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Alternate oscillations of self-terminating and recombination lasers in univalent calcium and strontium ions*

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The univalent calcium and strontium ions have been confirmed as ideal lasing substances both for self-terminating laser and recombination laser by theoretically analysing their energy level structures and lasing mechanisms. With the optimization of the excitation circuit and the improvement of the laser cavity as well as the laser discharge tube, the alternate laser oscillations of the two laser mechanisms were successfully realized by longitudinal pulsed discharge in mixture vapours of helium and univalent ions of calcium or strontium, respectively. The dependences of laser performance on working parameters, together with the characteristics of the photoelectric pulse waveforms were elementally studied and analysed.

Keywords: univalent calcium and strontium ions, self-terminating laser, recombination laser, alternate laser oscillations

PACC: 4255H, 4260D, 4260H

1. Introduction

The study on self-terminating metal vapour lasers has made a great progress in the past several decades since Fowles and Silfvast first reported pulsed lead vapour laser in 1965.[1] Subsequently, Walter *et al* made further analysis of the mechanisms of this class of lasers and summarized five criteria that the self-terminating laser should follow for efficient operation when he first reported copper and calcium ion vapour lasers in 1966.[2] In the same paper, he also predicted the strontium ionic self-terminating laser at 1.03/1.09μm wavelengths due to transitions from the resonance levels $5p^2P^0_{3/2,1/2}$ to the metastable states $4d^2D_{5/2,3/2}$. It is well known that typical representatives of metal atomic and ionic self-terminating lasers are copper vapour laser, calcium and strontium ionic lasers, respectively. These lasers have found many applications such as in medicine and infrared technology. This class of lasers should be operated in pulsed discharge mode, because the lower laser level is the metastable state, laser transition and electron-species collisions will result in population accumulation in the lower laser level and self-termination of lasing. The key on the discharge excitation is that the current pulse rise time should be shorter than the radiative lifetime of the laser transition.

Nowadays, calcium and strontium ionic recombination lasers are probably the only ones among pulsed gas lasers to deliver average powers of several watts in blue-violet and UV band at pulsed repetition frequency (PRF) of the order of ten kHz,[3] which can be applied in microelectronics, micro-biology and genetic engineering, etc. These lasers were first obtained in experiment by Latush and Sem in 1973.[4] Later, they made a detailed investigation on mechanisms of this class of lasers and also concluded with five criteria on efficient operation of recombination lasers.[5] The primary two of them can be summarized as follows: 1) In most cases the levels of metal ions form more or less close groups and the gaps between such groups are considerably greater than the separations between the levels in each group. The upper laser level should be one of the lowest in the higher group of closely spaced levels, and the lower laser level should be one of the highest in the lower group of closely spaced levels. 2) The electron density should be sufficiently high

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to ensure that the probability of collisional transitions within groups exceeds the probability of optical transitions.

In order to increase the recombination rate, the electron temperature should be cooled as fast as possible after the end of discharge. Therefore, helium is regarded as an optimal buffer gas, and the falling edge of the current pulse is ensured to be short and free from second oscillations.

Though the self-terminating and the recombination lasers of calcium and strontium, univalent ions have been well investigated and developed in the past decades;[6−9] they were treated relatively independently. On the basis of our previous work and theoretical analysis on calcium and strontium atomic and ionic lasers,[10−12] we confirmed the univalent ions of calcium and strontium to be ideal lasants both for self-terminating and recombination lasers. Although the excitation mechanisms of the two classes of lasers are completely different, there exist similarities in the pulsed discharge circuit, the demand to the buffer gas and working temperature. In this paper, through improving our previous experimental setup, the laser cavity structure and the parameters of the discharge circuit, we combine the two classes of lasers together and successfully obtain the alternate oscillations of self-terminating laser and recombination laser of calcium and strontium univalent ions in experiment.

2. Analysis of energy level structures of Ca^+ and Sr^+

It is well known that both calcium and strontium belong to alkaline-earth metals and have similar physical and chemical properties. During the high voltage and high current pulsed discharge in a mixture of higher-pressure helium buffer gas and calcium or strontium vapours, above 90% Ca or Sr atoms are ionized to doubly charged ions via direct or stepwise ionizing due to their lower single and double ionization potentials. The energy level structures and the laser wavelengths of univalent calcium and strontium ions are shown in Fig.1 (a) and (b), respectively. As their energy level structures are similar, we focus on the Ca^+ level structure in this section. It is known from Fig.1 (a) that the upper group contains the levels from $5s\ ^2S_{1/2}$ to $5p\ ^2P^0_{1/2,3/2}$, while the lower group is formed by levels from $4s\ ^2S_{1/2}$ to $4p\ ^2P^0_{1/2,3/2}$. The level structure of the lower group in Ca^+ ions is similar to that of neutral atomic copper. The self-terminating transitions from the resonance levels to the metastable levels are

$4p\ ^2P^0_{3/2} \rightarrow 3d\ ^2D_{5/2}$ at 854.2nm wavelength, and
$4p\ ^2P^0_{1/2} \rightarrow 3d\ ^2D_{3/2}$ at 866.2nm wavelength.

