

Acceleration of neutral molecular beams and jets by resonant excitation of the molecules by high-power infrared laser radiation in the gas-dynamic expansion zone

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The ability of molecules in an intense resonant infrared (IR) laser field, under conditions of collisional excitation, to absorb a large number of photons can be used to obtain accelerated neutral molecular beams (supersonic jets). A method is described for accelerating pulsed molecular beams using TEA radiation from a CO₂-laser. Results of such an experiment are presented. The method allows one to obtain intense molecular beams with kinetic energies ≈ 1 eV, and can be combined with aerodynamic acceleration. The spectral and energetic characteristics of CO₂-laser-induced acceleration of neutral molecular beams are examined in the case of SF₆. Possible applications are discussed. © 1995 American Institute of Physics.

1. INTRODUCTION

In recent years, molecular (atomic) beams have found a continually expanding range of applications in scientific research work,¹ including high-energy research² in which the kinetic energy of the molecules (atoms) significantly exceeds their thermal energy (≈ 0.1 eV).

Intense beams of neutral molecules (atoms) with kinetic energy from one to a several electron volts are required in various areas of fundamental and applied research¹ (in the study of chemical reactions with energy barriers, the interaction of molecular beams with a surface, modeling of circumterrestrial orbital space, etc.). At the same time, this energy range is the most difficult to work with, and to the present day there is no universal method for obtaining molecular (atomic) beams with kinetic energies in this range.

There exist a few ways of solving this problem.^{2,3} They can be classified according to the process used to control the energy of the molecules: 1) the electrostatic process, which uses neutralization of ionic beams,⁴ and 2) the thermodynamic process, in which the molecular (atomic) beams are extracted from supersonic jets with the help of skimmers. In the first case the kinetic energy of the molecules is controlled by varying the voltage on electrodes placed in the path of the beam. In the second, the kinetic energy is controlled by the temperature T_0 of the gas before it exits the nozzle:

$$\frac{1}{2} m v^2 = \frac{\gamma}{\gamma - 1} k (T_0 - T), \quad (1)$$

where v is the steady-state velocity of the beam, m is the mass of a molecule, $\gamma = C_p/C_v$ is the ratio of the specific heats of the expanding gas, k is the Boltzmann constant, and T is the steady-state temperature.

Thus, at room temperature the kinetic energy of the molecules cannot be increased, with the exception of cases in which the interesting gas is mixed with a light gas—the carrier.^{5,6} This method is not too effective when the ratio of the masses of the interesting gas and the carrier is small. It can be combined with heating of the nozzle to ≈ 3000 K. Such a system allows one, for example, to accelerate argon atoms, diluted with helium, to several electron volts.⁷ How-

ever, this method is also not universal, since at high temperatures molecular dissociation, breakdown of the material of the nozzle, etc. are possible.

Other methods have been found to obtain energetic beams of hydrogen and oxygen atoms. Photolysis of HBr (Refs. 8 and 9) or HI (Refs. 10 and 11) has been used to generate high-energy hydrogen atoms. These methods make use of pulsed lasers and the hydrogen atom fluxes obtained are very low. To obtain high-energy atomic beams, it is also possible to simultaneously combine dissociation and heating of the gas with the help of a discharge³ ignited in the gaseous medium. Microwave,¹² radio frequency,¹³ and other^{14,15} discharges have been successfully used to generate beams of oxygen atoms in order to study chemical reactions. The kinetic energies of the atomic-oxygen beams obtained by these means are ≈ 0.8 eV, and it is quite difficult to control the energy of the atoms.

In Ref. 16 a significant increase in the velocity of a beam of SF₆ molecules was observed when the molecules were irradiated by resonant cw CO₂-laser radiation inside the capillary of the nozzle, which was transparent to IR radiation, directly before the exit opening. The kinetic energy of the laser-excited molecules was ≈ 0.21 eV.

Pulsed and continuous optical discharges^{17,18} have been successfully used^{19–23} to generate high-energy atomic beams. In the method using a continuous optical discharge, plasma is ignited by a pulsed laser or an electric spark, and is maintained by a cw CO₂-laser. The plasma is ignited inside the nozzle directly before the exit opening. Beams of argon atoms with kinetic energy up to 2.55 eV were obtained in Ref. 23 by this method. However, this method is also not universal, and is technically quite difficult.

