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Relaxation of the $6p\,^2P^0_{3/2}$ metastable state in a self-terminating thallium laser

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***Abstract*. We present the results of an experimental study of a Tl vapour/Ar mixture as an active medium. Under optical excitation of thallium to the $6d\,^2D_{3/2}$ state, we have achieved lasing at $\lambda = 1.3$ μm ($7p\,^2P^0_{1/2} - 7s\,^2S_{1/2}$ transition) and 535 nm ($7s\,^2S_{1/2} - 6p\,^2P^0_{3/2}$) through cascade transitions from the $6d\,^2D_{3/2}$ state to the upper laser levels. The electron deexcitation rate of the $6p\,^2P^0_{3/2}$ metastable state is no slower than 1.5×10^{-8} cm^3 s^{-1}, meaning that this process cannot prevent self-terminating laser action at the $7s\,^2S_{1/2} - 6p\,^2P^0_{3/2}$ transition under electron-beam pumping at pulse repetition rates up to 100 kHz.**

Keywords: *thallium laser, optical pumping, relaxation of metastable states.*

1. Introduction

Lasers on the thallium $7s\,^2S_{1/2} - 6p\,^2P^0_{3/2}$ transition [1] are expected to be attractive coherent radiation sources for two reasons: 535-nm laser radiation is fairly convenient for a variety of applications, and the operating temperature of thallium lasers is about 800 °C, which allows inexpensive silica tubes to be used in the fabrication of such lasers. In spite of these obvious advantages, the properties of thallium lasers have been examined in a few papers [2], and there are no reports devoted to their potential applications.

One possible reason for this is that such lasers are difficult to operate, in particular because of the short lifetime (~ 14 ns) of the $7s\,^2S_{1/2}$ resonance state at the laser transition [3]. So thallium lasers require more perfect power supplies than do conventional copper vapour lasers. Even under the assumption that the pump energy density is the same for copper vapour lasers and thallium lasers, the latter require an order of magnitude higher peak pump power density at pump pulse durations no longer than 10 – 15 ns. Like with other self-terminating metal atom lasers, this problem is difficult to resolve in the case of a gas-discharge Tl laser.

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An alternative approach is to excite thallium vapour with an electron beam generated by an open discharge. Recently proposed laser cells with ceramic – metal cathodes [4] are capable of operating at ~ 1000 °C and ensure an electron pulse duration of ~ 15 ns, which suggests the possibility of creating an electron-beam-pumped Tl laser.

In designing a thallium laser, it is desirable to know the basic parameters of thallium vapour as an active medium. Such data are however missing. A key issue in creating a high-power laser is its ability to operate at high pump pulse repetition rates. To this end, atoms in the metastable lower laser level must rapidly relax to the ground state. One general – and rather efficient – deexcitation mechanism for the lower laser levels in self-terminating lasers is electron deexcitation [5]. An important feature of the thallium atom is that the $6p\,^2P^0_{3/2}$ metastable state, a fine-structure component of the $6p\,^2P^0$ ground state, lies well above (~ 1 eV) the other component ($6p\,^2P^0_{1/2}$). Given that the electron deexcitation efficiency in such systems had not been reported previously, the purpose of this work was to study the effect of the relaxation of the thallium $6p\,^2P^0_{3/2}$ metastable state on the lasing process in thallium vapour.

2. Investigation technique and experimental setup

Figure 1 shows a diagram of the thallium atom levels of interest in this study. Repetitive 276.7-nm laser pulses excite thallium atoms to the $6d\,^2D_{3/2}$ state. The excited atoms are stimulated to decay to the $7s\,^2S_{1/2}$ resonance state and then undergo a laser transition to the $6p\,^2P^0_{3/2}$ metastable state ($\lambda = 535$ nm). IR lasing at the $6d\,^2D_{3/2} - 7p\,^2P^0$ and $7p\,^2P^0 - 7s\,^2S_{1/2}$ transitions differs from visible lasing at the $7s\,^2S_{1/2} - 6p\,^2P^0_{3/2}$ transition in that the lower laser levels of the first two transitions have much shorter lifetimes than do their upper laser levels. In contrast, the lower level of the third transition is metastable, and only self-terminating lasing is possible. The study of the IR and visible lasing parameters under various conditions provides insight into the relaxation processes involving thallium atoms in their metastable state.

