Peculiarities of collisional excitation transfer with excited screened energy levels of atoms

V. A. Gerasimov,[1] V. V. Gerasimov,[1,2,*] and A. V. Pavlinskiy[3]
[1]*Institute of Atmospheric Optics, Siberian Branch of the Russian Academy of Sciences, Tomsk, 634055 Russia*
[2]*Department of Physics, Tomsk State University, Tomsk, 634050 Russia*
[3]*Institute of High Current Electronics, Siberian Branch of the Russian Academy of Sciences, Tomsk, 634055, Russia*
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We report an experimental discovery of deviations from the known regularities in collisional excitation transfer processes for metal atoms. The collisional excitation transfer with excited screened energy levels of thulium and dysprosium atoms is studied. The selecting role of the screening 6s shell in collisional excitation transfer is shown.

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The collisional excitation transfer, and particularly the excitation transfer in collisions of metal atoms between themselves and metal atoms with atoms of inert gases, is one of the most studied processes in atomic collisions physics. These processes include

$$M_i^* + M_0 \rightleftarrows M_k^* + M_0 \pm \Delta E, \tag{1}$$

$$M_i^* + R \rightleftarrows M_k^* + R \pm \Delta E, \tag{2}$$

where M_i^*, M_k^*, M_0 are the metal atoms in the i and k excited states and the ground state respectively, R is the atom of an inert gas, and ΔE is the energy defect between the i and k states.

Studies of the processes mentioned above were performed mainly for atoms with unscreened excited atomic levels. A considerable amount of experimental data obtained in studies of mentioned processes by other authors, shows the following principal regularity. The cross sections of processes (1) and (2) are defined mainly by energy defect ΔE, and cross sections of both processes decrease with increasing ΔE. However the cross section $\sigma_{M\text{-He}}$ of process (2) decreases significantly faster with increase of ΔE than the cross section $\sigma_{M\text{-}M}$ of process (1) and, hence, the efficiency of process (2) is always lower than that of Eq. (1) at the same ΔE. Thus, the contribution of process (2) in excitation transfer becomes negligible already at low ΔE. For example, Table I contains the ratio of cross sections of processes (1) and (2) depending on ΔE. The cross sections are taken from Refs. [1–6]. We can see from Table I that the ratio of cross sections $\sigma_{M\text{-}M}/\sigma_{M\text{-He}}$ of processes (1) and (2) increase with the increasing ΔE. For example, for Rb at $\Delta E = 238$ cm^{-1} $\sigma_{\text{Rb-Rb}} = 740\sigma_{\text{Rb-He}}$.

The processes (1) and (2) with excited screened atomic levels remain unstudied. These atoms with a screened shell include rare-earth metals (REM) with an incomplete $4f$ shell (Ce-Yb). They are characterized by the presence of excited 5 d states screened by the outer 6s shell. Interest to processes (1) and (2) is also defined by studies on broadening of screened transitions in REM atoms due to collisions of these atoms with atoms of inert gases [7,8]. It is shown experimentally in Refs. [7,8] that screened transitions broaden considerably less than unscreened transitions.

In this work, we present the results of an experimental study of collisional excitation transfer processes (1) and (2) with excited screened energy levels of REM atoms. To carry out our experiment we use the fact that some REM atoms (Sm, Eu, Dy, Ho, Er, Tm, Yb) have laser transitions when they are excited by electron impact in gas discharge. A distinctive feature of these lasers is that the most part of upper laser levels has the same parity that the ground state, and thus are not populated by electron impact in discharge [9–11]. The most likely way of upper level population is the collisional excitation transfer from close-lying resonant levels. Figure 1 shows schematically the population inversion creation. Here 1 and 2 are lower and upper laser levels respectively. Resonant level 3 is excited by electron impact from the ground level 0. Level 2 is populated in collisions (1) and (2) from level 3.

Laser amplification I and, therefore, lasing power P is known to be defined by inversion population density. In this case,

$$I \equiv N_2 - \frac{g_2}{g_1} N_1, \tag{3}$$

where N_1 and N_2 are the populations and g_1 and g_2 are the statistical weights of lower and upper laser levels, respectively. As a rule, $N_2 \gg N_1$, therefore the lasing power is defined only by the population of upper laser level N_2. So we

TABLE I. Cross sections ratio of collisional excitation transfer processes for metal-metal $\sigma_{M\text{-}M}$ (1) and metal-helium $\sigma_{M\text{-He}}$ (2) for different energy defects ΔE.