In Ref. 24 we proposed a method for accelerating intense neutral molecular beams (supersonic jets), based on excitation of the molecules by IR radiation in the gas-dynamic expansion zone at the exit from the nozzle. In the present paper we present a detailed description of the method and present results of a more complete study of IR-laser-induced acceleration of neutral molecular beams.

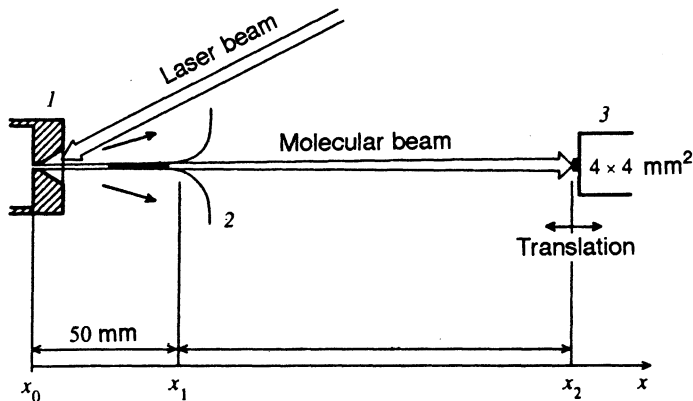


FIG. 1. Diagram of the experimental setup: 1) pulsed nozzle, 2) skimmer, 3) pyroelectric detector.

2. METHOD

The essence of the proposed method of accelerating neutral molecular beams (supersonic jets) consists in the following. The molecules flowing out of the nozzle into a vacuum chamber are excited by high-power resonant IR-laser radiation in the gas-dynamic expansion zone immediately beyond the nozzle. Infrared absorption by the molecules leads to a significant increase in their internal (mainly vibrational) energy.²⁵ At the same time, rapid vibrational-translational ($V-T$) relaxation, effectively transfers energy from the internal degrees of freedom to the translational, as a result of which the molecules are accelerated, including those not absorbing the IR radiation.

Since the number density of the molecules immediately beyond the nozzle is quite high (effective pressure ≥ 100 Torr), the processes of excitation and $V-T$ relaxation are both quite efficient.

3. EXPERIMENT

A diagram of the experiment is given in Fig. 1. The main parameters of the setup used in the pulsed-molecular-beam experiments are described in detail in Refs. 26 and 27. Here we present only a brief description.

In the experiments we used a pulsed "current loop nozzle",²⁸ working at room temperature. The diameter of the opening of the nozzle was 0.75 mm. The opening time was $\approx 60 \mu\text{s}$ (at half maximum). The gas pressure in the nozzle varied within from ≈ 0.2 to 5 atm. The nozzle itself was made from duraluminum. The nozzle was cut in the shape of a cone with opening angle 60 deg. The length of the cone was 15 mm. The vacuum chamber in which the molecular beam was formed was pumped down to a pressure of $\approx 1 \cdot 10^{-6}$ Torr. The nozzle operated with a frequency of 0.2 Hz. To extract the molecule beam from the jet, we used a conical diaphragm (skimmer) with an opening diameter of 1.5 mm, which was placed 50 mm from the cut of the nozzle.

The molecules were excited^{26,27} by a pulsed TEA CO_2 -laser, which was tunable with the help of a diffraction grating. The laser generated pulses with energy up to 3 J. The laser pulse consisted of a peak width with half-maximum ≈ 100 ns, and a tail, of duration $\approx 0.6 \mu\text{s}$, that contained $\approx 50\%$ of the total energy. To avoid optical breakdown inside the cone of the nozzle, the energy was lowered to ≈ 0.1 J through separation of the TEM_{00} mode and additional attenu-

ation with sheets of CaF_2 . The laser radiation was directed into the gas-dynamic expansion zone without focusing. The nozzle, CO_2 -laser, and detecting system were powered by a GI-1 generator. The time delay between the nozzle pulse and the CO_2 -laser was set by means of a G5-54 pulse delay generator.

The molecule beam was detected^{26,27} with the help of a pyroelectric receiver with temporal resolution $\approx 3-5 \mu\text{s}$. The receiver is described in detail in Ref. 29. The receiver could be translated along the beam axis with the help of a mechanical device. This allowed us to obtain time-of-flight spectra of the molecules at various points along the beam axis²⁷ and directly measure the most probable velocity of the molecules (at the maximum of the time-of-flight distribution). The signal from the receiver was amplified ($\times 100$) and then fed to the input of a S9-16 digital oscilloscope. The receiver measured the energy of the molecules spreading out inside a solid angle defined by the dimensions of the active element of the receiver ($4 \times 4 \text{ mm}^2$) and the distance $x_0 x_2$ from the cut of the nozzle to the detector.