Our experimental setup is shown schematically in Fig. 2. The 40-W output pulses from a 510.6-nm copper vapour laser with a repetition rate $f = 11$ kHz are amplified and directed to two transversely pumped Pyrromethene 556 dye laser cells. The first cell is a master oscillator (MO) utilising the Hänsch scheme with a reflector – lens (about 30-fold) beam expander [6]. After the expander, the required wavelength is selected by an evacuated Fabry – Perot etalon with

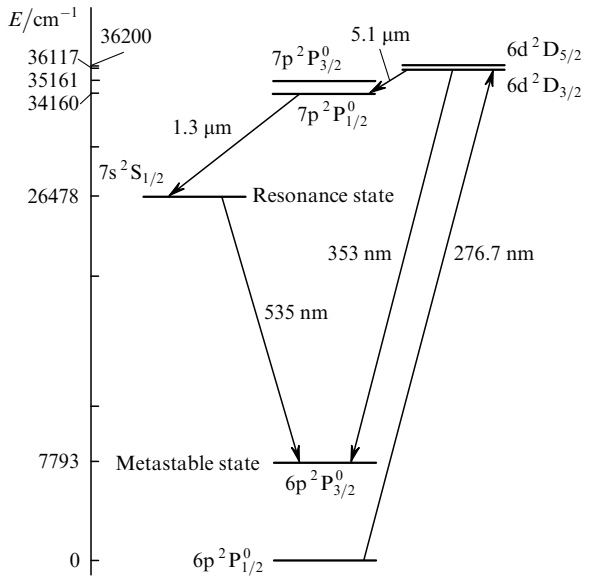

Figure 1. Diagram of thallium energy levels and laser transitions.

a 97.5 % reflectance of the mirrors at 553 nm and a cavity length of 7.5 mm, and is redirected to the cell by a diffraction grating (angle of incidence, $\sim 85^\circ$). One of the faces of a quartz wedge is used as the output coupler. At a 15-W pump power, the dye laser in this cavity has an output power from 50 to 100 mW and a linewidth of ~ 75 MHz. The laser frequency is maintained constant or tuned by fixing or varying the pressure in the etalon. At a fixed pressure, the reproducibility of the lasing wavelength during repeated on/off cycling of the pump and dye lasers is within the stated accuracy (10^{-7}) of the wavelength meter we use (Angstrem, Novosibirsk), with no changes in dye master oscillator settings.

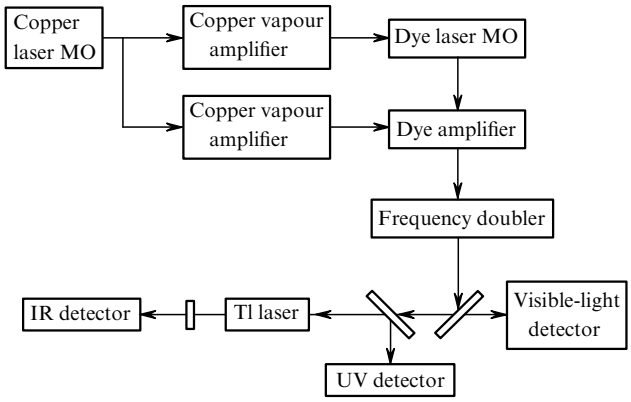

Figure 2. Schematic diagram of the experimental setup.

After the master oscillator, the beam is sent to an amplifier and then to a BBO frequency doubler. At a dye amplifier output power of 0.5–1 W and UV pulse duration of ~ 10 ns, the 553.4 to 276.7 nm conversion efficiency was 20 %–25 %. The beam was then collimated to a diameter of 1 mm and directed to an externally heated BeO ceramic tube with an inner diameter $D = 1.5$ cm and a heated length of 40 cm. The tube contained metallic thallium on four spaced tantalum substrates and was enclosed in a vacuum-tight quartz casing. In addition, the casing contained tantalum foil electrodes, which enabled periodic discharges to be generated in the Tl vapour/Ar mixture. In the case of electron-beam pumping, the choice of argon as the buffer gas is not critical and is dictated by the higher electron beam generation efficiency. The gas-discharge power supply was synchronised with the pump laser power supply, so that the pulse repetition rate was the same for optical and discharge excitation, with the possibility of varying the optical pulse delay over the entire quiescent period.

