Atomic total angular momentum as a measure of inversion in pulsed vapor lasers

V.M. Klimkin

Institute of Atmospheric Optics,
Siberian Branch of the Russian Academy of Sciences, Tomsk

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The possibility to produce the population inversion at self-limiting transitions of atoms and ions of chemical elements is connected with spectroscopic properties of working transitions.

Pulsed gas-discharge lasers based on transitions from resonance to metastable states of atoms and ions have passed a long way of development – more than 30 years. By now the self-limiting lasing is obtained at ~50 spectral transitions of 15 chemical elements, including *p*-, *d*-, and *f*-elements. At the same time, some negative results have not found proper explanation up to now. Therefore, the obtained data call for analysis and generalization. The goal of this paper is to pay attention of researchers to the relation between the spectroscopic properties of working transitions and the possibility of producing the population inversion, as well as the lasing efficiency in these lasers.

Most currently known self-limiting laser transitions and the ground states of the corresponding atoms and ions (44) are classified in the Table presented below. The transitions are systematized by chemical elements. In addition, lasing wavelengths and estimates of the specific output energy expressed in the number of quanta of laser transitions by a ten-grade scale are given. It is seen from the Table that there are some regularities in the classification of laser transitions and ground states of particles, as well as in the energetic efficiency of transitions. In particular, 31 laser transitions in this Table, i.e., the overwhelming majority, can be combined by the following characteristic: the total moments of atoms and ions in the metastable states are higher than in the resonant states. These transitions are characterized by high output energy. The list includes only 11 transitions, which have total moments of working states are equal to or smaller than the moments of resonant states. The output energy at these transitions is very low.

Thus, based on the available experimental data obtained from observation of self-limiting lasing, we should admit that the highly efficient laser transitions have a spectroscopic feature – the total moments of atomic particles in the metastable states should be higher than in the resonant states.

This criterion of efficiency of laser transitions can be formulated in terms of statistical weights – the statistical weight of the metastable state should be higher than that of the resonant state.

Objectively, the existence of this feature is confirmed by the negative results – the absence of lasing at some "promising" transitions from resonant to metastable levels. By promising transitions, we mean the transitions meeting five well-known conditions.[1,2] These cases can hardly be seen from the tabulated data, therefore we consider them separately.

As an example, we take the most thoroughly studied and efficient copper and gold vapor lasers. Figure 1 depicts the energy level diagrams for the copper and gold atoms, as well as the known lasing transitions of copper with $\lambda = 510$ and 578 nm and gold with 312.2 and 627 nm. The peculiarity of the lasing in the copper atom is that at one of three spectral transitions in the system of resonant and metastable states, namely, between the states with the equal moments of momentum $^2P^0_{3/2} - {}^2D_{3/2}$ at $\lambda = 570$ nm, the lasing is not observed, although these states completely meet the above conditions of the efficient lasing[1,2] and the line at $\lambda = 570$ nm is one of the brightest lines in the spontaneous emission in the copper vapor. The similar situation takes place in the gold atom at $\lambda = 506.4$ nm. Lasing is not observed also at the similar transitions in spectra of ions of the rare-earth elements Ba^+, Sr^+, and Ca^+ with equal moments of the resonant and metastable states. Thus, from analysis of the negative results, we can see some regularity, according to which the lasing, as a rule, is not observed between the states with equal moments. In the general case, it is much more difficult to obtain the inversion at these states than at the states meeting the criterion of efficient transitions formulated above. It is obvious that the resonant states with the high moment have no prospects as working levels of the highly efficient pulsed lasers.

The nature of the above regularity in the list of laser transitions and the causes of absence of lasing at $\lambda = 570$ nm Cu, 506 nm Au, and so on are in the fact that the rates of electron excitation of resonant and metastable states of atomic systems in the electric discharge are close, and the considerable inversion arises only due to the difference between total moments (statistical weights) of working states.