Element	ΔE (cm^{-1})	$\sigma_{M\text{-}M}/\sigma_{M\text{-He}}$
Na	17	3.8
K	58	9.04
Rb	78	14.41
Rb	238	740
Cs	554	$\sim 10^5$

*GVVSnake@mail.ru

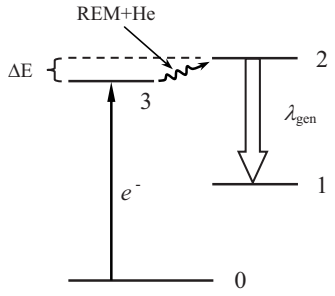

FIG. 1. Scheme of populating of upper laser level 2 by the collisional excitation transfer from close-lying resonant level 3. λ_{gen} is the laser transition wavelength; ΔE is the energy defect.

can make a conclusion about the efficiency of processes (1) and (2) measuring lasing power.

For our experiment, we have chosen a gas-discharge thulium vapor laser. Helium and neon were used as buffer gases. Thulium was selected due to the following reasons.

(1) There are many (about 20) laser transitions [11] with different energy defects (ΔE=30–500 cm^{-1}) between upper (acceptors) and close-lying resonant (donors) laser levels in thulium atom.

(2) The excitation efficiency of screened $5d$ levels from the ground state by electron impact is comparable to the efficiency of unscreened level excitation.

To find out peculiarities of the collisional excitation transfer with screened and unscreened excited energy levels of atoms we studied two strongest laser transitions with wavelengths λ_1=1310.06 nm and λ_2=1495.78 nm (Fig. 2). The first laser transition with λ_1 occurs between the upper laser level UL1 with energy $E_{UL1}=22902_{(J=13/2)}$ cm^{-1} ($4f^{12}6s^26p_{1/2}$) and the lower laser level LL1 with energy $E_{LL1}=15271_{15/2}$ cm^{-1} ($4f^{12}5d_{3/2}6s^2$). Population of LL1 level from the ground state by electron impact is prohibited by the selection rules (ΔJ=4). The second laser transition with λ_2 occurs between the upper laser level UL2 with energy $E_{UL2}=25520_{11/2}$ cm^{-1} ($4f^{13}5d6s$) and the lower laser level LL2 with energy $E_{LL2}=18837_{9/2}$ cm^{-1} ($4f^{12}5d_{5/2}6s^2$) [12]. The donor level for the transition with λ_1 is the resonant screened level R1 with energy $E_{R1}=22929_{5/2}$ cm^{-1} ($4f^{12}5d_{5/2}6s^2$), close-lying to UL1 (ΔE=27 cm^{-1}). The donor level for the transition with λ_2 is the resonant unscreened level R2 with energy $E_{R2}=25656_{5/2}$ cm^{-1} ($4f^{13}6s6p$), close-lying to UL2 (ΔE=136 cm^{-1}).

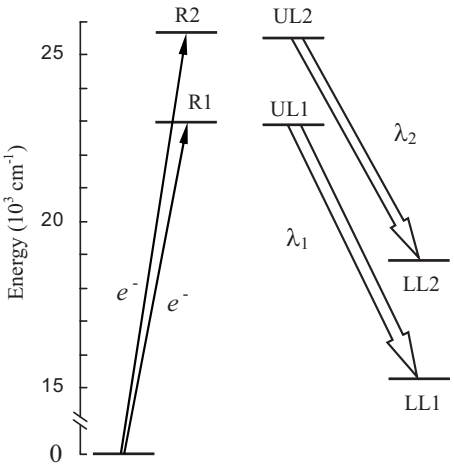

FIG. 2. Populating of upper laser levels UL1 and UL2 by the collisional excitation transfer from close-lying screened resonant level R1 and unscreened level R2, respectively, Doubled arrows show the laser transitions.

