

ATMOSPHERIC ABSORPTION OF RADIATION FROM VARIOUS TYPES OF IODINE PHOTODISSOCIATION LASERS

A.A. Artemov, N.F. Borisov, and V.M. Osipov

S. I. Vavilov State Optical Institute, Leningrad

Received August 15, 1989

The spectral composition of radiation for various types of iodine photodissociation lasers is observed. The spectral behavior of the absorption coefficient is determined on the basis of experimental data on the atmospheric absorption lines. It is shown that the lowest atmospheric absorption exists for the laser pumped by the emission of a high-current discharge plasma.

In the construction of lasers suitable for operation on extended atmospheric paths it is necessary to use laser emitters whose frequencies lie within the atmospheric transparency windows.

These include the iodine photodissociation lasers (PL) in which population inversion occurs on the levels of the hyperfine structure (HFS) of the Iodine $^2P_{1/2}-^2P_{3/2}$ magnetic dipole optical transition. The spectral composition of the photodissociation laser radiation depends to a strong degree on the magnetic field magnitude. The degree of magnetic field localization in the PL active zone is determined in turn by the design features and means of pumping of the laser.

The features peculiar to the PL make it possible to tune the laser spectral composition in the range of $\Delta\nu \sim 0.5 \text{ cm}^{-1}$ by relatively simple and accessible means. Consequently, the possibility exists of controlling the losses attendant to the transfer of PL radiation through the atmosphere since the atmospheric molecular absorption coefficient has a strongly pronounced selective nature² and is therefore

quite sensitive to the laser spectral composition.

In this paper we estimate the absorption of laser radiation for various atmospheric paths on the basis of the experimental data on the spectral composition of most typical photodissociation lasers and the atmospheric molecular absorption coefficient.

1. SPECTRAL CHARACTERISTICS OF THE MAIN TYPES OF PHOTODISSOCIATION LASERS

a. In the absence of external magnetic fields the structure of the $^2P_{1/2}-^2P_{3/2}$ working transition on the only stable isotope I^{127} of the iodine atom, which possesses nuclear spin $I = 5/2$, is determined by the hyperfine splitting of the $^2P_{1/2}$ and $^2P_{3/2}$ terms on $(2I + 1) = 6$ sublevels (see Fig. 1a). In accordance with the $\Delta F = 0, \pm 1$ selection rules, where F is the total atomic angular momentum, the luminescence spectrum has 6 lines³ and the most intense of these corresponds to the $F' = 3 F = 4$ transition.

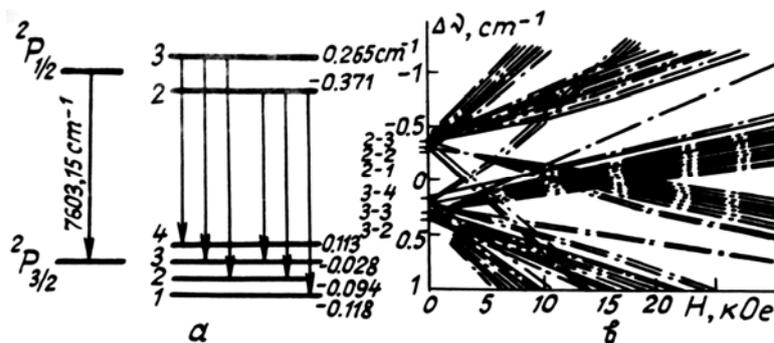


FIG. 1. Hyperfine structure of iodine photodissociation laser transitions (a — in the absence of a magnetic field; b — Zeeman splitting in a constant magnetic field up to 30 kOe)