Table. Lasing transitions

Transition #	Wavelength, nm	Atom (ion)	Working transition	Ground state	Specific output energy,* rel. units
	1	2	3	4	5
1	854.2	CaII	$4p^2P^0_{3/2} - 3d^2D_{5/2}$	$^2S_{1/2}$	5
2	866.2	CaII	$4p^2P^0_{1/2} - 3d^2D_{3/2}$	$^2S_{1/2}$	5
3	5546.0	CaI	$4p^1P^0_1 - 3d^1D_2$	$^1S_0$	10
4	534.1	MnI	$y^6P^0_{7/2} - a^6D_{9/2}$	$^6S_{5/2}$	5
5	542.0	MnI	$y^6P^0_{5/2} - a^6D_{7/2}$	$^6S_{5/2}$	5
6	547.0	MnI	$y^6P^0_{5/2} - a^6D_{5/2}$	$^6S_{5/2}$	1
7	551.7	MnI	$y^6P^0_{3/2} - a^6D_{3/2}$	$^6S_{5/2}$	1
8	553.8	MnI	$y^6P^0_{3/2} - a^6D_{1/2}$	$^6S_{5/2}$	1
9	1290.0	MnI	$z^6P^0_{7/2} - a^6D_{9/2}$	$^6S_{5/2}$	2
10	1329.4	MnI	$z^6P^0_{7/2} - a^6D_{7/2}$	$^6S_{5/2}$	2
11	1331.9	MnI	$z^6P^0_{5/2} - a^6D_{7/2}$	$^6S_{5/2}$	2
12	1362.7	MnI	$z^6P^0_{5/2} - a^6D_{5/2}$	$^6S_{5/2}$	1
13	1386.4	MnI	$z^6P^0_{3/2} - a^6D_{3/2}$	$^6S_{5/2}$	1
14	1399.7	MnI	$z^6P^0_{3/2} - a^6D_{1/2}$	$^6S_{5/2}$	1
15	452.9	FeI	$x^3D^0_4 - a^5P_3$	$^5D_4$	1
16	510.6	CuI	$4p^2P^0_{3/2} - 4s^{22}D_{5/2}$	$^2S_{1/2}$	10
17	578.2	CuI	$4p^2P^0_{1/2} - 4s^{22}D_{3/2}$	$^2S_{1/2}$	9
18	1032.7	SrII	$5p^2P^0_{3/2} - 4d^2D_{5/2}$	$^2S_{1/2}$	5
19	1091.4	SrII	$5p^2P^0_{1/2} - 4d^2D_{3/2}$	$^2S_{1/2}$	5
20	6456.0	SrI	$5p^1P^0_1 - 4d^1D_2$	$^1S_0$	10
21	614.2	BaII	$6p^2P^0_{3/2} - 5d^2D_{5/2}$	$^2S_{1/2}$	3
22	649.7	BaII	$6p^2P^0_{1/2} - 5d^2D_{3/2}$	$^2S_{1/2}$	3
23	1130.3	BaI	$6p^1p^0_1 - 5d^3D_2$	$^1S_0$	10
24	1499.9	BaI	$6p^1P^0_1 - 5d^1D_2$	$^1S_0$	10
25	312.2	AuI	$6p^2P^0_{3/2} - 6s^{22}D_{5/2}$	$^2S_{1/2}$	1
26	627.8	AuI	$6p^2P_{1/2} - 6\mathrm{s}^{22}D_{3/2}$	$^2S_{1/2}$	10
27	535.0	TlI	$7s^2S_{1/2} - 6p^2P^0_{3/2}$	$^3P_{1/2}$	1
28	363.9	PbI	$6p7\mathrm{s}^3P^0_1 - 6s^{23}P_1$	$^3P_0$	1
29	405.7	PbI	$6p7\mathrm{s}^3P^0_1 - 6s^{23}P_2$	$^3P_0$	2
30	722.9	PbI	$6p7s^3P^0_1 - 6p^{21}D_2$	$^3P_0$	5
31	472.2	BiI	$7s^4P_{1/2} - 6p^{32}D^0_{3/2}$	$^4S_{3/2}$	1
32	664.5	EuII	$z^9P_5 - a^9D^0_6$	$^9S_4$	1
33	1001.9	EuI	$z^7P_4 - a^7D^0_5$	$^9S_4$	1
34	1016.8	EuII	$z^7P_4 - a^7D^0_4$	$^9S_4$	1
35	1361.1	EuII	$z^9P_4 - a^7D^0_5$	$^9S_4$	1
36	1664.8	EuI	$y^8P_{5/2} - a^8D^0_{7/2}$	$^8S_{7/2}$	2
37	1760.1	EuI	$y^8P_{9/2} - a^8D^0_{11/2}$	$^8S_{7/2}$	2
38	4321.4	EuI	$y^8P_{9/2} - b^8D^0_{11/2}$	$^8S_{7/2}$	2
39	5066.1	EuI	$y^8P_{7/2} - b^8D^0_{9/2}$	$^8S_{7/2}$	2
40	5430.7	EuI	$y^8P_{7/2} - b^8D^0_{5/2}$	$^8S_{7/2}$	1
41	6059.2	EuI	$y^8P_{5/2} - b^8D^0_{3/2}$	$^8S_{7/2}$	1
42	1345.3	YbII	$5p^2P^0_{3/2} - 5d^2D_{3/2}$	$^2S_{1/2}$	1
43	1649.8	YbII	$5p^2P^0_{3/2} - 5d^2D_{5/2}$	$^2S_{7/2}$	2
44	2437.7	YbII	$5p^2P^0_{1/2} - 5d^2D_{3/2}$	$^2S_{1/2}$	2