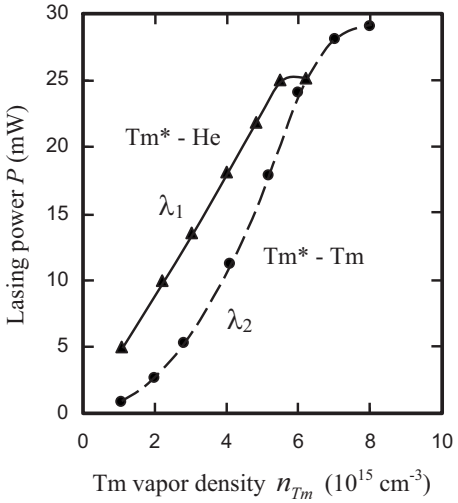

FIG. 3. Lasing power vs Tm vapor density. The solid line corresponds to the excitation transfer in collisions of Tm^* atoms with screened excited $5d6s^2$ level and the ground state He atoms. The dashed line corresponds to the excitation transfer in collisions of Tm^* atoms with unscreened excited $6s6p$ level and the ground state Tm atoms.

Thulium vapor was created in an aluminum oxide gas-discharge tube with the diameter of 20 mm and the active zone length of 400 mm. Pieces of metallic thulium with purity of 99.83% were placed inside the tube and distributed over the entire tube length. The tube was heated with an outer high-temperature heating coil. The exciting pulsed discharge repetition rate was of 100 Hz. A 2.35 nF storage capacitor was charged up to 10 kV and then discharged to the tube via a hydrogen-filled thyratron of the TGI1-1000/25 type. Such a discharge could not have any influence on the tube temperature. The tube temperature was changed with a heating coil ranging from 1000 °C to 1150 °C that corresponded to thulium vapor density change from 1×10^{15} cm^{-3} to 9×10^{15} cm^{-3}. This temperature range complies with the condition $\Delta E < kT_g$, where k is the Boltzmann constant and T_g is the gas temperature. The buffer gas was helium at the pressure of 2 Torr.

The spectral emission lines were selected using a tunable cavity with a 300 grooves/mm diffraction grating as the totally reflecting mirror and the output transparent glass plate. The experimental setup is described in details in Ref. [13]. The lasing power was measured with a calorimetric detector IMO-2M with total accuracy of 7%.

The experimental results are shown in Fig. 3. We can see that functional dependences of the lasing power from Tm vapor density are different for transitions with λ_1 and λ_2. The dependence $P(n_{Tm})$ for spectral line with λ_1 has a linear

profile [i.e., $P(n_{\mathrm{Tm}}) \sim n_{\mathrm{Tm}}$], while the dependence $P(n_{\mathrm{Tm}})$ for spectral line with λ_2 has a quadratic one [$P(n_{\mathrm{Tm}}) \sim (n_{\mathrm{Tm}})^2$]. This fact points out that population inversion on the transition with λ_1 is formed in process (2), while for the transition with λ_2 it is process (1). The distinctive feature of the results observed is that for the laser transition with λ_1 (its upper level is populated from the screened resonant level) the efficiency of process (2) is significantly higher than the efficiency of process (1), i.e., $\sigma_{\mathrm{Tm\text{-}Tm}} < \sigma_{\mathrm{Tm\text{-}He}}$.This fact is inconsistent with the known regularities in atomic collision physics described above. Note that the upper parts of both curves (Fig. 3) have significant deviations from functional dependences due to the decrease of electron temperature (i.e., the decrease of population rate of the resonant levels by electron impact from the ground state) with the increase of Tm vapor density. Carrying out of direct experiment to discover the contribution of Tm^*-Tm collisions to excitation transfer in the absence of buffer gas is impossible due to some technical problems. The main problem is the rapid metal vapor condensation on the output windows of gas-discharge laser tube in the absence of buffer gas.