The ratio of spontaneous emission intensity of the 854.2nm line to the 866.2nm is 9:5 according to the total sum rule,[13] but the laser action will be concentrated at the 854.2nm line because of the modulus competition.

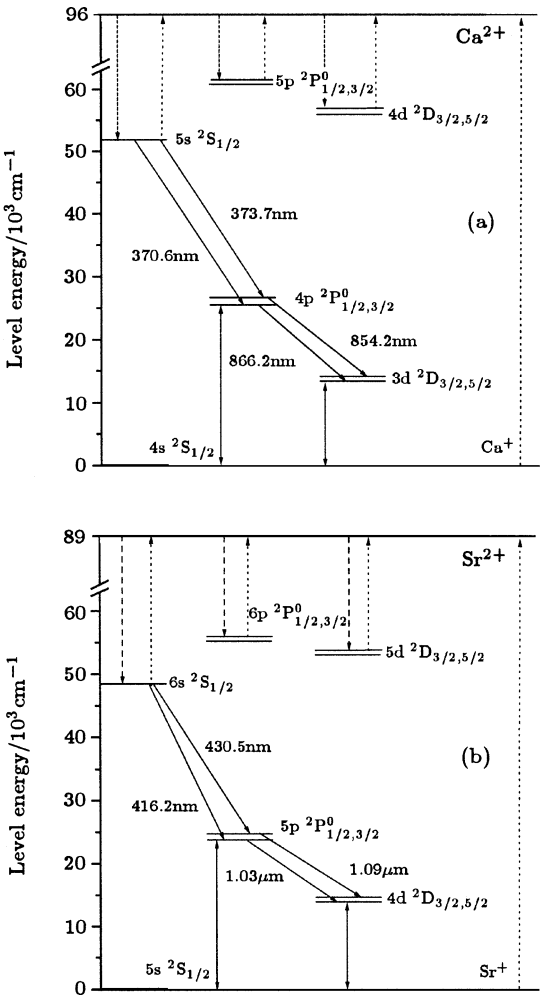


Fig.1. Schematic energy level diagram showing the laser transitions and the collisional processes in Ca^+ and Sr^+. – – → electron impact ionization, - - → electron/ion recombination, ⟷ electron/ion collision, ⟶ laser transition.

The upper laser level of the recombination laser is the lowest level $5s\ ^2S_{1/2}$ in the upper group, while the lower laser levels are the two of the highest levels $4p\ ^2P^0_{1/2,3/2}$ in the lower group. They fully meet the requirement to the energy level structures of recombination lasers. The recombination laser lines are

$5s^2S_{1/2} \rightarrow 4p\ ^2P^0_{3/2}$ at 373.7nm line, and
$5s^2S_{1/2} \rightarrow 4p\ ^2P^0_{1/2}$ at 370.6nm line.

The spontaneous emission intensity ratio evaluated by the sum rule of the two lines is 2:1, but the former will dominate in the recombination laser action because of the modulus competition.

Though both the self-terminating and the recombination lasers in univalent calcium ions share the same level $4p\,^2P^0_{1/2,3/2}$, their laser oscillations occur alternatively. The former appears at the rising edge of the current pulse while the latter at the early afterglow. Under condition of pulsed repetition discharge, calcium or strontium vapours by themselves are mixtures of their atoms and ions due to the accumulation effect, and the ratio of atoms to ions depends on the discharge conditions. Therefore, there are sufficient Ca^+ remainders on the ground state before each pulsed discharge so that the electron–ion collisions during the early period of discharge can be correctly described by the Born approximation. Thus, the first resonance levels of Ca^+ ($4p\,^2P^0_{1/2,3/2}$) will be preferentially excited over the metastable states $3d\,^2D_{3/2,5/2}$ via electron impact whenever the discharge is initiated; as a result, the population inversion between the two levels is established and the self-terminating lasing of Ca^+ is achieved. During the remainder of the pulsed discharge (~200 ns), stepwise pumping of the ground state Ca^+ and the excitation levels Ca^{+*} continues until a large population of the ground state Ca^{++} is established at the centre of the tube corresponding to about 98% of the original Ca density at the start of the current pulse.[14] During the period of recombination after the end of pulsed discharge, the decay rate of the concentration of multiply charged calcium ions N_i can be described by the following equation (which ignores the diffusion losses):

$$dN_i/dt = -\alpha N_i n_e^2. \qquad (1)$$

The triple recombination coefficient α depends on the ionic charge z and the electron temperature T_e,[5]

$$\alpha = 1.8 \times 10^{-8} z^3 L T_e^{-4.5} = c(z) T_e^{-4.5}, \qquad (2)$$

where L=$\ln(z^2+1)^{1/2}$ is the Coulomb logarithm, n_e is the electron density. Hence, the rate of recombination and, consequently, the rate of pumping of the levels increase strongly with decreasing T_e and increasing n_e and N_i, and also with increasing z.