At the same time the induced emission spectrum of an "unmagnetic" pumped PL (which includes also iodine lasers with solar⁴ and chemical⁵ pumping) is formed by competition of transitions, which is

caused by different rates of relaxation between the sublevels of the ground state and the excited state. Therefore under real conditions of laser operation when the rate of sublevel mixing for the lowest

working state ${}^2P_{3/2}$ significantly exceeds the rate of sublevel relaxation for the ${}^2P_{1/2}$ state,⁶ the spectrum consists of mainly one component corresponding to the strongest transition $F' = 3 \rightarrow F = 4$. The photodissociation laser line shape (see curve "a" in Fig. 2) is caused by collisional broadening and has a Lorentz shape with half-width $j = 0.02\text{--}0.03 \text{ cm}^{-1}$ at half-maximum. The experimental position of the laser line shape center differs slightly from the calculated one and is equal to $\nu_0^a = 7603.314 \text{ cm}^{-1}$.

b. Another group of atomic-iodine lasers contains sources of optical pumping which produce considerable magnetic fields in the PL active medium. The structure of the working transition in a magnetic field is characterized by radical changes caused by Zeeman splitting, and shift and overlap of the HFS components for the ${}^2P_{1/2}\text{--}{}^2P_{3/2}$ transition. Figure 1b shows the frequency spectrum of the ${}^2P_{1/2}\text{--}{}^2P_{3/2}$ transition in a constant magnetic field with magnitude up to 30 kOe, which was calculated in Ref. 7 for the σ - and π -transitions in accordance with the selection rules for weak, intermediate, and strong magnetic fields. Under real conditions of laser operation when optical pumping sources produce magnetic fields that are transient in time and nonuniform across the section of the active medium, the induced radiation spectrum for the ${}^2P_{1/2}\text{--}{}^2P_{3/2}$ transition has in general a multicomponent and transient nature.

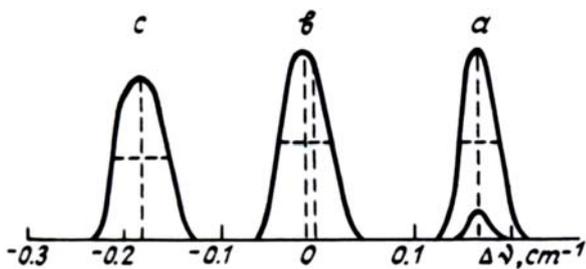


FIG. 2. Line shapes for various types of PL's (the shift $\Delta\nu$ is given with respect to the center of the ${}^2P_{1/2}\text{--}{}^2P_{3/2}$ multiplet $\nu = 7603.13 \text{ cm}^{-1}$).

A photodissociation laser pumped by plasma emission from a high-current discharge, developing directly in the active medium,⁸ is most typical of "magnetic" pumped iodine lasers. In this case the lasing region is an annular zone bordering on the plasma volume which moves in the active medium as the plasma channel expands. Using powerful high-current discharges in these lasers it is possible to realize conditions under which the magnitude of the magnetic field produced in the active region by the plasma current is localized in the range $10 \text{ kOe} < H < 20 \text{ kOe}$. In this case, due to competition between the transitions, the experimentally observed spectrum of PL radiation consists mainly of one magnetically broadened quasi-monochromatic laser line which corresponds to the region of overlap of the $F' = 2 \rightarrow F = 2$ and $F' = 3 \rightarrow F = 3$ transitions. The shape of this line with frequency

$\nu = 7603.15 \text{ cm}^{-1}$ which corresponds to the multiplet center, and with half-width $j = 0.05\text{--}0.06 \text{ cm}^{-1}$, is well approximated by the Lorentz dependence (see curve b in Fig. 2).

c. Iodine lasers pumped by low-pressure z -pinch xenon lamps are another typical example of PL's which are of interest due to their high efficiency (maximum for PL's) and the possibility of a pulsed periodic regime of laser operation. The xenon plasma is formed in the lamp by a high-current oscillatory discharge which produces a transient magnetic field with intensity up to $H \leq 10 \text{ kOe}$ in the laser active region.

