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FEASIBILITY OF AN ADSORPTION-GASDYNAMIC LASER

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The operation of the gasdynamic laser [1 - 4] is based on processes of vibrational relaxation of polyatomic molecules, which result from paired collisions of the molecules in the volume of the gas.

The purpose of the present article is to show that non-equilibrium expansion of a two-phase gas-aerosol system in a supersonic nozzle can be accompanied by inversion of the population of the vibrational levels of polyatomic anisotropic molecules, as a result of vibrational relaxation of the molecules in the adsorbed state on the surface of the aerosol particles. The surface relaxation greatly increases the number of molecular gases in which population inversion can be obtained by the gasdynamic method.

We assume that the adsorption has a dynamic character, i.e., within the time scale characteristic of the problem, the molecule is in the adsorbed state many times, and that physical adsorption on the surface occurs at a temperature higher than the critical two-dimensional-condensation temperature [5].

The dependence of the average lifetime of the molecule in a two-phase gas-aerosol system on the type of vibrational level of the molecule is due to the joint action of three factors: 1) the adsorbed molecule is oriented in a definite manner relative to the surface of the adsorbent; 2) the damping of different vibrational modes of the molecule depends on its orientation in the adsorbed state; 3) the time of stay on the surface also depends on the orientation of the molecule. For clarity, we shall consider the CO_2 molecule, and to simplify the calculations we assume that the rotational motion of the molecule in the adsorbed state is suppressed and that the molecule can be oriented either parallel or perpendicular to the surface.

Let us determine the dependence of the time of stay of the molecule in the adsorbed state on its orientation. The density of the fluctuating electromagnetic fields, which exists near the surface of the adsorbent, decreases like $1/r^3$ with increasing distance r from the surface. The energy of interaction of the molecule with this field gives the dispersion component of the Van-der-Waals forces. If we assume that the main cause of the difference between the absorption energies for two orientations is the change of the distance between the center of the molecule and the surface of the absorbent, then

$$\gamma = E_{\perp}/E_{\parallel} = (b/a)^3,$$

where a and b are the dimensions of the electron cloud of the molecule parallel and perpendicular to the molecule axis. Taking b/a = 0.7 for the CO_2 molecule,

we get $\gamma = 0.34$. The time that the molecule stays in the adsorbed state depends exponentially on the adsorption energy

$$\tau = \tau_0 \exp(E/kT),$$

$$\eta = \frac{\tau_\perp}{\tau_\parallel} = \exp\left(\frac{E_\perp - E_\parallel}{kT}\right) = \exp\left[-\frac{E_\parallel}{kT}(1-\gamma)\right],$$

where $1/\tau_0 = 10^{12} - 10^{15}$ sec^{-1} is the frequency of the oscillations of the molecules relative to the adsorbent surface [5]. In the adsorption of CO_2 on charcoal we have $E = 6.5$ kcal/mole [5], $\eta = 0.1$ at $T = 900°K$, and $\eta = 10^{-3}$ at $T = 300°K$. If the molecule is strongly elongated in one direction, then the difference between the adsorption times is large.

Let us determine the damping of different vibrational modes of the molecule as a function of the molecule orientation on the surface. We assume to this end that the oscillatory motion of the CO_2 molecule can be represented by four linear oscillators, two of which, the symmetrical (m00) and the antisymmetrical (00m) valence oscillations, are directed along the molecule axis, the z axis, and the directions of the two others, comprising a doubly degenerate deformation (0m0) oscillation, coincide with the directions of the x and y axes; we assume that the damping of all four oscillators is the same and equal to δ if the direction of the oscillations is perpendicular to the adsorbing surface, and equal to zero if the direction is parallel to the surface. We assume also that the probability of deactivation of the molecule is small within the time of a single stay in the adsorbed state, $p = \tau\delta \ll 1$. The dependence of the damping of the different vibrational modes on the molecule orientation is shown in the table.

Orientation	(m00)	(0m0)		(00m)	Lifetime in adsorbed state
$\parallel$	0	0	δ	0	$\tau_\parallel$
$\perp$	δ	0	0	δ	$\tau_\perp$

Let us determine the ratio of the lifetime τ_u of the CO_2 molecule in the two-phase system at the upper laser level (00^01) to the average lifetime τ_ℓ at the four lower levels (10^00), (02^00), (02^20), which have equal populations owing to the resonant exchange in the gas phase

$$\frac{\tau_u}{\tau_\ell} = \frac{\tau_\perp \delta + 3\tau_\parallel \delta}{4\tau_\perp \delta} = \frac{3+\eta}{4\eta} \approx \frac{1}{\eta}.$$

The formula shows that the lifetime of the molecule at the lower level is shorter by a factor $1/\eta$ than the lifetime at the upper laser level.

In supersonic wind tunnels and in gasdynamic lasers there is a common reason why flow in the supersonic part becomes two-phase. The reason is the volume condensation of the vapors of those substances which are contained in the form of slight admixtures in the gas and have a noticeable vapor tension compared with the total gas pressure.

The presence of aerosol particles in the gas stream causes attenuation of the transmitted infrared radiation, owing to the absorption and scattering by the minute particles. Let us estimate the total attenuation of a plane

wave for two cases: water droplets, $N \sim 10^{13}$ cm^{-3}, D = 30 Å, using the value of the complex dielectric constant for a radiation frequency ν = 1000 cm^{-1}, and also metallic spheres of copper having the same dimension and the same density, at the same frequency. The photon free path in a two-phase medium is $\ell \sim 1/N\sigma$, and in the former case $\ell \sim 0.1$ km while in the latter it is 30 times larger (σ is the total-attenuation cross section).

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ELECTROSTATIC ENERGY OF A DISORDERED ALLOY

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The electronic theory of alloys, based on the method of pseudopotentials, is presently being successfully developed (cf., e.g., [1, 2]). As follows from the theory, an important contribution to the total energy of the alloy is made by the electrostatic energy of the ion lattice in the field of the uniformly-distributed negative charge. The expression for the electrostatic energy E_{es} of a completely disordered binary alloy was obtained in [1] by generalizing the corresponding expression for the E_{bs} of the band structure of the alloy. It turned out that both E_{es} and E_{bs} consist of two parts each, the energy of the "average" ion lattice (which contains the sum over the reciprocal-lattice points) and the so-called difference energy, which is proportional to $(Z_A - Z_B)^2$ (Z_A and Z_B are the charges of the ions of type A and B), and expressed in the form of a sum over all the reciprocal-space points allowed by the cyclic boundary condition, with the exception of the reciprocal-lattice vectors.

As is well known, the electrostatic energy of a system of charges in a homogeneous compensating field is conveniently determined with the aid of the Ewald transformation. Ewald's method employs a parameter η, which characterizes the dimensions of the Gaussian charge-distribution "caps" over the lattice points. The value of η is chosen such as to ensure optimally rapid convergence of the series. The final result should in this case be independent of η. However, the expression given in [1] for E_{es} does not satisfy this requirement, since the first term (the electrostatic energy of the "average" crystal) does not depend on η, whereas the second is a function of η.

We shall show in this paper how to effect the Ewald transformation for an alloy with an arbitrary ion placement. It turns out here that in a disordered alloy, in the absence of short-range order (the case considered in [1]), the expression for E_{es} has no difference term at all. If, on the other hand, there is a correlation in the placement of the ions (short-range order), then the