POPULATION INVERSION IN NITROUS OXIDE AT HIGH PRESSURES AND TEMPERATURES

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Submitted 12 July 1973
ZhETF Pis. Red. 18, No. 4, 249 - 253 (20 August 1973)

Absolute population inversion is obtained for a number of lines of the 00^01 - 10^00 transition of the N_2O molecule through resonant energy transfer from the vibrationally-excited nitrogen produced when some of the N_2O molecules are decomposed. The inversion reached 10^{17} cm^{-3} at a total pressure 28 atm and a gas temperature 2000°K.

The prospects for using exothermal decomposition of polyatomic molecules to produce population inversion were indicated in [1, 2]. It is proposed there to produce inversion between rotational-vibrational level of the initial decomposing molecule as a result of the fact that the vibrationally-excited product of its decay has an oscillation frequency close to one of the fundamental frequency of the dissociating molecule, and this can lead to selective pumping to vibrational levels of the corresponding mode.

Such a situation is realized in the dissociation of the N_2O molecule if it is rapidly heated to temperatures above 1600°K. In the exothermal dissociation of N_2O, an appreciable fraction of the energy can be released in the form of vibrational energy of the nitrogen molecules, the fundamental frequency of which is $\nu = 2359$ cm^{-1} and is close to the frequency $\nu_3 = 2224$ cm^{-1} of the antisymmetrical vibration of N_2O (the level scheme of N_2O is similar to the widely known level scheme of CO_2). When the 00^01 level of N_2O is populated at a sufficiently high rate by collisions with the excited nitrogen molecules, inversion can occur between this level and the group of levels 10^00 and 02^00.

For inversion to set in, it is necessary that the rate of energy release, which is determined by the rate v_{chem} of the chemical reaction, be larger than or equal to the rate of the vibrational-vibrational exchange between the different oscillation modes v_{VV}, and greatly exceed the rate v_{VT} of the vibrational-translational relaxation of the 00^01 level of the N O molecule.

We have established experimentally the existence of inversion between the vibrational-rotational levels of N_2O molecules; this inversion is due to the chemical energy released when a fraction of these molecules is dissociated.

The experiments were performed with a shock tube. We used the procedure of amplifying the beam of a low-power electric-discharge N_2O laser. The setup is illustrated in Fig. 1. We used in the experiments mixtures of N_2O with argon, helium, and nitrogen. The translational temperature T in the inverted medium was 1600 - 2000°K, and the pressure P was 9 - 28 atm. A typical oscillogram obtained for an $N_2O:N_2$ (1:4) mixture is shown in Fig. 2. The signals of the laser and of the pressure pickup are constant between the start of the oscilloscope and the arrival of the incident wave (point A). The arrival of the incident wave is characterized by a jumplike increase in pressure, and also by the start of a noticeable absorption of the laser beam (point A). The arrival of the reflected wave (point B) also corresponds to a rapid change in the pressure-pickup signal and a sharp minimum of the laser signal, corresponding to deflection of the beam as a result of the density gradient in the front of the reflected wave. This minimum is followed by the start of the amplification peak. The duration of the amplification pulse (C) was usually 20 - 30 μsec. In the succeeding instants of time, a monotonic attenuation of the sounding-laser beam intensity was observed.

The main results of the experiments are listed in the table. In our first experiments we tended to duplicate the operating conditions of [1] with an N_2O:Ar mixture. Under the conditions in which vibrational unbalance was observed in [1] (line 1 of the table), we recorded amplification. The maximum gain, however, 9.2×10^{-3} cm^{-1}, we obtained with the $N_2O:N_2$ (1:4) mixture, line 6 of the table. A gain α of this size is sufficient to produce lasing in this mixture, but we were unable to observe it, probably because of technical difficulties.

The dependence of α on the temperature, for the same optimal mixture, is shown in Fig. 3. The extremal character of this dependence is evident. This means that the condition $v_{chem} > v_{VT}$ is satisfied only in a limited range of temperatures. It is seen from a comparison of the data in lines 5, 6, and 7 of the table that the maximum gain increases with increasing pressure.