Measurements of the kinetics of the spectrum of this type of PL shows that during the first half-period of the discharge current (when about 90% of the laser output is emitted) the laser operates on components shifted from the multiplet center to lower wave numbers ($\Delta\nu > 0.18 \text{ cm}^{-1}$). These components correspond to the overlap region of the $F' = 2 \rightarrow F = 2$ and $F' = 3 \rightarrow F = 4$ Zeeman sublevels when the magnetic field is localized in the lasing region within the range $5 \text{ kOe} < H < 10 \text{ kOe}$. A short-duration spike of radiation with frequency corresponding to the frequency of the $F' = 3 \rightarrow F = 4$ transition unperturbed by a magnetic field is observed when the discharge current passes through zero. In this case the energy of the laser monopulse is about 10–15% of the total laser output. As follows from the above, it may be possible to approximate the spectrum of a powerful-lamp-pumped PL by two Lorentz lines, one of which ($\nu_1^a = 7602.97 \text{ cm}^{-1}$) is shifted by the amount $\Delta\nu = -0.18 \text{ cm}^{-1}$ from the multiplet center and contains 85–90% of the output and the second ($\nu_2^b = 7603.314 \text{ cm}^{-1}$) coincides with the frequency of the $F' = 3 \rightarrow F = 4$ transition unperturbed by a magnetic field (see curve c in Fig. 2).

2. ABSORPTION OF PL RADIATION ON ATMOSPHERIC PATHS

At present there are quite a few papers devoted to the determination of the atmospheric gas absorption coefficients on the iodine laser frequency $\nu_0 = 7603.314 \text{ cm}^{-1}$ (see, e.g., Ref. 10). To estimate the absorption of quasi-monochromatic PL radiation with various spectral composition, it is necessary to know the spectral behavior of the absorption coefficients (i.e., the shape of the function K_ν) in the appropriate spectral range and also its variation with altitude. Such information may be obtained from data on the spectral lines of the various atmospheric gases. Now almost all such tasks have been solved using the AFGL compilation¹¹ of spectral line parameters of atmospheric gases. The parameters of the relatively weak spectral lines often have considerable errors in spite of unceasing work on correction of the existing data and addition of new data.¹¹ Therefore in the creation of the initial databank of spectral line

parameters¹¹ we used the experimental results of Refs. 2 and 10 to correct the spectral parameters of water vapor. In this spectral region the absorption by CO₂, CH₄ and H₃ lines is considerably weaker. Therefore, possible errors in the specification of their spectral parameters have hardly any influence on the accuracy of calculation of the total atmosphere absorption.

The absolute Intensities and half-widths of the seven strongest water vapor lines in the region 7600–7608 cm⁻¹ are given in Ref. 2. The relative contributions of the weaker H₂O lines (including the lines of the H₂O isotope) to the absorption at the frequency 7603.14 cm⁻¹ are given in Ref. 10. Many of these lines are absent in Ref. 11. Two of them lie immediately in the region of PL operation and, consequently, determine the absorption in this region.

The data given in Ref. 10 allow one to calculate the relative intensities of these lines. For their normalization we used the value of the line intensity at the frequency $\nu = 7602.352 \text{ cm}^{-1}$ given in Ref. 2, which is equal to $S = 1.15 \cdot 10^{-23} \text{ cm}^{-1}/\text{mole} \cdot \text{cm}^{-2}$. Table I presents the parameters of the H₂O spectral lines calculated in this way (S_{calc}) and used in Ref. 11 to correct the data of the compilation.

TABLE I.

Parameters of the H₂O Spectral Lines In the Region of PL Operation.

frequency, cm ⁻¹	S_{calc}	S_{comp}	S	air
7602.352	$1.15 \cdot 10^{-23}$	$1.52 \cdot 10^{-23}$	$1.15 \cdot 10^{-23}$	0.08
7602.82	$5.41 \cdot 10^{-25}$	-	-	0.08
7603.32	$1.32 \cdot 10^{-25}$	-	-	0.08
7603.58	$0.17 \cdot 10^{-25}$	-	-	0.046
7604.992	$1.1 \cdot 10^{-23}$	$2.26 \cdot 10^{-23}$	$1.34 \cdot 10^{-23}$	0.067
7605.809	$0.93 \cdot 10^{-23}$	$0.787 \cdot 10^{-23}$	$1.04 \cdot 10^{-23}$	0.081

Note: The value of S is given in units of $\text{cm}^{-1}/\text{mole} \cdot \text{cm}^{-2}$, and of γ in $\text{cm}^{-1} \cdot \text{atm}^{-1}$.

