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NONEQUILIBRIUM DISSOCIATION OF A MOLECULAR GAS UNDER THE INFLUENCE OF RESONANT LASER RADIATION, WITH ALLOWANCE FOR VIBRATION-VIBRATION COLLISIONS

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1. With the appearance of powerful lasers, new possibilities were uncovered for acting on intermolecular processes, namely excitation and dissociation of molecules, initiation and control of chemical reactions, etc. Research on the excitation and dissociation of isolated molecules in a laser field is the subject of a number of papers [1 - 5]. In [1] is considered the interaction between a molecule and a field in the case when the acting force either coincides with the field frequency and resonant transitions are then possible through several levels, or in the case when the acting force is non-linear in the field, and different field harmonics take part in the excitation. The probabilities of two- and three-quantum dissociation of a molecule were calculated in [2, 5]. Reference [4] is devoted to excitation of a Morse oscillator. In all the cited papers, the threshold density of the laser radiation was $10^{14} - 10^{16}$ W/cm².

Dissociation and chemical reactions in molecular gases, induced by resonantly absorbed infrared laser radiation, were experimentally investigated in [6]. The threshold fluxes were of the order of 10^2 W/cm². Physically this circumstance, unlike in [1 - 5], is due to selective interaction of the radiation with one of the vibrational degrees of freedom of the molecule and with vibration-vibration relaxation of the excitations. In this case the vibrational temperature can noticeably exceed the translational temperature of the medium, as was pointed out, for example, in [7].

In the present paper we solve the nonstationary self-consistent problem of dissociation of a molecular gas under the influence of resonant laser radiation under conditions when the duration of the pulse is limited only by the time t_m of the vibration-translation relaxation. It is assumed that the considered vibrational branch of the molecule is almost harmonic for vibrational exchange up to levels from which dissociation proceeds with a probability close to unity. On the other hand, the anharmonicity is always important for the absorbed laser radiation, so that absorption is possible only between the first two levels of the molecule.

2. We assume that the vibrational energy exceeds the quantum energy $h\omega$ of the harmonic oscillator. In this case the time dependence of the molecule energy distribution function $F(t, \epsilon)$ (ϵ is the energy and t is the time) can be described by a kinetic equation in the diffusion approximation:

$$\frac{\partial \Phi}{\partial t} = - \frac{\partial J}{\partial \epsilon},$$

$$J = -\epsilon \left[\Phi + (1 + \nu(\epsilon - \epsilon_*)) \frac{\partial \Phi}{\partial \epsilon} \right]. \quad (1)$$

Here

$$\Phi = \theta F / N_0, \epsilon = \mathcal{E} / \theta, \tau = t / t_c, \theta = \frac{G}{\int_0^{\epsilon^*} \mathcal{E} F d\mathcal{E}} / \int_0^{\epsilon^*} F d\mathcal{E}$$

is the vibrational "temperature," t_c is the time of vibration-vibration relaxation, N_0 is the initial density of the molecules, $\kappa = I\sigma/\theta$, I is the laser-radiation flux density, σ is the resonant-absorption cross section, \mathcal{E}_* and \mathcal{E}^* are respectively the limit energy above which the molecules are populated only by resonant collisions, and the dissociation energy, $\nu(x)$ is a function equal to zero or unity depending on the sign of its argument.

We seek the function $\Phi(\tau, \epsilon)$ in the form

$$\Phi(\tau, \epsilon) = e^{-k\tau} \phi(\epsilon), \int_0^{\epsilon^*} \phi(\epsilon) d\epsilon = 1, \phi(\epsilon^*) = 0, \quad (2)$$

where $\epsilon^* = \mathcal{E}^*/\theta$ is the reduced dissociation energy of the molecule. From (1), with (2) taken into account, we can obtain two relations for determining the dissociation-rate constant k and the vibrational "temperature" θ :

$$k = J(\epsilon^*), \quad k(\epsilon^* - 1) = \kappa \left[\int_0^{\epsilon^*} \phi(\epsilon) d\epsilon - \epsilon \phi(\epsilon) \right]_{\epsilon = \epsilon^*}. \quad (3)$$

Relations (3) have a simple physical meaning: the dissociation constant is equal to the particle flux at the point $\epsilon = \epsilon^*$, and the energy loss as a result of the dissociation is offset by the energy absorbed from the laser radiation.

