1) Henceforth, we shall use the atomic system of units.

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Isomerization of molecules under multiphonon vibrational and subsequent electronic excitation by laser radiation

I. N. Knyazev, Yu. A. Kudryavtsev, N. P. Kuz'mina, and V. S. Letokhov

Institute of Spectroscopy, USSR Academy of Sciences, Moscow

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A description is given of the method used to achieve the isomerization of trans-dichloroethylene molecules under multiphoton vibrational excitation with CO_2 laser radiation, followed by excitation to an electronic state by the radiation from an excimer KrF laser. The multiphoton vibrational excitation of molecules results in a shift of the uv absorption band of transi-dichloroethylene and a sharp increase in the rate of isomerization. An examination is made of an isomerization model in which the leading mechanism is a chain reaction involving Cl atoms from the dissociation of dichloroethylene. Experimental results are found to be in satisfactory agreement with this model. Methods of increasing the selectivity of molecular isomerization by laser radiation are discussed.

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1. INTRODUCTION

There has been considerable interest in the processes involved in the selective effect of laser radiation on matter.[1] Discrete processes ensuring highly selective excitation of atoms and molecules have attracted particular attention. As an example of studies in this area, we note the two-step excitation and dissociation of ammonia molecules produced by infrared laser and noncoherent ultraviolet radiation[2] and the multiphoton selective excitation of molecules by infrared laser radiation.[3]

Laser radiation produces a great variety of selective effects in matter.[4] Among them, selective photoisomerization of molecules is of particular interest. Isomerization is a multimolecular process not requiring collisions and is therefore expected to be highly selective.

Until quite recently, the isomerization of molecules was investigated by two traditional methods, namely, thermal excitation from the electronic ground states[6] and excitation by noncoherent radiation to the upper electronic states.[7] Thermal excitation is nonselective whereas the main difficulty in achieving selective isomerization of molecules in the case of noncoherent sources of radiation is that it is difficult to produce high radiation density in a narrow spectral range.

Several methods have been proposed to achieve selective photoisomerization (see Fig. 1). The first approach (Fig. 1a) is based on the multiphoton excitation of molecules in a strong infrared field, so that the energy barrier in the electronic ground state can be overcome.[8,9] The second approach (Fig. 1b) is based on the laser excitation of the lower electronic states of the molecules from which they can undergo transitions to the electronic ground state with a change in the isomeric form. Selective ionization is difficult to achieve in this case because, as a rule, there is a lack of resolved structures in the electronic absorption spectra. The most general approach, which is investigated in the present paper, is the stepwise excitation of molecules illustrated in Fig. 1c.

The radiation from a moderate-intensity infrared laser will excite the molecules of only one isomeric form (because of the sharp difference between the infrared absorption spectra of the isomers) in a multiphoton fashion to a number of vibrational levels. A substantial fraction of the molecules undergoes transitions to the vibrational excited states, and the uv absorption edge of

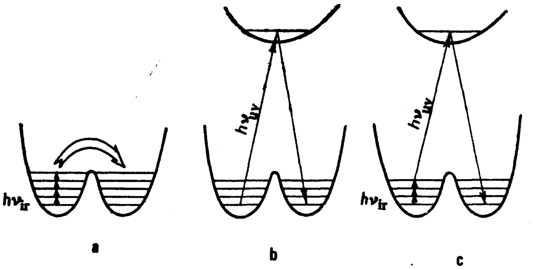

FIG. 1. Possible isomerization schemes.

the molecules shifts toward the red. A uv laser is then used to take these molecules to an electronic excited state from which the molecules execute transitions to the ground state. In this case, the selectivity of the isomerization process for the monomolecular mechanism is completely determined by the infrared excitation channel.

In this paper, we report experiments on the isomerization of 1,2-dichloroethylene under multiphoton excitation of vibrational states by CO_2 laser radiation, followed by the excitation of the molecules to an electronic state by the radiation from an excimer KrF laser. The dichloroethylene molecules were chosen because the CO_2 laser radiation is absorbed by the trans-isomer but not by the cis-isomer,[10] whereas the uv absorption edge of trans-dichloroethylene[11] lies within the generation range of the excimer KrF laser. The power density of the infrared laser beam was chosen so that there was no isomerization of dissociation of dichloroethylene molecules in the presence of infrared radiation alone.

