eous burnout of the inverted population can lead to development of radiation pulsations, in agreement with the large accumulated experimental material,[12] and is confirmed by theoretical calculations.

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Quasiclassical calculation of transition probabilities

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A simple general formula is derived for the probability of an allowed transition between quasiclassical states.

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It is well known that the calculation of a transition probability between quasiclassical states reduces to the calculation, by the method of steepest descents, of matrix elements of the perturbation potential. If the transition is classically forbidden, the saddle point lies in the complex plane and the result depends on details of the analytic behavior of the potential in which the quasiclassical motion occurs (Ref. 1, Sec. 51). If the transition is forbidden, simple general formulas can be derived; the present note deals with this case.

We shall calculate the square of the absolute value of the matrix element

$$\varphi_{nn'}(q)=\int\varphi_n(x)e^{iqx}\varphi_{n'}(x)\,dx \tag{1}$$

with the quasiclassical functions

$$\varphi_n(x)=\frac{c_n}{\sqrt{p_n}}\cos\left(\frac{1}{\hbar}\int_a^x p_n dx-\frac{\pi}{4}\right),\quad p_n=\{2m[\varepsilon_n-U(x)]\}^{1/2}, \tag{2}$$

where $U(x)$ is the potential in which the quasiclassical motion of a particle with mass m occurs, ε_n is an energy eigenvalue, a is the position of one of the classical turning points, and c_n is a normalization constant. We write the integral (1) in the form of a sum

$$\varphi_{nn'}(q)=\frac{c_nc_{n'}}{4}\sum_{s_1,s_2=\pm1}\int\exp\left[if(x)-i\frac{\pi}{4}(s_1+s_2)\right]\frac{dx}{(p_np_{n'})^{1/2}}, \tag{3}$$

where

$$f(x)=\frac{1}{\hbar}\int_a^x(s_1p_n+s_2p_{n'})\,dx+qx. \tag{4}$$

In the sense of the quasiclassical approximation, $f(x)$ is a rapidly varying function. Therefore in calculating the integrals in Eq. (3) we can use a steepest descent (or stationary phase) method. The position of the saddle points is given by the equation

$$\hbar f'(x_l)=s_1p_n(x_l)+s_2p_{n'}(x_l)+\hbar q=0,$$

which expresses the law of conservation of momentum. For allowed transitions all of the x_1 are real. The result of the calculations is

$$|\varphi_{nn'}(q)|^2=\pi\frac{|c_nc_{n'}|^2}{8}\sum_{s_1,s_2=\pm1}\sum[|f''(x_l)|p_n(x_l)p_{n'}(x_l)]^{-1}. \tag{5}$$

Differentiating the quasiclassical quantization condition with respect to n and comparing the result with the well known expression for the normalization constant (Ref. 1, Sec. 48), we obtain the relation

$$|c_n|^2=\frac{2m}{\pi\hbar}\frac{d\varepsilon_n}{dn}. \tag{6}$$

Equations (5) and (6) give the required result. This expression can be written in a more intuitive, and in some cases much more convenient form:

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$$|\varphi_{nn'}(q)|^2=\frac{1}{2\pi\hbar}\frac{d\varepsilon_n}{dn}\frac{d\varepsilon\,'}{dn'}\int dx\,dp_1\,dp_2\,\delta(p_1+p_2+\hbar q)$$
$$\times\delta\left(\varepsilon_n-\frac{p_1^2}{2m}-U(x)\right)\delta\left(\varepsilon_{n'}-\frac{p_2^2}{2m}-U(x)\right). \qquad (7)$$

To verify that (5) and (7) are identical, one needs only to perform the integrations in Eq. (7) first over the momenta p_1 and p_2 and then over the coordinate x. The expression (7) is especially convenient in calculating sums over the states n or n', which in the classical limit reduce to integration over an energy, which can be performed easily by means of a δ function.

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Dependence of the rate of vibrational relaxation of CH_3F on the equilibrium and vibrational temperatures

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The double infrared resonance method was used to investigate the rates of vibrational–translational (V–T) relaxation of CH_3F (by interaction with argon) as a function of the equilibrium and vibrational temperatures. It was established that the vibrational temperature increased the V–T relaxation rate more effectively than did the translational temperature because the relaxation from the upper vibrational levels was fast.

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The CH_3F molecule is one of the most slowly relaxing polyatomic molecules. Therefore, it is a convenient object for investigating the influence of the vibrational relaxation on its reactivity.[1] In such investigations one requires data on the rate of the vibrational–translational (V–T) relaxation of the CH_3F molecules interacting with various gases in a wide temperature range. However, the published rate constants of the relaxation processes are available only for room temperature.[2] Moreover, as shown in Ref. 3, the V–T relaxation rate of CH_3F depends on the populations of the vibrationally excited states even for cw infrared radiation intensities of ~10 W/cm^2. Similar dependences have been observed earlier in the case of pulsed excitation of SF_6 (Ref. 4) by CO_2 laser radiation of 10–10^5 W/cm^2 intensity, i.e., at high degrees of occupancy of the vibrational levels. The present paper describes an investigation of the temperature dependence of the rate of the V–T relaxation of CH_3F under uniform heating conditions and also in the case of vibrational heating at a fixed translational temperature.

EXPERIMENTAL METHOD AND RESULTS

The double infrared resonance method was used to determine the vibrational relaxation time. The apparatus was basically the same as that described in Refs. 5 and 6. A mixture of gases was admitted to a glass cylindrical cell, which was 80 cm long and had an internal diameter of 2.5 cm; the windows were made of KBr. The cell was placed inside an oven 70 cm long where the temperature was kept constant to within +3°. Two laser beams passed along the cell axis. The probe beam was provided by an He–Ne laser tuned by a magnetic field near the $\lambda = 3.39\ \mu$ wavelength. Molecules were excited either by a Q-switched CO_2 laser (E~1 mJ, $\tau = 1$–$3\ \mu$**sec**) or by a cw CO_2 laser with an output power of ~10 W. The cw CO_2 laser beam was interrupted in a time of ~10 μsec at a frequency of 80 Hz by a chopper placed at the focus of the telescope. Before entering the cell the beam was attenuated (if necessary) by a polarizer. Use was made of the $P(20)$ laser line due to the 00^01–02^00 transition, coinciding with the $Q(12, 2)$ line of $^{12}CH_3F$ and the $P(32)$ line of the same laser transition coinciding with the $R(4, 3)$ line of $^{13}CH_3F$.

The signal was recorded with a Ge:Au photoresistor. The signal/noise ratio was improved by the use of a 64-channel digital store with response up to 1 μsec/channel. The stored signal was used to drive a chart plotter.

The temperature dependences of the relaxation constants of CH_3F interacting with argon and helium were determined with the probe laser beam tuned to an absorption line of $^{12}CH_3F$ in a field of 380 G (the absorption spectra of $^{12}CH_3F$ and $^{13}CH_3F$ in the tuning range of the He–Ne laser were reported in Ref. 6). After a CO_2 pulse there was an increase in the intensity of the probe laser beam and this was followed by exponential relaxation. Figure 1 shows a typical signal and its semilogarithmic anamorphosis. The slope of this anamorphosis can be used to find the vibrational relaxation time. Other figures give the dependences of the reciprocal of the vibrational relaxation time on the argon (Fig. 2) and helium (Fig. 3) pressures obtained at various tempera-

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