Without preliminary excitation of the molecules, the signal S_0 from the detector is proportional to the quantity²⁷

$$S_0 \propto nv \left(E_a + E + \frac{mv^2}{2} \right) \equiv nvE_0, \quad (2)$$

where n is the number density of the molecules at the surface of the detector, E_a is the heat of adsorption per molecule, and E is the energy of a molecule (sum of vibrational, rotational, and "local" translation energy).

In the case in which the beam molecules are excited by a laser pulse, the signal S_L is proportional to the quantity^{26,27}

$$S_L \propto nv(E_0 + E_{ab}), \quad (3)$$

where E_{ab} is the energy absorbed by the molecule from the laser field.

When the beam molecules are excited far from the nozzle ($x \geq 50$ mm), where collisions, and consequently the $V-T$ relaxation process do not take place, the velocity of the excited molecules does not differ from that of the unexcited ones.^{26,27} If, on the other hand, the molecules are excited in the gas-dynamic expansion zone, where the collision frequency is large, then as a result of the $V-T$ relaxation process the absorbed energy is either completely or partially

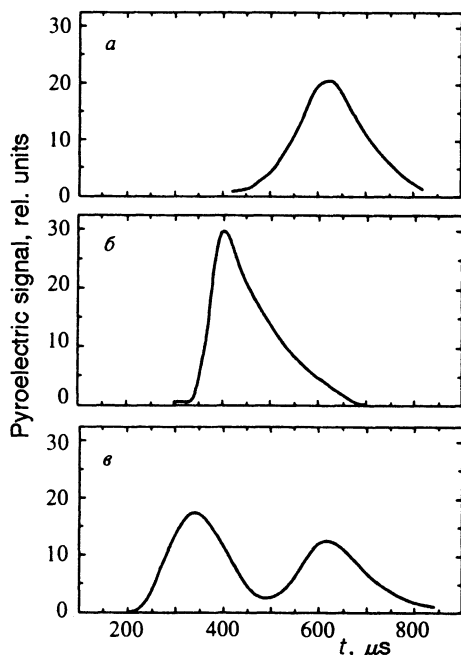


FIG. 2. Time-of-flight spectra of SF₆ molecules without laser, acceleration (a) and with laser acceleration (b,c). In case (b) all of the molecules in the beam are accelerated; in case (c) only the molecules leaving the nozzle at the initial moment are accelerated. The distance from the nozzle to the receiver is 25 cm. The SF₆ pressure in the nozzle is 4 atm. Energy of the CO₂-laser: b) 0.1 J, c) 0.05 J, at the IOP(20) line.

channeled into the translational degrees of freedom, and acceleration of the molecules takes place, including those that have not absorbed IR radiation.

4. RESULTS AND DISCUSSION

In the experiments we measured the time-of-flight spectra of the beam molecules at various distances from the nozzle and determined the most probable velocity of the molecules in unaccelerated and accelerated beams. The experiments were carried out with molecules of SF₆, CF₃I, NH₃, CF₂HCl, both with and without carriers (H₂, D₂, N₂, Ar, CH₄). Figures 2a–c present time-plots of signals from the detector (time-of-flight spectra of the molecule SF₆) without laser acceleration (a), and with laser accelera-

tion (b, c). The molecules were excited on the line IOP(20) of the CO₂-laser (944.2 cm⁻¹), which is closely in resonance with the ν₃ vibration of the molecule.^{26,30} In Fig. 2b the delay between the pulses leaving the nozzle and the pulses leaving the laser was chosen (t_d = 120 μs) to accelerate all molecules in the beam. In Fig. 2c the delay was shortened to 60 μs in order to accelerate only those molecules that escape the nozzle in the first moment, and leave the others unaccelerated. When, on the other hand, the delay is increased (τ_d = 180 μs) and the laser pulse excites the molecules leaving the nozzle at the last moment, the accelerated molecules catch up with and overtake the unaccelerated molecules.