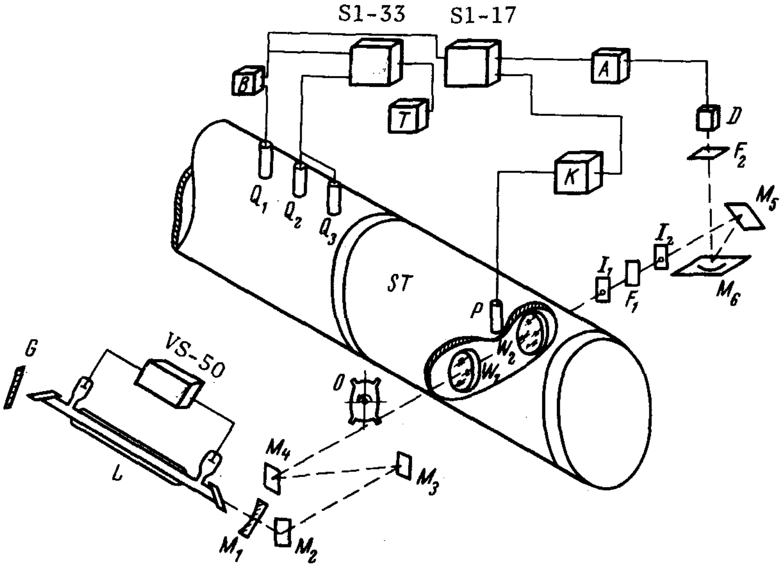

Fig. 1

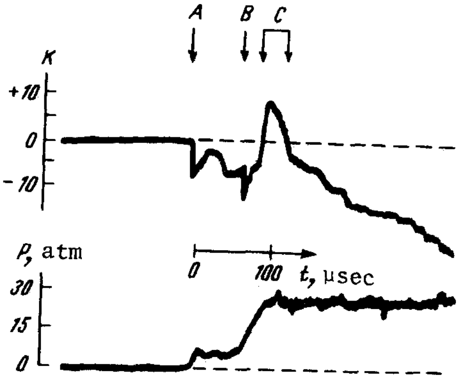

Fig. 2

Fig. 1. Experimental setup: ST - shock tube, L - sounding laser with type VS-50 supply, G - diffraction grating (100 lines/mm), M_1 - laser mirror, M_2 - M_6 - rotary mirrors, O - shutter, W_1 and W_2 - germanium observation windows, I_1 and I_2 - diaphragms of 5 mm diam, F_1 and F_2 - filters of teflon and InSb, respectively, D - Ge-Au photoresistor, A - amplifier, P - type LKh-601 pressure pickup, K - cathode follower, Q_1 - contact pickup triggering oscilloscopes S1-33 and S1-17 through a delay block B, Q_2 and Q_3 - contact pickups to measure the shock-wave velocity, T - quartz oscillator.

Fig. 2. Typical oscillogram for N_2O:N_2 mixture (1:4): T = 1950°K, p = 28 atm, I - pressure-pickup signal, II - sounding-laser signal, A - arrival of incident shock wave, B - arrival of reflected shock wave, C - zone where inversion exists.

Experimental conditions and results

Run number	Mixture comp., rel.un.				$\alpha_{max}\cdot 10^2$ cm^{-1}	T_{max} °K	P_{max} atm
	N_2O	N_2	Ar	He			
1	1	–	4	–	0.49	1710	8
2	1	–	9	–	0.28	1680	6
3	1	–	–	4	0.05	2100	11
4	1	4	–	4	0.23	2080	13
5	1	4	–	–	0.33	2300	12
6	1	4	–	–	0.77	2050	19
7	1	4	–	–	0.92	1950	28

α_{max} - maximum gain for given mixture, T_{max}, P_{max} - temperature and pressure corresonding to α_{max}.

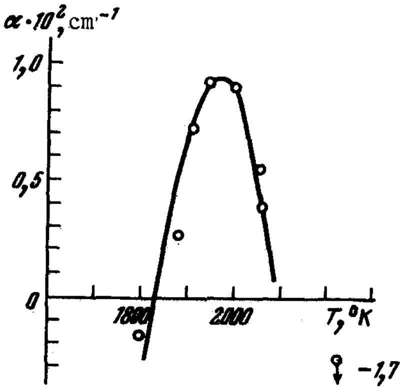

Fig. 3. Temperature dependence of gain α for N_2O:N_2 (1:4) mixture at approximate pressure 28 atm.