From (1) we obtain for the distribution function $\phi(\epsilon)$ the following solution

$$\phi(\epsilon) = \begin{cases} C_0 \exp\left(-\frac{\epsilon}{1+\kappa}\right) {}_1F_1\left(-k, 1, \frac{\epsilon}{1+\kappa}\right), & \epsilon < \epsilon_* \\ C_1 \exp(-\epsilon) {}_1F_1(-k, 1, \epsilon) + C_2 \exp(-\epsilon) \psi(k, \epsilon), & \epsilon > \epsilon_* \end{cases}, \quad (4)$$

where ${}_1F_1(x, y, z)$ is the confluent hypergeometric function,

$$\psi(k, \epsilon) = {}_1F_1(-k, 1, \epsilon) \ln \epsilon + \exp(\epsilon) \sum_{m=1}^{\infty} C_{m+k}^m \frac{(-\epsilon)^m}{m!} \sum_{\nu=0}^{m-1} \left(\frac{1}{1+\nu+k} - \frac{2}{1+\nu} \right),$$

C_{m+k}^m are the binomial coefficients, and C_0 , C_1 , and C_2 are constants determined by relations (2) and by the condition for the continuity of the flux $J(\epsilon)$ at the point ϵ_* .

In the case of greatest practical interest $\epsilon_* = h\nu/\theta$, i.e., there is weak anharmonicity, which does not affect strongly the resonant-exchange processes but which excludes absorption of field quanta between all levels except the ground and first-excited levels. We can then obtain with the aid of (3) and (4) analytic expressions for the vibrational "temperature" θ and for the dissociation constant $\gamma = k/t_c$:

$$\gamma = \frac{1}{2t_c} a \left(\frac{h\nu}{\mathcal{E}^*} \right)^2 \frac{(\mathcal{E}^*/\theta)^2}{1 + a\mathcal{E}^*/\theta}, \quad (5)$$

$$\exp\left(-\frac{\mathcal{E}^*}{\theta}\right) = \frac{a}{2} \left(\frac{h\nu}{\mathcal{E}^*} \right)^2 \frac{\mathcal{E}^*/\theta}{1 + a\mathcal{E}^*/\theta}, \quad a = \frac{I\sigma t_c}{\mathcal{E}^*}. \quad (6)$$

3. It follows from (5) and (6) that in weak fields ($\alpha \rightarrow 0$) the dissociation constant is actually proportional to the radiation flux density. On the other hand, in strong fields ($\alpha \rightarrow \infty$), the vibrational "temperature" and the dissociation constant tend to a finite limit, corresponding to a bleaching of the system, connected with the saturation effect. In conclusion, we present numerical calculations for a case close to the experimental conditions of [6, 7], i.e., $t_c \approx 10^{-6}$ sec, $t_m \approx 10^{-3}$ sec, $hw \approx 0.1$ eV, $\mathcal{E}^* \approx 2$ eV, $\sigma \approx 10^{-17}$ cm² (pressure ~ 100 Torr), $I \approx 10^2$ W/cm². Then $\theta \approx 0.2$ eV and $\gamma \approx 10^3$ sec⁻¹. Thus, $\gamma t_m \sim 1$ under these conditions, and consequently a noticeable fraction of the molecules will be dissociated prior to the onset of the vibrational-translation relaxation.

Obviously, the results can be directly applied to the case of decay, resulting from a chemical reaction, of a vibrationally excited molecule. In formulas (5) and (6) it is then necessary to take \mathcal{E}^* to mean an energy close to the activation energy of the chemical reaction.

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INFLUENCE OF NONEQUILIBRIUM EXCITATIONS ON THE PROPERTIES OF SUPERCONDUCTING FILMS IN A HIGH-FREQUENCY FIELD

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A number of experiments have revealed a certain increase of the critical current of thin films [1, 2], and also of their transition temperature [3] under the influence of a high-frequency field. As already noted [4], effects of this type may be due to a redistribution of the electronic excitations, which inevitably occurs under conditions of stationary irradiation. What remained uncertain, however, was the possible scale of the effect, since the calculations were made only in first order in the field intensity. The results presented below fill this gap.

We used for the calculation a model in which the film was assumed to be so thin that the density of the high-frequency current in the energy gap Δ remained constant over its cross section. At the same time we assumed that the mean free path of the electron is small compared with the film thickness, making it unnecessary to take into account the singularities due to reflection from the walls.

The principal role is played in what follows by the assumption that the lifetime of the excitation τ_0 is large in relation to the energy transfer. In the case of a metal at $T \ll \theta$ (the Debye temperature) this time, as is well