The most probable velocity of the SF₆ molecules without laser acceleration was v₀ = (470 ± 10) m/s, which corresponds to a kinetic energy of E_{kin}⁰ ≈ 0.17 eV, while the velocity of the accelerated molecules was v_L = (815 ± 15) m/s, which corresponds to a kinetic energy of E_{kin}^L ≈ 0.51 eV. In combination with aerodynamic acceleration of SF₆ molecules in a mixture with methane (SF₆:CH₄ = 1:10, total pressure p_Σ = 1 atm at the nozzle) we obtained v₀ ≈ 1000 m/s and v_L ≈ 1200 m/s, which corresponds to a kinetic energy of the SF₆ molecules of E_{kin}^L ≈ 1.0 eV.

In analogous experiments with CF₃I (nozzle pressure was p₀ = 1 atm, excitation was produced in the ν₁ vibrational band of the molecule³¹ by the 9R(12) line of the CO₂-laser at frequency 1073.3 cm⁻¹) the velocity of the molecules without laser acceleration was v₀ = (415 ± 10) m/s (E_{kin}⁰ ≈ 0.18 eV), and with acceleration the kinetic energy was E_{kin}^L ≈ 0.74 eV. In combination with aerodynamic acceleration for the CF₃I in mixture with methane (CF₃I:CH₄ = 1:15, p_Σ = 1.3 atm) the most probable velocity of the molecules without laser acceleration was v_L = (1065 ± 20) m/s, which corresponds to a kinetic energy of the CF₃I molecules of E_{kin}^L = 1.2 eV.

Results of our experiments on laser acceleration of molecular beams are displayed in Table I. Note that the efficiency of the laser-induced acceleration depends both on the amount of energy absorbed from the laser field and on the V–T relaxation rate. Polyatomic molecules should be accelerated more efficiently by this method, since a large amount of energy can be deposited in the internal degrees of freedom of such molecules by way of multiphoton IR absorption.²⁵ Besides, as the absorbed energy increases, the V–T relax-

TABLE I. Results on laser-induced acceleration of molecular beams.

Composition of gas	Nozzle pressure (atm)	Mean flow velocity (m/s)		Kinetic energy of molecules (eV)		CO ₂ -laser line
		v ₀	v _L	E _{kin} ⁰	E _{kin} ^L	
SF ₆	4.0	470	815	0.17	0.51	10P(20)
SF ₆	1.5	455	800	0.16	0.50	10P(20)
SF ₆ +H ₂ (1:10)	2.0	1220	1530	1.15	1.80	10P(20)
SF ₆ +D ₂ (1:10)	1.1	1170	1450	1.05	1.60	10P(20)
SF ₆ +N ₂ (1:12.5)	1.0	790	890	0.48	0.61	10P(20)
SF ₆ +Ar (1:15)	1.3	650	750	0.32	0.42	10P(20)
SF ₆ +CH ₄ (1:10)	1.0	980	1165	0.74	1.05	10P(20)
CF ₃ I	1.0	415	845	0.18	0.74	9R(12)
CF ₃ I+CH ₄ (1:15)	1.3	815	1065	0.70	1.19	9R(16)
CF ₂ HCl	1.0	600	720	0.16	0.24	9R(30)
NH ₃ +CH ₄ (1:1.5)	1.2	1040	1420	0.09	0.18	9R(30)

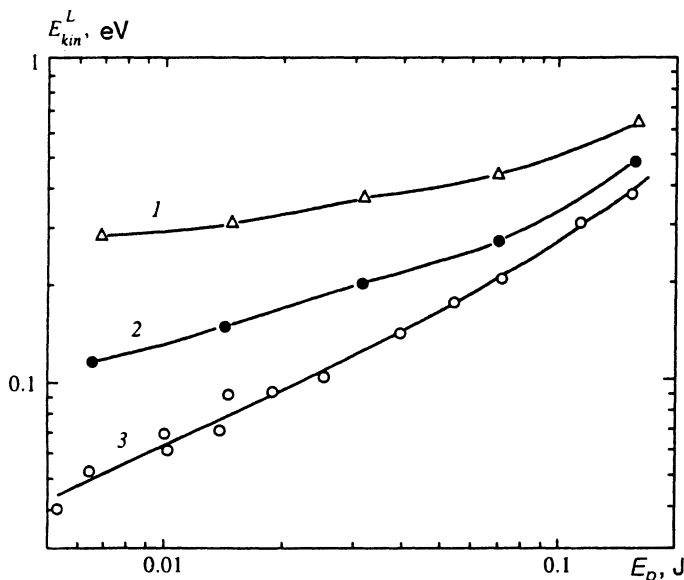


FIG. 3. Dependence of the kinetic energy of the accelerated SF₆ molecules on the energy of the exciting CO₂-laser pulse: 1) total kinetic energy of an SF₆ molecule, 2) kinetic energy induced by the laser. The SF₆ pressure in the nozzle is 2.5 atm. The laser line IOP(16). For comparison, curve 3 plots the dependence of the energy absorbed by the SF₆ molecules (in arb. units) on the energy density of the exciting pulse (in J/cm², numerical values correspond to the labelling of the axis in joules), obtained in Ref. 34 at T≈300 K and SF₆ pressure 0.15 torr.