2. EXPERIMENTAL SETUP

General arrangement. The experimental setup (Fig. 2) consists of the ultraviolet and infrared channels, the cell containing the gas under investigation, and the detection system. The procedure was as follows: when the voltage pulse from the triggering unit *14* is applied, the discharge gap P_1 in the supply circuit for the CO_2 laser fires and then the discharge gap P_2 in the photopredissociation unit of the KrF laser fires. Stable synchronization of the CO_2 and KrF lasers is ensured by the optically controlled discharge gap P_3 in the supply circuit feeding the main discharge in the KrF laser. This is achieved by reflecting some of the CO_2 laser radiation from the plate 12 and focusing it with the lens *15* on the discharge-gap electrode. This synchronization scheme ensures that the CO_2 laser pulse leads the KrF pulse by about 100 ± 10 nsec, and the photopredissociation pulse from the excimer laser leads the main discharge by about 0.5–1.0 μsec.

Infrared channel. The vibrational levels of trans-dichloroethylene were excited by the CO_2 laser *13* working at atmospheric pressure with photopreionization. The laser cavity was defined by a copper mirror with a radius of curvature of 10 m and a flat semitransparent dielectric mirror with a reflection coefficient of 80% near 10.6 μ. The laser emission spectrum consisted of the single $P(20)$ line of the 10.6-μ band. The maximum laser pulse energy was 0.3 J for a pulse length of 100 nsec. The laser radiation was focused on the cell containing the gas under investigation by the lens *11* (focal length 35 cm). Teflon films were used to reduce the laser intensity in photoabsorption experiments. The spot area at the center of the lens caustic curve was about 10^{-2} cm².

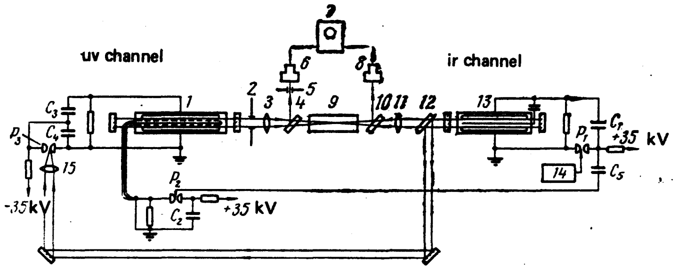

FIG. 2. Experimental setup: *1*—excimer KrF laser; *2, 5*—stops; *3*—quartz lens (f=25 cm); *4, 10*—BaF_2 plane-parallel plates; *6, 8*—FÉK-14 photocell; *7*—oscillograph; *9*—cell; *11*—NaCl lens (f=35 cm); *12*—KRS-5 plane-parallel plate; *13*—CO_2 laser with transverse discharge; *14*—triggering unit; *15*—NaCl lens (f=3 cm).

Ultraviolet channel. The excitation of the molecules to the electronic state was produced by the gas-discharge excimer KrF laser with photopreionization. The laser emission spectrum consisted of the single line at 2485 Å with a spectrum width of about 3 Å. The laser pulse length was 25 nsec and the energy per pulse did not exceed 20 mJ.

In contrast to our previous work on the excitation of gas lasers in a self-sustained discharge,[12-14] in the present experiments, we used photopreionization of the working mixture. The photopreionization of the gas was achieved by discharging the capacitor C_2 into a cable at the end of which the braiding was replaced by 40 copper foil rings in steps of 0.5 mm. The last ring was connected to the core of the cable. The discharge between the rings produced the ionization of the gas in the region of the main discharge. Two such cables located symmetrically relative to the electrodes at a distance of 3 cm from the center of the cell were employed for preionization.

The size of the active region was 60×2×0.4 cm. The 60-cm long electrodes were in the form of brass rods, 18 and 5 mm in diameter (anode and cathode, respectively).

