

# Conditions for efficient operation of metal-vapor lasers with indirect excitation of the upper lasing levels

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We analyze the mechanism of creating population inversion in gas-discharge lasers on the transitions with the upper lasing level populated due to collisional excitation. The conditions for efficient operation of rare-earth vapor lasers are described, and their power capabilities are evaluated using thulium as an example.

## Introduction

One of the ways to create the population inversion in metal-vapor lasers is collisional transfer of the excitation energy.

In this case the part of a donor is played by the resonance level, which is well excited by the electron impact from the ground atomic state in discharge, while the nearby nonresonance upper laser level plays the part of an acceptor. Atoms in the ground state as well as molecules and atoms of rare gases can be used as collisional partners. Under the gas discharge conditions, the transfer of the excitation energy by collisions among identical atoms is the most efficient way.<sup>1</sup> The choice of rare-earth elements as object for the study was caused by the complexity of their atomic spectra and, consequently, by a great number of indirectly excited levels nearby the resonance ones.

In this paper we evaluate the conditions for efficient operation of rare-earth gas-discharge lasers at the transitions with the population inversion created by the above-described method.

## Scheme of creating the population inversion

In the diagram shown in Fig. 1 the following designations are used:  $A_{ij}$  is the probability of a spontaneous transition;  $S_{ij}$  and  $S_{ji}$  are the rate constants of de-excitation and excitation at atom-atom collisions;  $X_{ji}$  is the rate constant of excitation by the electron impact;  $Y_{ij}$  is the rate constant of de-excitation by the electron impact;  $P$  is the lasing transition. Since the level 1 is metastable, the probability of the spontaneous transition  $A_{10}$  is negligibly low; the processes of excitation at atom-atom collisions leading to the transitions  $0 \rightarrow 1$ ,  $0 \rightarrow 2$ ,  $0 \rightarrow 3$ ,  $1 \rightarrow 2$ , and  $1 \rightarrow 3$  are ignored because of a large energy separation between these levels.

The equations for the population of levels have the following form:

$$dN_3/dt = -N_3(Y_{32} + Y_{31} + Y_{30} + A_{31} + A_{30} + S_{32}) + N_2(S_{23} + X_{23}) + N_1X_{13} + N_0X_{03}, \quad (1)$$

$$dN_2/dt = N_3(Y_{32} + S_{32} + A_{32}) - N_2(Y_{21} + Y_{20} + S_{21} + S_{20} + S_{23} + A_{21} + A_{20} - X_{23}) + N_1X_{12} + N_0X_{02} - P, \quad (2)$$

$$dN_1/dt = N_3(Y_{31} + A_{31} + S_{31}) + N_2(Y_{21} + A_{21} + S_{21}) - N_1(X_{12} + X_{13} + S_{10} + A_{10} + Y_{10}) + N_0X_{01} + P, \quad (3)$$

$$dN_0/dt = N_3(A_{30} + S_{30} + Y_{30}) + N_2(A_{20} + S_{20} + Y_{20}) + N_1(Y_{10} + S_{10}) - N_1(X_{01} + X_{02} + X_{03}). \quad (4)$$

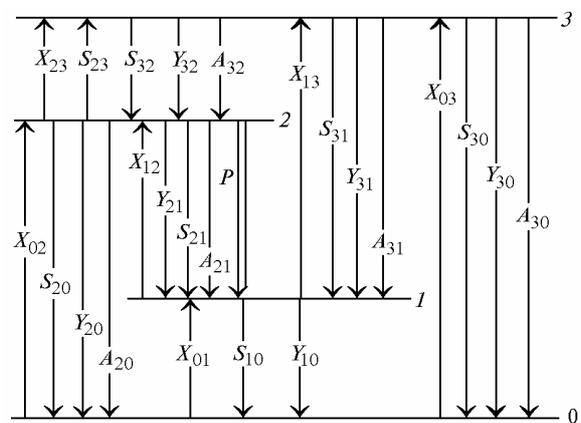
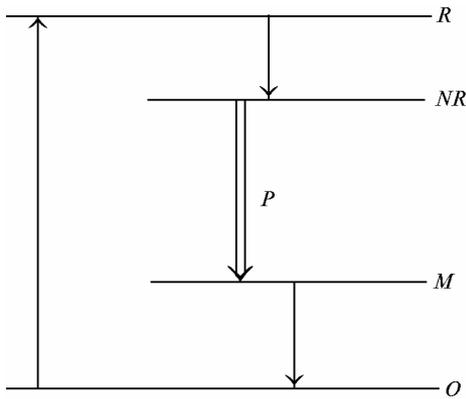


Fig. 1. Creation of population inversion in lasers at transitions with indirect excitation of upper laser levels.