ation rate grows substantially. Thus, for example, for SF₆ we have $p\tau_{V-T} \approx 122 \mu\text{s} \cdot \text{torr}$ in a weak IR field and $p\tau_{V-T} \approx 22 \mu\text{s} \cdot \text{torr}$ in the field of a CO₂-laser with energy density $\Phi \approx 0.5 \text{ J/cm}^2$ (see Ref. 32 and the citations therein).

From the table it is clear that SF₆ and CF₃I are quite efficiently accelerated even without a carrier. This, undoubtedly, is a result of the multiphoton nature of the absorption of these molecules^{30,31} i.e., the absorption of a large number of photons per molecule, and the significant increase of the V-T relaxation rate of the highly excited molecules.³² The acceleration of the CF₂HCl molecules is rather inefficient, mainly because the frequency of the exciting CO₂-laser pulse is quite strongly (by more than 30 cm⁻¹) detuned from the center of the absorption band of the molecules,³³ as a result of which efficient excitation of the molecules is hindered. In the case of small molecules (e.g., NH₃, etc.), which do not undergo multiphoton absorption, the efficiency of laser acceleration depends mainly on the rates of V-T and rotational acceleration. The latter is important for the entrainment when a large fraction of the molecules interact with the laser pulse. Rapid rotational relaxation and the cyclic nature of the interaction of the molecules with the laser radiation (rapid V-T relaxation) can ensure efficient acceleration of the molecules.

Figure 3 plots the dependence of the kinetic energy of SF₆ molecules on the energy of the exciting pulse in the case of pumping of the SF₆ molecules at the IOP(16) line of the CO₂-laser, which coincides with the Q-branch of the transition $\nu_3 = 1 \leftarrow \nu_3 = 0$ of the ν_3 vibration of the molecule.^{26,30} Curve 1 gives the total kinetic energy of the molecules in the beam, and curve 2, the energy induced by the laser. Curve 3 gives, for comparison, the dependence, obtained in Ref. 34 of the energy absorbed by the SF₆ molecules (in arbitrary units) on the energy density of the exciting pulse (in J/cm², in the same numerical scale as indicated for the energy variable) in the case of exciting SF₆ molecules in a cuvette at room temperature and a pressure of 0.15 torr. The values of the energy density in our experiments were probably only insignificantly higher than in Ref. 34.

The absorbed energy is equal to the product of the frac-

tion of excited molecules q and the mean level of excitation ε_q ($E_{ab} = q\varepsilon_q$).³⁵ Since in our case the number density of molecules in the excitation band is quite large and the rotational relaxation time is less than the duration of the laser pulse, we have $q \approx 1$ (Ref. 35). Consequently, in our case, as the pump energy rises only the level of vibrational excitation grows. In the case of curve 3, as the pump energy density rises, both the fraction of excited molecules and the mean level of excitation grow. For this reason, curve 2 is less steep than curve 3.

It is clear from Fig. 3 that the laser-induced kinetic energy E_{kin}^L increases with the energy of the exciting pulse E_p in the energy range under study roughly as $\sqrt{E_p}$, without saturation. Absorption by SF₆ molecules behaves almost identically.^{26,30} Similar results were also obtained²⁴ in the case of excitation of SF₆ at the IOP(20) line of the CO₂-laser. We can therefore assume that at higher energy densities of the exciting pulse (which are simple to achieve with a conical nozzle²⁷) and gas pressures in the nozzle it is possible to obtain molecular beams with kinetic energy >1 eV. Indeed, the dissociation energy of the majority of molecules lies in the region 2–5 eV. Consequently, even without taking account of the cyclic nature of the interaction, the molecules can absorb energy from the IR field comparable with the dissociation energy. If we take cyclic nature of the interaction into account, which must be done when the vibrational-translational relaxation time is much less than the duration of the exciting pulse ($\tau_{V-T} \ll \tau_p$), the total energy absorbed by the molecule can significantly exceed the dissociation energy.