The electrodes were 20 mm apart. The optimum

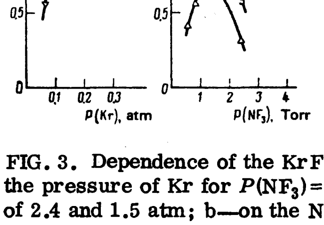

FIG. 3. Dependence of the KrF laser radiation energy: a—on the pressure of Kr for $P(NF_3)$=1.25 Torr and overall pressure of 2.4 and 1.5 atm; b—on the NF_3 pressure for P(Kr)=0.2 atm and overall pressure of 2.4 and 1.2 atm. ○—2.4, ▽—1.5, and △—1.2 atm.

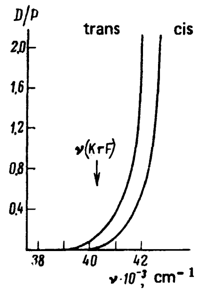

FIG. 4. Ultraviolet absorption spectrum of trans- and cis-dichloroethylene[9]; $D=\log(I/I_0)$, P—pressure in atm, arrow shows the KrF laser generation frequency.

composition of the mixture was determined for this excitation system. Figures 3a and b show the laser energy as a function of NF_3 and Kr pressure for different values of the overall pressure of the mixture. The optimum composition was found to be 1.25 Torr NF_3 + 0.2 atm Kr + 2.2 atm He. The energy was measured with a calibrated thermopile. The working mixture was held in a separate flask for about 24 h. The mixture was allowed to flow slowly through the working chamber while the laser was operating.

The KrF laser cavity was 1 m long and was formed by a dielectric mirror with a radius of curvature of 3 m (reflection coefficient of 95% at 2485 Å) and a plane-parallel quartz plate. The laser radiation was focused on the cell *9* by the quartz lens *3* (focal length 25 cm).

Registration system. In experiments on photoabsorption by dichloroethylene at 2485 Å under infrared excitation, the KrF laser radiation was defined by the 1-mm stop *2* and was directed onto the cell in the region with high infrared intensity. Lens *3* was removed for these experiments. The reference radiation and the radiation transmitted by the cell were reflected by plates *4* and *12* and were finally received by photocells *6* and *8*. Signals from these photocells were fed into the differential amplifier of the S1-15 oscillograph *7*. Equalization of photocell signals in the absence of absorption was performed with the aid of the stop *5*.

In material-production experiments, the stop *2* was removed and the entire KrF laser radiation was focused by lens *3* at the center of the cell with the focal points of lenses *3* and *11* made coincident. The cell was made of glass and had a volume of 40 cm^3 (length 16 cm). The cell was equipped with BaF_2 windows transparent to the CO_2 laser radiation and the radiation from the excimer KrF laser. The relative concentrations of cis- and trans-dichloroethylene were determined from the infrared absorption spectra in the region between 1450 and 600 cm^{-1} before and after irradiation. This was done by freezing the gas into a measuring cell, 10 cm long (volume 15 cm^3), with KBr windows. The change in the concentration of the trans-isomer was estimated from the absorption peak at 1200 cm^{-1} while the corresponding determinations for the cis-isomer were based on the 700 cm^{-1} peak. Measurements based on the stronger peaks at 830, 860, and 900 cm^{-1} were different because of the overlapping of these peaks.

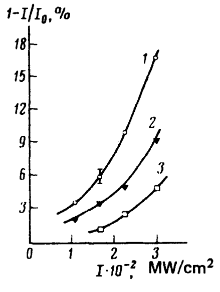

FIG. 5. Absorbed energy as a percentage of ir power density at the focal point of the lens: 1—106 Torr, 2—76 Torr, 3—46 Torr.

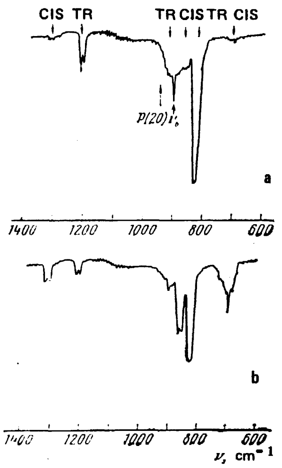

FIG. 6. Ir absorption spectrum of 1,2-dichloroethylene: a—prior to irradiation; b—after exposure to 5400 pulses of CO_2 and KrF laser radiation.