3. Results and discussion

At ordinary UV pump powers, $P_{UV} \sim 50$ mW, the thallium laser is superfluorescent starting at a metal vapour pressure of $\sim 10^{-3}$ Torr, with lasing at $\lambda = 1.3$ μm ($7p\,^2P^0_{1/2}$–$7s\,^2S_{1/2}$) and 535 nm ($77s\,^2S_{1/2} - 6p\,^2P^0_{3/2}$). Note that the 353-nm ($6d\,^2D_{3/2} - 6p\,^2P^0_{3/2}$) lasing is more than two orders of magnitude weaker than the visible lasing. Clearly, superfluorescence must also develop at the $6d\,^2D_{3/2}$–$7p\,^2P^0$ transitions around 5 and 10 μm. We however did not study lasing in this spectral region for lack of a suitable detector. At an about 90 % absorption of the UV pump pulse energy, the conversion efficiency from the absorbed pump energy to the lasing output is 20 %–25 %. Most of the superfluorescent emission (about 90 %) is directed along the pump beam. Under the optimal conditions, using mirrors highly reflecting at the Tl lasing wavelengths or a plane–parallel plate has an insignificant effect on the overall output power but enables redistribution of the power between the ends of the laser tube.

Figure 3 shows the relative output power of the Tl laser at 535 nm (P_{GR}) and 1.3 μm (P_{IR}) as a function of argon pressure p_{Ar} at a ground-state thallium atom concentration of $\sim 10^{13}$ cm^{-3} (vapour pressure, $\sim 6.5 \times 10^{-3}$ Torr). The curves are seen to differ in shape. The reason for this is that raising the buffer gas pressure inhibits diffusion of the thallium atoms in the $6p\,^2P^0_{3/2}$ metastable state, thereby impeding the 535-nm lasing process. For $p_{Ar} \geqslant 45$ Torr, the concentration of thallium atoms in this state is so high that no lasing occurs.

The increased population of the Tl $6p\,^2P^0_{3/2}$ metastable level inhibits the decay of the $7s\,^2S_{1/2}$ lower (for IR lasing)

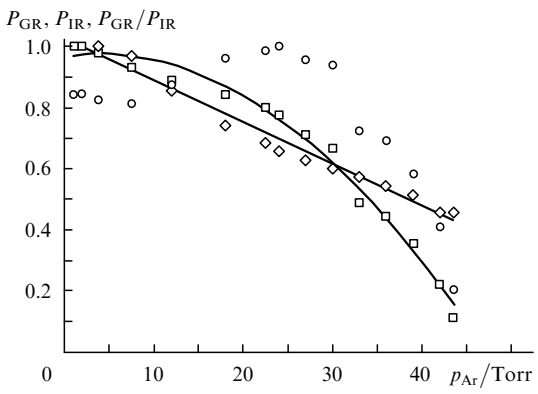

Figure 3. Relative output power of the Tl laser at 535 nm (□, P_{GR}) and 1.3 μm (◇, P_{IR}) and the P_{GR}/P_{IR} ratio (○) as functions of argon pressure p_{Ar} ($p_{Tl} \sim 6.5 \times 10^{-3}$ Torr).

level through imprisonment, but lasing persists up to $p_{Ar} \approx 80$ Torr. At $P_{UV} \sim 50$ mW and a 50 % pump power conversion efficiency, the concentration of atoms in the metastable state after a lasing pulse is $N = 0.5P_{UV} \times (fV_1hc/\lambda)^{-1} \sim 2 \times 10^{13}$ cm^{-3} (V_1 is the interaction volume). This corresponds to a 535-nm absorption coefficient $k_0 = \sigma_0 N \sim 0.8 \times 10^2$ cm^{-1} for Doppler broadening (σ_0 is the absorption cross section). According to Holstein [7], this increases the lifetime of the $7s\,^2S_{1/2}$ level through reabsorption by a factor of g^{-1} [$g = 1.6/\{(k_0D/2)[\pi \log(k_0D/2)]^{1/2}\} = 0.29$ is the extraction factor], i.e., to ~ 48 ns, which is close to the lifetime of the $7p\,^2P^0$ state (~ 63 ns [8]). All this impedes the IR lasing process, without completely suppressing it.