The transmittances of a vertical atmospheric path for PL radiation with various spectral composition were calculated on the basis of these data. The calculational program, briefly described in Ref. 12, was modified somewhat. To increase the speed of calculation of the transmission function $T_{\Delta\nu}$ for quasi-monochromatic radiation with laser line shape $g(\nu, \nu_0)$ along the path between the height z_1 and the height z_2 :

$$\begin{aligned}
 T_{\Delta\nu} &= \int_{\Delta\nu} g(\nu, \nu_0) T(\nu) d\nu = \\
 &= \int_{\Delta\nu} g(\nu, \nu_0) \cdot \exp\left\{-\int_{z_1}^{z_2} \sum_{i,j} K_i^j(\nu, z) dz\right\} d\nu.
 \end{aligned}
 \tag{1}$$

the following technique was used to limit the number of lines i summed over in this expression. The equivalent linewidth in the "weak line" approximation (i.e., the value $S_{ij} \cdot U_j$ where S_{ij} is the line intensity and U_j is the concentration of j th gas) was calculated for every absorption line i of the j th atmospheric gas lying within the integration interval $\Delta\nu$. For the lines, lying outside the integration interval, their contribution to the absorption coefficient at the nearest endpoint of the interval was calculated.

If the obtained values exceeded some given level, then these lines were included in the calculation. The integration was carried out using the Simpson rule with constant (for the frequency integration) or automatically chosen step (for the altitude integration). The atmospheric model for summer in the middle latitudes was used in the calculation.

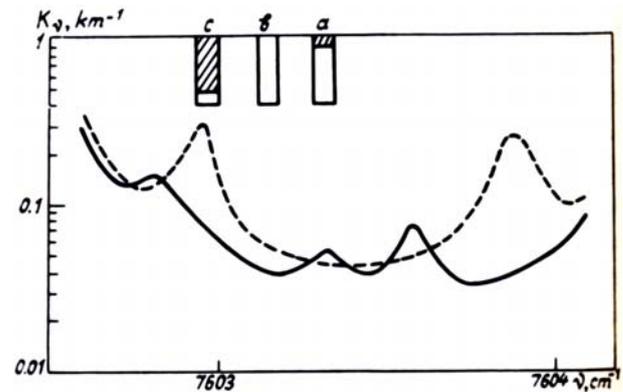


Fig. 3

Figure 3 (solid curve) shows the calculated spectral behavior of the absorption coefficient for the atmospheric boundary layer for a relative humidity of 9.8 g/kg. The dashed curve shows the results calculated using the AFGL compilation.¹¹ One can see that taking into account the experimental data on the H₂O line parameters from Refs. 2 and 10 resulted in a considerable change of the shape of the function K_ν in this spectral range. It should be noted that the calculated value of at the frequency 7603.14 cm⁻¹ corresponds to the value of the absorption cross section $\sigma = 1.07 \cdot 10^{-24} \text{ cm}^2$, which is in very good agreement with the experimental results of Refs. 10 and 13, which are equal to $1.1 \cdot 10^{-24} \text{ cm}^2$ and $1.15 \cdot 10^{-24} \text{ cm}^2$, respectively.

Let us now consider the influence of the peculiarities of the PL spectrum on the transmittance. The lasing regions of the type a , b , and c lines (the dashed part) are indicated in Fig. 3 with their boundaries corresponding to the Lorentz line width. As can be seen from Fig. 3, the minimum atmospheric absorption must be for radiation with frequency 7603.13 cm⁻¹ (region b), i.e., the photodissociation laser pumped by the plasma emission of a high-current discharge. The xenon-lamp pumped laser has slightly worse prospects (for problems of

radiation transfer) since the laser line is located nearer the H_2O line center at $\nu = 6702.82 \text{ cm}^{-1}$ due to a shift of the laser line to the lower frequency region.