We have also investigated the dependence of the gain on the spectral composition of the sounding radiation. Gain was obtained for both the P and the R branch of the $00^01 - 10^00$ transition. We can therefore conclude that the inversion in our case was complete and not partial.

According to preliminary estimates, for conditions not optimized with respect to the most important parameters, the inversion under the conditions of the described experiments can reach 10^{17} cm^{-3} and the per-unit lasing energy ∿5 J/l.

The authors thank A. M. Prokhorov, I. S. Zaslonko, and B. F. Gordiets for useful discussions and advice.

[1] I. S. Zaslonko, S. M. Kogarko, E. V. Mozzhukhin, and A. I. Demin, Dokl. Akad. Nauk SSSR 202, 1121 (1972).
[2] I. S. Zaslonko, S. M. Kogarko, E. V. Mozzhukhin, and A. I. Demin, Gorenie i vzryv (Combustion and Explosion), Proc. of 3rd All-union Symposium on Combustion and Explosion, 5 - 10 July 1971, Leningrad, Nauka, 1972, p. 685.

SPONTANEOUS SINGLE-FREQUENCY GENERATION OF A SOLID-STATE RING LASER

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Submitted 13 July 1973
ZhETF Pis. Red. 18, No. 4, 253 - 255 (20 August 1973)

1. The known methods of obtaining single-frequency1) emission from solid-state lasers consist either of Q-discrimination of the axial modes, or of elimination of the spatial inhomogeneity of the field in the active medium, and special optical elements or mechanical motions are required in all cases. We consider here the possibility of obtaining single-frequency emission from a solid-state laser with homogeneously-broadened luminescence line. This possibility is based on the instability of the standing-wave regime in a ring resonator. We show that under certain conditions single-frequency lasing occurs in such a resonator spontaneously, i.e., without the use of the aforementioned special means.

2. At a sufficiently low scatter-induced coupling between the opposing waves, the generation of one axial mode is unstable, and a nonstationary self-oscillatory regime can set in [1 - 3], wherein the radiation becomes periodically unidirectional, and the phase difference between the opposing waves oscillates; this can be treated as the motion of the field maxima and minima along the resonator axis. It is natural to expect these singularities in the field pattern, which contribute to smoothing of the spatial inhomogeneity, to lead to single-frequency generation. On the other hand, stable stationary generation of standing waves of several axial modes remains feasible [1]. Which of these possibilities is realized depends on the relations between a number of factors, the character of the competition between them can be explained by means of the following qualitative considerations: Assume that at the initial instant there exists in the resonator a standing wave of one axial index at the frequency of the luminescence-line center, spatially modulating the inverted population. In accordance with the first possibility, a traveling wave of the same frequency and with a growth increment F_0 should appear in the resonator, and in accordance with the second possibility there should appear one more standing wave with an axial index that differs by ξ, shifted in space relative to the first at the center of the active medium by $\lambda/4$, with a growth increment F_ξ. One can expect the realized regime to be the one in which the growth increment of the corresponding wave is larger, this being more favored energywise. By comparing the increments, we obtain for their difference, after integrating over the length of the active medium:

$$F_\xi - F_0 \sim a(\sin 2\pi\xi\zeta / 2\pi\xi\zeta) - [b(1/g_\xi - 1) + (\gamma_\xi - \gamma_0)/g_\xi]. \qquad (1)$$

Here a and b are positive pump functions, g_ξ is the form factor of the luminescence line of the active medium, ζ is the coefficient of the filling of the resonator with the active medium along the optical length, and γ_0 and γ_ξ are the losses in the resonators at the corresponding frequencies. On the basis of [1] we can conclude that when the filling coefficient increases upward from zero, the single-frequency automodulation regime becomes more favored, and if $\zeta = 1/2$ and 1, this regime becomes preferred at all values of the pump regardless of the value of ξ. The single-frequency regime becomes more favored energywise also when selective losses $\gamma_\xi - \gamma_0 > 0$ are introduced into the resonator.

3. The experiments were performed with a three-mirror resonator having an optical length 50 cm, with generation of a fundamental transverse mode, using YAG:Nd^{3+} crystals at 1.06 μ