Taking into account nonequivalence of the direct and inverse processes and radiation trapping by the resonance transitions, we can consider a simplified scheme of creating the population inversion (Fig. 2).

In this scheme the main factors determining its efficiency are radiation trapping by the resonance transition, which transforms the resonance level into the effective donor, and energy defect between the resonance and upper lasing levels.



**Fig. 2.** Generalized scheme of the lasing levels of self-limited transitions with indirect excitation of the upper lasing levels: resonance level ( $R$ ); nonresonance (upper lasing) level ( $NR$ ); metastable (lower lasing) level ( $M$ ); ground state ( $O$ ); the lasing transition ( $P$ ).

### Calculation of radiation trapping factor

Radiation trapping can be characterized by the factor  $F$ , which decreases the probability of the spontaneous transition from the resonance level and, consequently, increases its lifetime:

$$A_{ij}(\text{with trapping}) = FA_{ij}(\text{without trapping}), \quad (5)$$

where  $A_{ij}$  is the probability of the spontaneous transition, and the factor  $F$  for a cylinder of radius  $R$  can be found from the equation

$$F = 1.6 / \{K_{ij} R [\pi \ln(K_{ij} R)]^{1/2}\}, \quad (6)$$

where  $K_{ij}$  is the absorption coefficient for laser radiation, and

$$K_{ij} = (1/8\pi)(\ln 2/\pi)^{1/2}(1/\Delta v_D)\lambda_{ij}^2 A_{ij} N_j g_i/g_j, \quad (7)$$

where  $\lambda_{ij}$  is the wavelength;  $\Delta v_D$  is the Doppler line width;  $N_j$  is the concentration of atoms in the ground state;  $g_i$  and  $g_j$  are the statistical weights of the levels. The probability of spontaneous transition  $A_{ij}$  and the Doppler line width can be found as:

$$A_{ij} = 0.67 g_j/g_i (1/\lambda_{ij}^2) f_{ij}, \quad (8)$$

$$\Delta v_D = 2v_0/c (2\ln 2 RT/\mu), \quad (9)$$

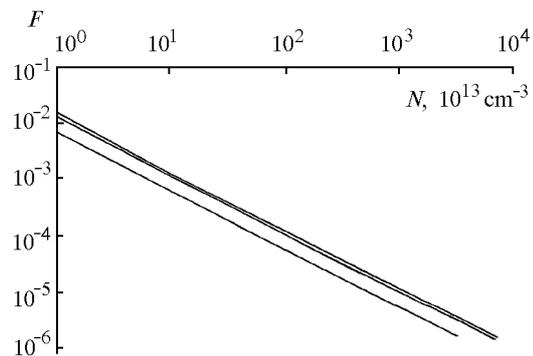
where  $f_{ij}$  is the oscillator strength;  $c$  is the speed of light;  $\mu$  is the molar weight of the atom;  $v_0$  is the frequency of the transition;  $R$  is the universal gas constant;  $T$  is the temperature. Upon substitution of the known parameters, we have

$$\Delta v_D = 7.162 \cdot 10^{-7} v_0(T/\mu). \quad (10)$$

By this method we have calculated the factors  $F$  for ten lanthanides: thulium, samarium, dysprosium, holmium, erbium, ytterbium, gadolinium, europium, neodymium, and praseodymium. The calculations were made for five strongest resonance lines of each element. The values of the oscillator strength were taken from Ref. 2.

Taking into account the capabilities of modern high-temperature equipment used in metal-vapor lasers and operating at  $T$  no higher than 2000 K as well as high reactivity of rare-earth elements in reactions of oxide substitution at high temperatures, we have selected the elements, in which radiation trapping occurs at  $T$  below 1800 K ( $F < 0.01$ ).