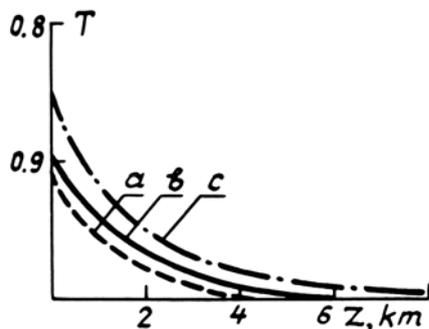


FIG. 4. Transmittance of a vertical atmospheric path from ground level to height z for radiation of various types of PL's.

Exact calculations of the transmission function for the above-considered PL lines confirm these conclusions. Figure 4 presents the results of calculations of the transmission for a vertical atmospheric path from the altitude z to the upper boundary of the atmosphere. It is seen that the atmospheric absorption doubles (from 7 to 14%) in going from the laser with type-b generation line to a laser with type-c generation line.

Thus, it is quite reasonable to use lasers operating in the region $7603.1\text{--}7603.2 \text{ cm}^{-1}$ or the region $7603.7\text{--}7603.9 \text{ cm}^{-1}$, e.g., PL pumped by radiation of a high-current discharge plasma, to reduce absorption losses attendant to the transport of radiation from an iodine-photodissociation laser through the atmosphere. Note that considerable progress in the field of superconducting materials has provided very real prospects of tuning PL radiation over a wider spectral range ($1\text{--}2 \text{ cm}^{-1}$).

REFERENCES

1. I.M. Belousova, V.D. Bodrov, V.M. Kisilev, V.M. Kurzenkov, and P.I. Krepostnov, *Zh. Eksp. Teor. Fiz.*, No. 65, 524 (1973).
2. N.F. Borisova, E.S. Bukova, K.P. Vasilevskii, and V.V. Tsukanov, in: *Abstracts of Reports at the Eighth All Union Symposium on High and Super-high Resolution Molecular Spectroscopy*, Krasnoyarsk, 1987, p. 157.
3. V.S. Zuev, V.A. Katulin, V.Yu. Nosach, and O.Yu. Nosach, *Zh. Eksp. Teor. Fiz.* **5**, No. 62, 1673 (1972).
4. V.Yu. Zalesskii, *Kvantovaya Electron.* **10**, No. 6, 1097 (1983).
5. D.J. Benard, W.E. McDermott, N.R. Pchelkin, and R. Bousek, *Appl. Phys. Lett.* **34**, 40 (1979).
6. V.A. Alekseev, G.A. Andreeva, V.N. Volkov, and E.A. Yukov, *Zh. Eksp. Teor. Fiz.* **63**, No. 2(8), 452 (1972).
7. V.N. Kurzenkov and O.P. Sklyarov, *Kvantovaya Electron.* **9**, No. 6, 1140 (1982).
8. B.L. Borovich, V.S. Zuev, V.A. Katulin, L.D. Mikheev, F.A. Nikolaev, O.Yu. Nosach, and V.B. Rosanov, *Itogi Nauki i Tekhniki, Ser. Rad iotekhnika* **15**, 155 (1978) (VINITI, Moscow).
9. O.B. Danilov, A.P. Zhevlakov, S.A. Tul'skii, and I.L. Yachev, *Kvantovaya Electron.* **9**, No. 6, 1245 (1982).
10. S.L. Bragg and J.D. Kelley, *Appl. Opt.* **26**, No 3, 506 (1987).
11. L.S. Rothman, *Appl. Opt.* **20**, No. 5, 791 (1981).
12. N.F. Borisova, V.M. Osipov, and N.I. Pavlov, *Kvantovaya Electron.* **12**, No. 12, 2505 (1985).
13. E.A. Korotchenko, A.Yu. Kurov, V.D. Nikolaev, Yu.N. Ponomarev, M.I. Svistun, and B.A. Tikhomirov, *Optika Atmosfery* **1**, No.1, 139 (1988).