Thus, to confirm the discovered regularities, we carried out the same studies for the laser transition of thulium from $E_{\mathrm{UL3}}=22559_{11/2}\ \mathrm{cm}^{-1}$ $(4f^{13}5d6s)$ to $E_{\mathrm{LL3}}=15587_{11/2}\ \mathrm{cm}^{-1}$ $(4f^{12}5d_{3/2}6s^2)$ with wavelength $\lambda_3=1433.97$ nm and the laser transition of dysprosium from $E_{\mathrm{UL4}}=24430_7\ \mathrm{cm}^{-1}$ $(4f^{10}5d6s)$ to $E_{\mathrm{LL4}}=12655_7\ \mathrm{cm}^{-1}$ $(4f^9 5d6s^2)$ with $\lambda_4=849.015$ nm. Populating of LL3 level from the ground state by electron impact is prohibited by the selection rules ($\Delta J=2$). The upper laser level UL3 in a thulium atom is populated from the resonant screened level R3 with energy $E_{\mathrm{R3}}=22791_{7/2}\ \mathrm{cm}^{-1}$ $(4f^{12}5d_{3/2}6s^2)$ and energy defect $\Delta E=232\ \mathrm{cm}^{-1}$. The upper laser level UL4 in a dysprosium atom is populated from the resonant unscreened level R4 with energy $E_{\mathrm{R4}}=24709_7\ \mathrm{cm}^{-1}$ $(4f^{10}6s6p)$ and energy defect $\Delta E=278.7\ \mathrm{cm}^{-1}$. This experiment has similarly showed that functional dependences of lasing power P for transition with λ_3 in a Tm atom is linear and for one with λ_4 in a Dy atom is quadratic.

No differences in functional dependences for $P(n_{\mathrm{Tm}})$ and $P(n_{\mathrm{Dy}})$ were observed when helium was replaced with neon. When the upper laser level is populated from the unscreened resonant level, we have a quadratic dependence for $P(n_{\mathrm{Tm}})$ and $P(n_{\mathrm{Dy}})$ and it means that they are not affected by process (2). The experimental data shown in Table I bring us to a conclusion that for $\Delta E=136\ \mathrm{cm}^{-1}$ in a Tm atom and for $\Delta E=278.7\ \mathrm{cm}^{-1}$ in a Dy atom cross section of process (2) is much lower than that of process (1).

Summarizing the above results, we have drawn the following conclusions.

(1) The collisional excitation transfer from excited screened levels of thulium atoms occurs in collisions with atoms of inert gases (He, Ne).

(2) The screening 6*s* shell prevents the excitation transfer in collisions with own thulium atoms.

(3) Processes of the collisional excitation transfer with unscreened excited levels of thulium and dysprosium atoms are the same as for other metals [1–6].

We do not exclude that the collisional excitation transfer with excited screened levels is the same for other REM atoms. It should be noted that linear dependence (instead of expected quadratic one) was also observed in Ref. [14] for the intensity of nonresonant fluorescence in changes of thulium and helium vapor densities. The optically excited $4f^{12}5d6s^2$ state was studied in that work [14]. No explanations for that fact were given.

[1] J. Huennekens and A. Gallagher, Phys. Rev. A **27**, 1851 (1983).
[2] J. Pitre and L. Krause, Can. J. Phys. **45**, 2671 (1967).
[3] M. Czajkowski, D. A. McGillis, and L. Krause, Can. J. Phys. **44**, 741 (1966).
[4] R. Boggy and F. A. Franz, Phys. Rev. A **25**, 1887 (1982).
[5] P. Münster and J. Marek, J. Phys. B **14**, 1009 (1981).
[6] T. J. Beahn, W. J. Condell, and H. I. Mandelberg, Phys. Rev. **141**, 83 (1966).
[7] V. D. Vedenin, Opt. Spektrosk. **70**, 956 (1991).
[8] N. I. Agladze *et al.*, Opt. Spektrosk. **63**, 12 (1987).
[9] V. A. Gerasimov, Opt. Spektrosk. **87**, 156 (1999) [Opt. Spectrosc. **87**, 144 (1999)].
[10] V. A. Gerasimov and L. N. Starkova, Opt. Spektrosk. **92**, 335 (2002) [Opt. Spectrosc. **92**, 298 (2002)].
[11] V. A. Gerasimov, B. P. Yunzhakov, Kvant. Elektron. **16**, 2386 (1989) [Sov. J. Quantum Electron. **19**, 1532 (1989)].
[12] W. C. Martin, R. Zalubas, and L. Hagan, Natl. Stand. Ref. Data Ser. (U.S., Natl. Bur. Stand.) **60**, 344 (1978).
[13] V. A. Gerasimov and L. N. Starkova, Pis'ma Zh. Tekh. Fiz. **27**, 51 (2001) [Tech. Phys. Lett. **27**, 65 (2001)].
[14] E. B. Aleksandrov *et al.*, Opt. Spektrosk. **60**, 881 (1986).