3. RESULTS

Preliminary experiments on the detection of increased uv absorption in the presence of infrared radiation were performed prior to the investigation of stepwise photoisomerization. The arrow in Fig. 4 shows the position of the KrF laser line and the long-wave edge of the uv absorption spectrum of the trans- and cis-isomers, taken from the literature.[11] Figure 5 shows the fraction of absorbed uv energy as a function of the ir power density in the focus of the lens for different gas pressures. It is clear from the figure that the absorbed energy was increased by a factor of 5–8. Because of the small amount of absorption, measurements could be carried out only at high gas pressures at which the processes governing the increase in photoabsorption were, of course, different from processes occurring at the low pressures at which photoisomerization was produced.

Figure 6a shows the infrared absorption spectrum of 1,2-dichloroethylene prior to irradiation. The frequency of the CO_2 laser $P(20)$ line is 944.2 cm^{-1} and corresponds to the R branch of the η_6 band associated with the stretching vibration of trans-1,2-dichloroethylene. The cis-isomer was not excited under CO_2 laser illumination. The initial ratio of trans- and cis-isomers was 0.9:0.1 at an initial pressure of 1.7 Torr.

Exposure to only CO_2 laser radiation produced no changes in the relative concentration of trans- and cis-isomers at maximum laser power and under exposure to 5400 pulses.

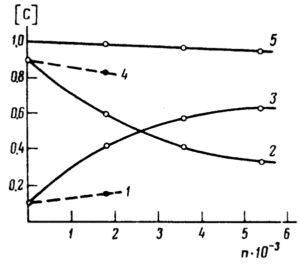

FIG. 7. Concentration C of trans-dichloroethylene (curves 2 and 4) and cis-dichloroethylene (curves 1 and 3) as a function of the number n of laser pulses: ○—exposure to both ir and uv; ●—uv only. Curve 5—resultant concentration of (trans + cis)-dichloroethylene.

When the gas under investigation was exposed to only the KrF laser radiation, a small reduction in the concentration of the trans-isomer and an increase in the concentration of the cis-isomer were observed. Simultaneous exposure of dichloroethylene to the CO_2 and KrF laser radiation pulses produced a sharp increase in the rate of production of cis-isomer from the trans-isomer. Figure 6b shows the infrared absorption spectrum of dichloroethylene after exposure of the gas to 5400 pulses. It is clear that the absorption peaks associated with the cis-isomer in the region of 1300, 860, and 700 cm^{-1} were higher, whereas the absorption peaks due to the trans-isomer in the region of 1300, 900 and 830 cm^{-1} were reduced. Moreover, an absorption peak due to acetylene, C_2H_2, appeared at 730 cm^{-1}.

The points in Fig. 7 show the measured dependence of the concentration of trans- and cis-isomers on the number of laser pulses. The solid lines show the results of calculations. The overall amount of dichlorineethylene at the end of the exposure was reduced by roughly 5%. When the gas was exposed to only the KrF laser radiation (curves 1 and 4), the isomerization rate was lower by a factor of 6.

An important characteristic of the process is the quantum yield. Our experimental data can be used to estimate the quantum yield of the photoisomerization reaction, defined as the ratio of the number of cis-dichloroethylene molecules produced to the number of photons absorbed by the trans-dichloroethylene molecules under exposure to only the KrF laser radiation:

$$\theta=\frac{\Delta N_{\text{cis}}}{\Delta I_{\text{uv}}/h\nu_{\text{uv}}}=\frac{\Delta N_{\text{cis}}}{\sigma^{\text{uv}}_{\text{trans}}N_{\text{trans}}lEn}=20, \qquad (1)$$

where $\sigma^{\text{uv}}_{\text{tr}}=6.25\times10^{-22}$ cm^2 [11] is the absorption cross section of trans-dichloroethylene at 2485 Å, $l=16$ cm is the length of the cell, $E=6.3\times10^{15}$ photons/pulse (5 mJ) is the laser radiation energy, and $n=1800$ is the number of pulses.

4. DISCUSSION OF RESULTS

The observed rate of production of cis-dichloroethylene cannot be explained exclusively in terms of the primary processes occurring during the absorption of laser radiation. This can be seen even from the fact that the quantum yield corresponding to the production of cis-dichloroethylene under absorption of the uv radiation is much less than unity. Secondary processes must, therefore, be taken into account in the interpretation of the results.