The following elements turned out to be suitable for use in the experiments: ytterbium, europium, samarium, thulium, dysprosium, erbium, holmium, and neodymium. The lasing in the first five elements have been obtained earlier.<sup>3,4</sup> Figure 3 shows the calculated dependence of the factor  $F$  on the concentration of thulium atoms for three transitions.



**Fig. 3.** Radiation trapping factor vs. concentration of thulium atoms for transitions with the wavelengths of 388.313, 374.407, and 371.792 nm.

### Collisional transfer of the excitation energy

Collisional transfer of the excitation energy was extensively studied in vapors of alkali and alkali-earth elements. The excitation energy transfer due to collisions of metal atoms with the excited atoms and molecules of rare gases was studied. Other elements were also studied, for example, zinc, mercury, cadmium, as well as excitation transfer between atoms of different elements: between magnesium and calcium, between magnesium and samarium, etc. The values of cross sections  $\sigma$  of such processes depend on the type of collisional partners and energy defect  $\Delta E$ ; these can achieve  $10^{-12} - 10^{-13} \text{ cm}^2$ . Thus, for example, at collisions of unexcited atoms with cesium atoms in an excited state the value of  $\sigma$  for excitation transfer is  $2.1 \cdot 10^{-14} \text{ cm}^2$  at  $\Delta E = 42.94 \text{ cm}^{-1}$ ,  $2.7 \cdot 10^{-14}$  and  $12.6 \cdot 10^{-14} \text{ cm}^2$  at, respectively,  $\Delta E = 20.97$  and  $7.16 \text{ cm}^{-1}$  (Ref. 1). In the same reaction for Ca and He at  $\Delta E = 105.9 \text{ cm}^{-1}$  it is  $3.19 \cdot 10^{-15} \text{ cm}^2$ , and for lithium at  $\Delta E = 357.7 \text{ cm}^{-1}$  it is  $4.3 \cdot 10^{-15} \text{ cm}^2$  (Ref. 1). From the above data it is seen that the cross section increases with decreasing energy defect. Thus, elements with very rich energy level structure and, consequently, small energy defects should be chosen as an object for the study. Rare-earth elements meet these requirements very well.

## Estimation of power characteristics of lasers with indirect excitation of the upper lasing levels

Power characteristics of these lasers were estimated for the laser transition with  $\lambda = 1338$  nm in thulium atoms. The upper level,  $E = 22742$  cm<sup>-1</sup>, of this transition is not a resonance one and it is populated through collisional transfer of excitation energy from two nearby resonance levels:  $E = 22929$  ( $\lambda = 435.993$  nm) and  $22791$  cm<sup>-1</sup> ( $\lambda = 438.643$  nm). The energy defects in these cases are 187 and 49 cm<sup>-1</sup>, respectively. The laser output power was estimated by the equation

$$P = V f_{\text{rep}} h N \nu, \quad (11)$$

where  $V$  is the laser tube volume;  $f_{\text{rep}}$  is the pulse repetition rate;  $h$  is the Planck's constant;  $N$  is the number of atoms involved in the laser transition;  $\nu$  is the laser transition frequency. The number of atoms contributing to laser emission is:

$$N = (N_1 + N_2)(g_{NR}/g_{NR} + g_M), \quad (12)$$

where  $N_1$  and  $N_2$  are the numbers of atoms participating in transitions from the above-listed resonance levels to the upper lasing level due to collisional transfer of the excitation energy;  $g_{NR}$  and  $g_M$  are the statistical weights of the nonresonance upper lasing and metastable levels (it was assumed that at the start of lasing the population of the metastable level is much less than that of the upper lasing level). The numbers of excitation atoms during the laser pulse  $N_1$  and  $N_2$  can be determined from the following equations:

$$N_1 = N_O V^{Tm} N_{R1} \sigma_{R1 \rightarrow NR}^{Tm, Tm} \tau g_{R1} / g_{NR}, \quad (13)$$