Increasing the inner tube diameter to a typical value of 1 cm increases the diffusion time of metastable thallium atoms in argon by a factor of 100. Consequently, to maintain laser action, p_{Ar} should be no higher than a fraction of a torr, which would have an adverse effect on the operational life of the laser because of the rapid removal of atoms to the cold zones of the tube. Moreover, at such argon pressures high-power electron beams cannot be generated. Therefore, in tubes with $d > 1$ cm the metastable state should be relaxed in a different way, in particular through electron deexcitation.

To ascertain whether this state can be depopulated through electron deexcitation, we examined electron generation in a Tl–Ar medium. To this end, a 0.85-μs discharge was struck in the tube before each UV laser pulse, and we assessed the effect of pumping conditions on 535-nm lasing parameters. Figure 4 shows the relative 535-nm lasing power at $p_{Ar} \approx 45$ Torr as a function of the separation between the optical pump pulse and the trailing edge of the discharge pulse. As seen, the 535-nm lasing is fully restored 25 μs after the discharge.

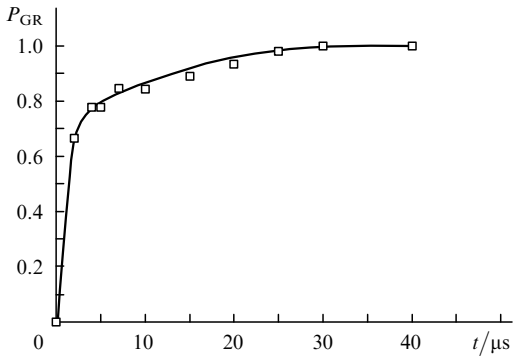

Figure 4. Relative 535-nm output power as a function of the pump pulse delay relative to the discharge pulse; $p_{Ar} \approx 45$ Torr, $p_{Tl} \sim 6.5 \times 10^{-3}$ Torr.

The rate constant k_e for electron deexcitation of the metastable state of thallium can be evaluated from the relation $k_e n_e = 4.6\tau_r^{-1}$ (τ_r is the time constant for deexcitation of the metastable state), which corresponds to a 99 % relative lasing output ($t = 25$ μs). Concentration n_e can be estimated from the discharge energy (under the assumption that half of the energy goes into ionisation):

$$n_e = \frac{0.5 \int UI\mathrm{d}t}{E_i V_2} = 1.2 \times 10^{13}\ \mathrm{cm}^{-3},$$

where U is the discharge voltage, I is the discharge current, E_i is the ionisation potential of argon, and V_2 is the discharge volume. Then, we have $k_e = 1.5 \times 10^{-8}$ cm^3 s^{-1}. At the same time, from the relation $k_e n_e = \tau_2^{-1} = 5 \times 10^5$ s^{-1}, where τ_2 is the time delay needed to restore the lasing output to a level of $1 - e^{-1} = 0.63$, we obtain $k_e \sim 4 \times 10^{-8}$ cm^3 s^{-1}. These values of k_e are typical of the deexcitation of the metastable state in self-terminating lasers [5]. Therefore, at moderate electron-beam pump intensities corresponding to $n_e \sim 10^{14}$ cm^{-3} [4], the deexcitation rate of the metastable state is high enough not to prevent lasing up to $f = 100$ kHz.

4. Conclusions

Atomic thallium lasing in an Ar–Tl medium was achieved at $\lambda = 1.3$ μm ($7p\,^2P^0_{1/2} - 7s\,^2S_{1/2}$ transition) and 535 nm ($7s\,^2S_{1/2} - 6p\,^2P^0_{3/2}$) through electron cascading from the $6d\,^2D_{3/2}$ state to the upper laser levels. The deexcitation rate of the $6p\,^2P^0_{3/2}$ metastable state is high enough not to prevent lasing at the $7s\,^2S_{1/2} - 6p\,^2P^0_{3/2}$ transition under electron-beam pumping up to 100 kHz.

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