480-nm region also changes. This is illustrated by two kinetic curves detected at the probe pulse wavelength of 450 nm for two chirps of the pump pulse, which are approximately equal in magnitude but have opposite signs (Fig. 8).



**Figure 7.** Photoinduced absorption spectrum in [2,2'-bipyridyl],3,3'-diol for the 1.5-ps time delay.

The time evolution of the optical density for delay times  $\Delta t > 4 \text{ ps}$  changes weakly. The variations in the kinetic curves show that the chirp changes the relation between reaction channels. The signal increase observed in the range from 100 fs to 200 fs is caused by the formation of diketo- and monoketo-tautomers in primary reactions. The maximum detectable optical density is proportional to  $\varepsilon_1 C_1 + \varepsilon_2 C_2$ , where  $C_1$  and  $C_2$  are the numbers of monoketo- and diketo-tautomers formed in two primary reaction channels, respectively; and  $\varepsilon_1$  and  $\varepsilon_2$  are the absorption coefficients of the corresponding tautomers. According to the literature and our data, the time evolution of the optical density is



**Figure 8.** Kinetic curves at the wavelength of the probe pulse of 450 nm measured at different values of  $\beta$ .

related to the kinetics of the monoketo–diketo-tautomer transition. This means that a plateau in the kinetic curves characterises the formation of diketo-tautomer only. Then, the measured optical density is proportional to  $(C_1 + C_2)\epsilon_2$ . In this case, the model proposed in Refs [23, 24] predicts that the value of  $1/(1 + C_2/C_1)$  for the positive chirp is two times smaller than that for the negative chirp.

Therefore, the phase characteristics of a femtosecond pump pulse can be used for controlling reactions in liquids. However, it seems that even shorter pulses are required to achieve a more efficient control of the reactions.

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