$$N_2 = N_O V^{Tm} N_{R2} \sigma_{R2 \rightarrow NR}^{Tm, Tm} \tau g_{R2} / g_{NR}, \quad (14)$$

where  $N_O$  is the concentration of atoms in the ground state;  $V^{Tm}$  is the mean velocity of atoms;  $N_{R1}$  and  $N_{R2}$  are the concentrations of atoms at the resonance levels;  $\sigma_{R1 \rightarrow NR}^{Tm, Tm}$  and  $\sigma_{R2 \rightarrow NR}^{Tm, Tm}$  are the cross sections of collisional excitation transfer from the resonance levels to the upper lasing level;  $\tau$  is the time of lasing;  $g_{R1}$ ,  $g_{R2}$ , and  $g_{NR}$  are the statistical weights of the resonance levels and the nonresonance (the upper lasing) one.

In their turn,

$$N_{R1} = N_O V^e n_e \sigma_{O \rightarrow R1}^{Tm, e} \tau, \quad (15)$$

$$N_{R2} = N_O V^e n_e \sigma_{O \rightarrow R2}^{Tm, e} \tau, \quad (16)$$

where (in our case  $\tau = 100$  ns)  $V^e$  is the mean velocity of electrons determined from the mean electron energy assuming that the electron temperature is 3.5 eV;  $\sigma_{O \rightarrow R1}^{Tm, e}$  and  $\sigma_{O \rightarrow R2}^{Tm, e}$  are the cross sections of the excitation energy transfer from the ground state to the resonance level at the electron impact;  $n_e$  is the concentration of electrons. The cross sections of excitation transfer from the ground state to the resonance level at the electron

impact were, respectively,  $\sigma_{O \rightarrow R1}^{Tm, e} = 1.64 \cdot 10^{-17}$  cm<sup>2</sup> and  $\sigma_{O \rightarrow R2}^{Tm, e} = 3.63 \cdot 10^{-17}$  cm<sup>2</sup> (Ref. 5).

Since there are no data on the cross sections of excitation transfer at atom-atom collisions of thulium available, their values were taken equal to those for other atoms but with the energy defects close to the above-said:  $\sigma_{R1 \rightarrow NR}^{Tm, Tm} = 2.1 \cdot 10^{-14}$  cm<sup>2</sup> (Ref. 6),  $\sigma_{R2 \rightarrow NR}^{Tm, Tm} = 5 \cdot 10^{-15}$  cm<sup>2</sup> (Ref. 7). The laser tube radius was 1 cm, the tube length was 50 cm, pulses were emitted at a repetition frequency of 10 kHz, and the pulse duration was 100 ns. In calculating we assumed that ionization of thulium atoms was 5%. The dependence of the calculated output power on the concentration of atoms, assuming constant electron temperature is shown in Fig. 4. This dependence was compared with the experimental data obtained with a similar laser tube. The needed wavelength was separated out in a dispersion cavity with a diffraction grating, and the power was measured with a calorimeter using reflection from a glass plate set in the cavity at some angle. At the temperature corresponding to the concentration of thulium atoms of  $10^{15}$  cm<sup>-3</sup>, the mean output power was 220 mW, what closely correlates with the calculated dependence.

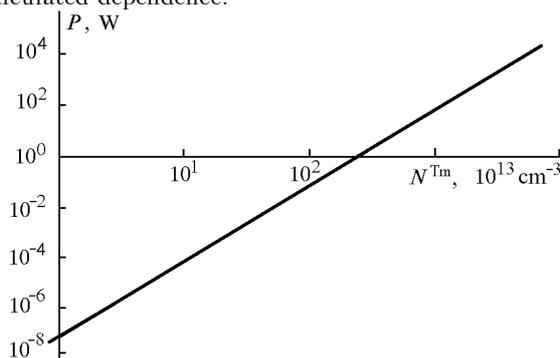


Fig. 4. Output power vs. concentration of thulium atoms.

## Conclusion

Thus, if the conditions of radiation trapping at resonance transitions are fulfilled and the energy defect between the resonance level and the upper lasing level does not exceed 400 cm<sup>-1</sup>, gas-discharge lasers with indirect excitation of the upper lasing level are promising sources of laser radiation.

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