In the example of an SF₆ molecule beam, we also examined the spectral characteristics of the acceleration. Figure 4 plots the dependence of the laser-induced kinetic energy of the SF₆ molecules on the frequency of the excitation, for laser pulse energy $E_p = 0.12 \text{ J}$ and SF₆ pressure $p = 2.5 \text{ atm}$ in the nozzle. For comparison, this figure also plots the dependence of the mean number of absorbed photons per molecule $\langle n \rangle$ on the frequency at energy density $\Phi \approx 0.12 \text{ J/cm}^2$

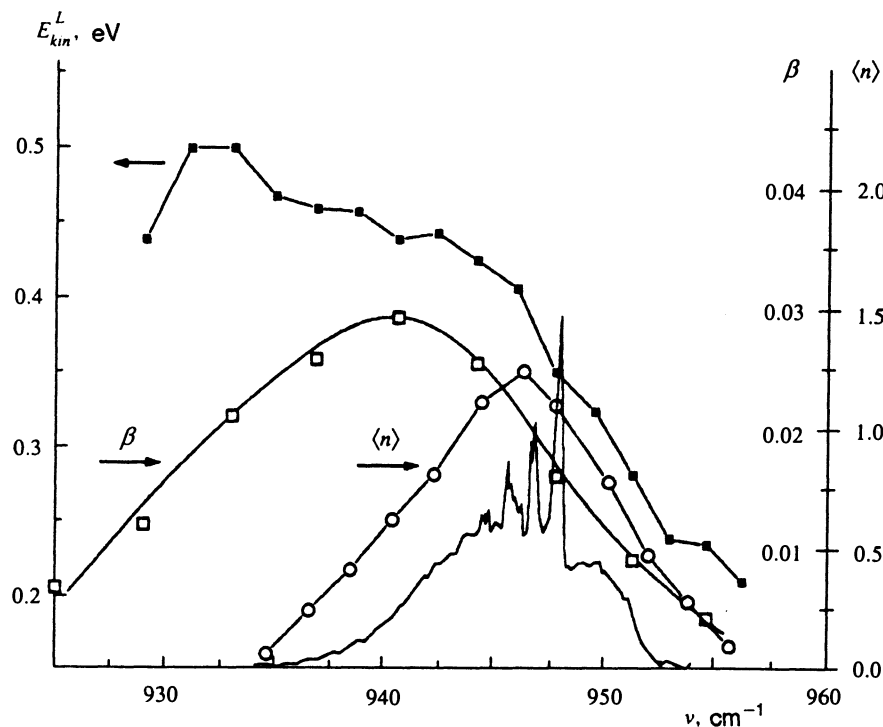


FIG. 4. Dependence of the laser-induced kinetic energy of the SF₆ molecules on the frequency of the exciting radiation. For comparison, also plotted are the frequency-dependences of the mean number of photons absorbed per molecule $\langle n \rangle$, (Ref. 36, SF₆ pressure 0.45 torr) and dissociation yield β of the SF₆ molecules (Ref. 37, SF₆ pressure 0.2 torr, energy density of the exciting pulse $\Phi \approx 3 \text{ J/cm}^2$). The linear absorption spectrum of the ν_3 vibration of SF₆ is shown below.³⁸

and SF₆ pressure $p=0.45$ torr, taken from Ref. 36, the frequency dependence of the dissociation yield β at $\Phi \approx 3 \text{ J/cm}^2$ and $p=0.2$ torr, taken from Ref. 37, and also the linear absorption spectrum of the ν_3 vibration of the SF₆ molecules at room temperature, taken from Ref. 38.

In Fig. 4 one can clearly see that the function $E_{\text{kin}}^L(\nu)$ is wider than the profiles of $\langle n \rangle(\nu)$ and $\beta(\nu)$. This is not remarkable, since in our case the number density of the molecules (the effective SF₆ pressure) in the excitation band was substantially higher ($p \approx 100$ Torr) than in Refs. 36 and 37 ($p \leq 0.5$ torr), and for this reason the spectral characteristics of the excitation are less pronounced. The strong shift of the maximum in $E_{\text{kin}}^L(\nu)$ toward the red is due to the anharmonicity of the vibrations of the molecules:³⁹ if the pump frequency is shifted toward the red, all of the higher-lying states of the molecule are excited^{25,40} (i.e., the mean level of vibrational excitation grows³⁵). As a result, the $V-T$ relaxation rate also grows,³² and, consequently, also the efficiency of laser acceleration of the molecules.