Note. * measured in the number of quanta.

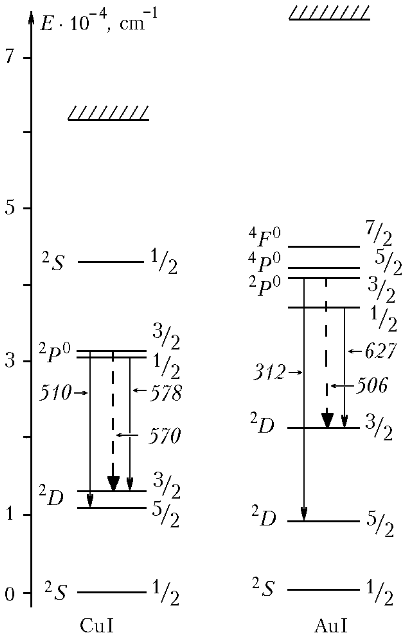

Fig. 1. Spectral transitions in systems of resonant and metastable states of copper and gold atoms.

This was demonstrated experimentally in Ref. 3. The idea of this experiment was the following. If we assume that metastable copper levels are not excited by electron impact, then the saturating light field in the laser cavity redistributes the populations of the working states in the inverse proportion to statistical weights. In this case, the population of the resonant copper level must decrease by 60% for the level $^2P_{3/2}$ and by 66% for the level $^2P_{1/2}$ (Ref. 4). This decrease corresponds to significant effect of quenching of the spontaneous emission, as observed at the corresponding resonant lines in the spectrum of working particles or at the working transitions over the tube radius. In Ref. 4 it was found experimentally that in the repetitively pulsed laser in the presence of the saturating optical field, the decrease in populations of the resonant Cu levels is not more than 14–15%, i.e., much less than the expected decrease. The analysis[4] has shown that the decrease (14–15%) in the population of the resonant state corresponds to the intense electron excitation of metastable states. In particular, it was found that the constants of the excitation rates of the resonant and metastable states are close for the Cu vapor laser.

For the model of an “ideal”[4] self-limiting laser (only direct electron excitation of working states is taken into account), the constants of the ultimate pumping rates k_r needed for obtaining the population inversion are determined as[4]:

$$k_r \geq k_m \frac{g_r}{g_m},$$

where g_m and g_r are the statistical weights of the working levels. With the increase of the moment of the ground state of atomic particles, $g_r/g_m \to 1$. Consequently, for elements with the high moment of the ground state, large differences between the excitation constants k_r and k_m are needed to achieve the lasing. Thus, with allowance for the rules of spectral transitions selection, we obtain one more criterion of the efficient laser transition – the efficient transition should be expected in atomic systems with the minimum total moment of atom or ion in the ground state, i.e., with $J = 0, 1/2$. As it is seen from the data given in the fourth column of the Table, most listed atoms and ions meet this criterion, but there are some exclusions. In particular, the atomic structures of manganese ($J = 5/2$), bismuth ($J = 3/2$), and europium ($J = 7/2$) are unfavorable for highly efficient lasing. The output energy is not high for these elements.

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