Experimental results can be interpreted within the framework of the following model (Fig. 8). Simultaneous exposure to the ir and uv radiation results in the appearance of electronically excited molecules of trans-dichloroethylene, the dissociation of which results in the appearance of chlorine atoms which, in turn, produce the isomerization chain reaction. The formation of the electronically excited molecules of trans-dichloroethylene proceeds through intermediate vibrationally excited states, as follows:

$$\text{trans-}C_2H_2Cl_2+h\nu_{\text{ir}}\rightarrow\text{trans-}C_2H_2Cl_2^{v}, \qquad (2)$$

$$\text{trans-}C_2H_2Cl_2^{v}+h\nu_{\text{uv}}\rightarrow\text{trans-}C_2H_2Cl_2^{*} \qquad (3)$$

and directly from the ground state:

$$\text{trans-}C_2H_2Cl_2+h\nu_{\text{uv}}\rightarrow\text{trans-}C_2H_2Cl_2^{*}. \qquad (4)$$

From the point of view of quantitative description, the most complicated process is the excitation of the molecules in the CO_2 laser field. The process of multiphoton absorption by the trans-dichloroethylene molecule was practically not investigated. We shall suppose that, during the excitation of the molecules to the first electronically-excited singlet state from both the ground and excited vibrational states, the processes governing the subsequent behavior of the molecules are all the same. There are three main channels for the decay of the electronically excited molecules of trans-dichloroethylene[15]:

$$\text{trans-}C_2H_2Cl_2^{*}\rightarrow C_2H_2+Cl_2, \qquad (5)$$

$$\text{trans-}C_2H_2Cl_2^{*}\rightarrow C_2HCl+HCl, \qquad (6)$$

$$\text{trans-}C_2H_2Cl_2^{*}\rightarrow C_2H_2Cl+Cl. \qquad (7)$$

Chlorine atoms produced in the reaction given by (7) play an important role in the process of trans-cis-isomerization.[6,17]

The yield of the chlorine atoms as a fraction of the number of excited $C_2H_2Cl_2^{*}$ molecules when only the processes (5)–(7) are taken into account is approximately 0.08.[15] The chlorine atoms produce a chain reaction through the formation of the vibrationally excited radical $C_2H_2Cl_3^{*}$ [18]:

$$Cl+\text{trans-}C_2H_2Cl_2\rightarrow C_2H_2Cl_3^{*}, \qquad (8)$$

$$Cl+\text{cis-}C_2H_2Cl_2\rightarrow C_2H_2Cl_3^{*}, \qquad (9)$$

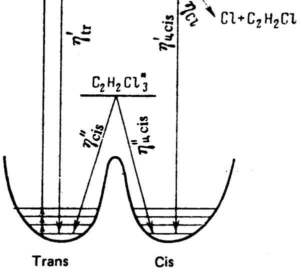

FIG. 8. Leading processes under ir and subsequent uv excitation of trans-dichloroethylene molecules.

which dissociates in a time of 5×10^{-11}–6×10^{-10} sec[18] into trans- and cis-dichloroethylene with probability $\eta''_{\text{trans}}=0.22$ and $\eta''_{\text{cis}}=0.78$.[19] This is accomplished by the release of the chlorine atom which continues the chain:

$$C_2H_2Cl_3^{*}\begin{array}{ll} \xrightarrow{\hspace{4em}} \text{cis -}C_2H_2Cl_2+Cl, & (10)\\ \xrightarrow[\eta''_{\text{trans}}]{\hspace{4em}} \text{trans -}C_2H_2Cl_2+Cl. & (11)\end{array}$$

A contribution is also provided by isomerization during the transition to the excited electronic state:

$$\text{trans-}C_2H_2Cl_2^{*}\begin{array}{ll} \xrightarrow{\eta'_{\text{cis}}} \text{cis -}C_2H_2Cl_2, & (12)\\ \xrightarrow[\eta'_{\text{trans}}]{\hspace{3em}} \text{trans -}C_2H_2Cl_2. & (13)\end{array}$$