The local maximum in the region 930 cm^{-1} is due to the fact that the absorption band of the other isotopic component of sulfur hexafluoride, ³⁴SF₆ (Ref. 30), is located in the region, the fraction of which in a natural isotopic mixture of SF₆ is around 4.2%. The absorption band of the ν_3 vibration of molecules of ³⁴SF₆ is shifted by 17 cm^{-1} toward the red relative to the absorption band of ³²SF₆ molecules.⁴¹

We also observed efficient acceleration of an SF₆ beam when the molecules were excited in the absorption band of the combined vibration $\nu_2 + \nu_6$ ($\approx 990 \text{ cm}^{-1}$, Refs. 42 and 43). The intensity of this band is roughly two orders of magnitude less than that of the vibrational band ν_3 (Ref. 43).

Let us estimate the intensity of the molecular beams which we have obtained. The accelerated beams are less intense than the unaccelerated ones since the radial velocity of the molecules in the accelerated beams is also higher. The

intensity of the unaccelerated molecular beams can be estimated if we know the total number of molecules N leaving the nozzle in each pulse, and the time-of-flight distribution of the molecules in the beam. In the case of SF₆ molecules, as was shown in Ref. 26, $N \approx 10^{17}$ for a nozzle pressure $p_0 = 5$ atm. The width (at half-maximum) of the time-of-flight distribution of the SF₆ molecules 25 cm from the nozzle (at the surface of the detector) is $\approx 150 \mu\text{s}$ (see Fig. 2a). If we assume that all the molecules leaving the nozzle are distributed inside the solid angle defined by the opening angle of the nozzle cone ($d\omega \approx 1 \text{ sr}$), then in our case the peak intensity of the unaccelerated beam 25 cm from the nozzle is $\approx 10^{21}$ molecules/sr·s. The intensity of the accelerated beam is somewhat less.⁴⁴ These intensities are quite high, in particular, for accelerated molecular beams.^{1,2}

V. CONCLUSION

In conclusion, let us summarize the main results and conclusions of our work.

A method has been proposed for accelerating intense neutral molecular beams (gas-dynamic jets), based on exciting the molecules with high-power resonant IR laser radiation in the gas-dynamic expansion zone just beyond the nozzle.

Intense molecule beams of SF₆ and CF₃I have been obtained, with kinetic energies $\approx 0.64 \text{ eV}$ and 0.74 eV , respectively, without a carrier gas, and $\approx 1.0 \text{ eV}$ and 1.2 eV , with the carrier gas CH₄.

For the SF₆ beams, we investigated the spectral and energetic characteristics of IR-laser-induced acceleration of the molecules:

1. We examined the dependence of the laser-induced kinetic energy of the SF₆ beam molecules on the frequency of the exciting pulse. We showed that the profile of the depen-

dence $E_{\text{kin}}^L(\nu)$ is wider than the profiles of the absorbed energy and the dissociation yield ($\langle n \rangle(\nu)$ and $\beta(\nu)$, respectively).

2. We examined the dependence of the laser-induced kinetic energy of the SF₆ molecules on the energy of the exciting pulse. We showed that the kinetic energy E_{kin}^L of the SF₆ molecules grows with increase of the pulse energy E_p in the energy range under study as $\sqrt{E_p}$, almost identical to the result for the absorbed energy.

Since molecules in an intense IR laser field can absorb energy comparable with or greater than their dissociation energy (2–5 eV) under collisional conditions of excitation, it can be assumed that this method can be used to obtain intense molecular beams with kinetic energies of from one to several electron volts.

It is easy to control the energy of the accelerated molecular beams by varying the energy, i.e., frequency, of the exciting laser radiation.

As a possible application of such beams, we may consider experiments with intersecting molecular beams examining the role of the internal and kinetic energies of the molecules in chemical reactions, and experiments on the interaction of molecular beams with a surface, including collisionally induced molecular dissociation,⁴⁵ in which a study of the role of the kinetic energy, along with the internal energy, is of interest.

As one practical application of the method, we may consider the possibility of powering rockets.

It should be noted that there also is interest in the possibility of accelerating molecular beams and jets by exciting the molecules with the radiation of high-power cw IR lasers.

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