This contribution depends on the ratio

$$\eta'_{\text{trans,cis}}\approx W_{\text{dec}}/2W_{\text{diss}}$$

where $W_{\text{dec}}=W_{\text{rad}}+W_{\text{col}}$ is the probability of decay of the excited electronic state through radiative transitions and collisions. The probability $W_{\text{rad}}\approx0.67f/\lambda^2$, where f is the oscillator strength and λ the transition wavelength, is equal to 2×10^8 sec^{-1} for $f\approx0.2$ and $\lambda=2485$ Å. The probability $W_{\text{col}}=N_0\langle\sigma v\rangle$ is approximately 5×10^6 sec^{-1} for $N_0=6\times10^{16}$ cm^{-1} (particle density), $\sigma=3.8\times10^{-15}$ cm^2 [18] (gas-kinetic cross section of dichloroethylene), and $v=2\times10^4$ cm/sec (mean particle velocity). We then have $W_{\text{diss}}\sim4\times10^8$ sec^{-1} since the lifetime of the excited dichloroethylene molecule is 2.4×10^{-9} sec.[20] Hence, $\eta'_{\text{trans,cis}}\approx0.25$.

Trans-dichloroethylene molecules excited to vibrational levels of the ground state by the CO_2 laser participate in uv absorption at a working pressure of 1.7 Torr because the V-T relaxation time in dichloroethylene is much greater than the delay between the ir and uv pulses (100 nsec). The precise value of the V-T relaxation time is not known but can be estimated from the empirical Lambert-Salter rule.[21] Assuming that the frequency of the lower vibrational level of trans-dichloroethylene is 227 cm^{-1},[22] we find that the V-T relaxation time is not less than 1 µsec Torr, and at a working pressure of 1.7 Torr is substantially greater than the time between the ir and uv pulses.

For the quantitative description of the concentrations of cis- and trans-isomers averaged over the period of 1 sec (laser frequency 1 Hz), we have the following set of equations:

$$\dot{N}_{\text{trans}}=-AN_{\text{trans}}+A\eta'_{\text{trans}}N_{\text{trans}}-A\eta_{\text{Cl}}\omega\alpha N_{\text{trans}}^2+A\eta_{\text{Cl}}\omega\eta''_{\text{trans}}N_{\text{trans}}, \qquad (14)$$

$$\dot{N}_{\text{cis}}=-A\eta_{\text{Cl}}\omega\alpha N_{\text{тр}}N_{\text{cis}}+A\eta_{\text{Cl}}\omega\eta''_{\text{cis}}N_{\text{trans}}+A\eta'_{\text{cis}}N_{\text{trans}}, \qquad (15)$$

where $A=\bar{\sigma}_{\text{tr}}lEf/V$ is the effective probability of excitation of the trans-dichloroethylene to the electronic state $C_2H_2Cl_2^{*}$ under the influence of laser radiation, ω is the chain length, i.e., the mean number of $C_2H_2Cl_3^{*}$ radicals produced under initiation by one chlorine atom, V is the cell volume in cm^3, f is the pulse repetition frequency in hertz, and $\alpha=1/(N_{\text{trans}}+N_{\text{cis}})$. The right-hand sides of these equations contain the sums of the rates of removal from and arrival of the molecules in the ground state in all the processes discussed above (see Fig. 6). The first term in (14) describes the rate of removal of trans-dichloroethylene molecules from the electronic ground state by laser radiation, and the second term in (14) and third in (15) represent the rate of formation of cis- and trans-dichloroethylene in the reactions defined by (12) and (13). The third term in (14) and first in (15) describe the rate of loss of molecules due to the formation of the vibrationally excited radical $C_2H_2Cl_3^{*}$ in reactions (8) and (9). The fourth term in (14) and the second in (15) represent the rate of formation of cis- and trans-dichloroethylene in the ground state from the radical $C_2H_2Cl_3^{*}$ in reactions (10) and (11).

The solution of this set of equations can be written in the following form:

$$N_{\text{trans}}(t)=\frac{b}{(b/N_{\text{trans}}(0)+a)e^{bt}-a},$$

$$N_{\text{cis}}(t)=N_{\text{cis}}(0)+N_{\text{trans}}(0)-\frac{b}{(b/N_{\text{trans}}(0)+a)e^{bt}-a}$$
$$+\frac{A}{a}\ln\left\{\frac{b}{N_{\text{trans}}(0)[(b/N_{\text{trans}}(0)+a]e^{bt}-a]}\right\}+\frac{Ab}{a}t,$$

where $a=A\eta_{\text{Cl}}\omega\alpha$, $b=A(1-\eta_{\text{Cl}}\eta''_{\text{trans}}\omega-\eta'_{\text{trans}})$.

The experimental results can be satisfactorily described by the theoretical curves for $a=7.68\times10^{-21}$ sec^{-1}.cm^3 and $b=-8.82\times10^{-5}$ sec^{-1}. The values of a and b correspond to an increase in uv absorption under CO_2 laser excitation by a factor of 6, $\bar{\sigma}_{\text{tr}}=6\sigma_{\text{tr}}^{\text{uv}}$, and chain length $\omega=1000$. The chain length was obtained from the experimentally determined amount of trans-dichloroethylene produced after 1800 pulses under exposure to the uv laser alone. In this case, $\eta_{\text{Cl}}\omega\eta''_{\text{tr}}\gg\eta'_{\text{tr}}$ and the formation of trans- and cis-dichloroethylene from the excited state in reactions (12) and (13) can be neglected. The above calculations of the concentrations of cis- and trans-isomers are therefore in agreement with experiment.

The particular feature of the diochloroethylene molecule is its rapid dissociation in the electronically excited state along several channels, including the release of chlorine atoms. These atoms are responsible for the chain reaction of isomerization of the molecules. The process is essentially selective only up to the second link in the chain. The chain length must be reduced in order to increase the selectivity of the process under these conditions, and this can be achieved by removing the chlorine atoms with the aid of suitable acceptors. This should, however, be accompanied by a sharp reduction in the quantum yield so that, to produce an amount of material sufficient for analysis, one must substantially increase the duration of exposure.

5. CONCLUSIONS

The isomerization of molecules under multistep excitation to electronically excited states under exposure to ir and uv (vuv) laser radiation is a very promising process, capable, at least in principle, of high selectivity. The particularly interesting feature is that, under isotopically selective excitation, the process of separation of isotopes reduces to the very simple separation of isomers. The development of powerful and efficient uv and vuv lasers will facilitate the practical realization of this method of selectively influencing the molecules and its practical application.

It is, however, important to remember the secondary chemical processes which may reduce the selectivity. Complex molecules usually decay into radicals from electronically excited states. It is, therefore, important to select molecules whose decay products have no appreciable effect on the isomerization reaction, or have low decay probability. The following excitation scheme can be used for such experiments. The molecule is first taken to the low-lying vibrational levels of the singlet electronic state and then undergoes transitions to the triplet state, followed by relaxation to the ground state. Molecular isomerization will be the leading process in this scheme.

Apart from its selective effect, multistep excitation of molecules can also be used in other fields, for example, in the spectroscopy of electronically excited states of complex isolated molecules.

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Anomalous Senftleben effect

A. S. Bruev

L. Ya. Karpov Physicochemical Research Institute, Moscow

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Some features of the Senftleben effect are considered for polar molecules. It is shown that, in contrast to the Kagan vector, the use of the Waldmann vector, previously employed in the description of nonspherical collisions in polar gases, is unjustified. It is established that molecular collisions leading to the violation of the principle of detailed balancing are accompanied by a change in the component of the angular momentum along the axis of symmetry of the molecule. The characteristic collision frequencies associated with the nonequilibrium polarization of the angular momenta of the molecules are determined for the CH_3CN gas.

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1. INTRODUCTION

Studies of phenomena such as the change in the transport coefficients in external fields, birefringence in a viscous flow of gas, the thermomagnetic rotation effect, depolarized Rayleigh scattering, NMR and ESR relaxation, and so on,[1] have resulted in an increased interest in the properties of nonspherical scattering by monatomic molecules. The principal aim of such investigations is to obtain information on the frequencies of elastic and inelastic collisions involving a change in the direction of the angular momentum of the molecule. This information is essential for the solution of many applied problems and, in particular, problems such as the development of masers using rotational transitions in molecules and controlled chemical reactions.