With the ending of discharge, the electrons are mainly cooled by elastic collisions with He and He^+ species. Once the electron temperature drops to around 5000K, the Ca^{++} begins to recombine, pumping the high lying Ca^{+*} levels, and at this stage, the electron density remains sufficiently high and the gaps between the levels in a group are comparable to or smaller than kT_e; the probabilities of direct and reverse collisional transitions in a group exceed the probabilities of optical transitions within the group and to levels of lower groups, so that the populations of the levels in a group N_{ig} obey the Boltzmann distribution with an electron temperature T_e,

$$N_{ig} = N_0 g_i \exp(-\Delta\varepsilon_i/kT_e), \qquad (3)$$

where N_0 and g_i represent the total population density and the statistical weight of the ith level in each group, respectively. $\Delta\varepsilon_i$ is the energy spacing between the ith level and the lowest level in a group, and k is the Boltzmann constant. Thus, the population inversion is established between the lowest level $5s\,^2S_{1/2}$ in the upper group and the highest levels $4p\,^2P^0_{1/2,3/2}$ in the lower group, and the recombination laser is realized. The characteristics of the energy levels and the kinetic process of the two classes of lasers in univalent strontium ions are similar to those of single calcium ions.

From the above analysis, it is shown that there is practically no energy level competition between the self-terminating and the recombination lasers both for singly ionized calcium and strontium ions, despite the fact that they share a common resonance level (the lower laser level of the recombination laser serves as the upper laser level of the self-terminating laser). If all requirements of the two laser classes, particularly the steepness of rising edge and the falling edge of the current pulse, were met simultaneously, the two classes of lasers with different mechanisms in singly ionized calcium and strontium ions could occur alternatively in the same apparatus.

3. Experimental setup

The experimental setup shown in Fig.2 is composed mainly of a laser discharge tube, a pulsed discharge circuit and a compound laser cavity. The laser tube is made of a quartz tube with 16mm outer diameter, 12mm inner diameter and 35cm electrode separation. Five shallow caves were uniformly distributed between two electrodes on the bottom of the tube to hold 3–5 pieces of calcium or strontium of 99 percent purity. This design not only can realize discharge self-heating, but also can avoid impeding the light path. There is a condenser on each side of the Brewster

windows in order to efficiently cool the vapour of calcium or strontium from the discharge zone and prevent deposition on the windows. A cylindrical metal heater separated 3cm from the outside of the discharge tube is coaxially placed along the laser tube to control the axial temperature of the laser tube. It has been confirmed that the whole working zone is uniformly heated in this apparatus and the axial temperature difference does not exceed 10°C. A Blumlein circuit was employed as a pulsed discharge circuit to ensure the current pulse can be applied both for self-terminating and recombination lasers. L_c is a charging inductance; $D_{1,2}$ is the 2DGL-2 A/20kV high voltage silicon diode for the purpose of charging capacitances C_1 and C_2 to doubled high voltage and disconnecting the discharge. The circuit is defined as an asymmetric Blumlein circuit with different storage capacitors C_1 and C_2. The inductance L_t ($\sim$1 μH) has the function of protecting the hydrogen-filled thyratron (ZQM-2000/25) and also has certain effect on diminishing the current oscillation. Because of the lack of a total reflector with a wide band range from near ultraviolet to near infrared, the optical cavity is composed of three external dielectric-coated mirrors M_1 to M_3. Considering that the gain coefficient of the self-terminating laser is larger than that of the recombination laser, the output coupler M_1 is of higher reflectivity at recombination laser lines and of lower reflectivity at self-terminating laser lines, the mirror M_2 with 3m radius of curvature serves as a total reflector to the recombination laser lines and has 90% transmissivity to the self-terminating lasers of Ca^+ and Sr^+, respectively; the mirror M_3 is a flat total reflector to the self-terminating lasers. Thus, M_1 and M_2 form the stable cavity for the recombination lasers, while M_1 and M_3 are for the self-terminating lasers if ignoring the loss of M_2. Table 1 lists the reflective coefficients of mirrors M_1–M_3 for the corresponding laser lines of Ca and Sr ionic lasers. Though this compound cavity is preferential to the recombination laser action, the self-terminating lasing is also obtained in our experiment. A Max 500 laser power meter with a set of optical filters measures the output power of the self-terminating and the recombination lasers. The waveforms of the discharge current and the laser pulses are detected by a Pearson model 410 current transformer and a plane photoelectric diode with fast response time (less than 2 ns), respectively. All pulses are displayed on a Tektronix TDS 754C oscilloscope.

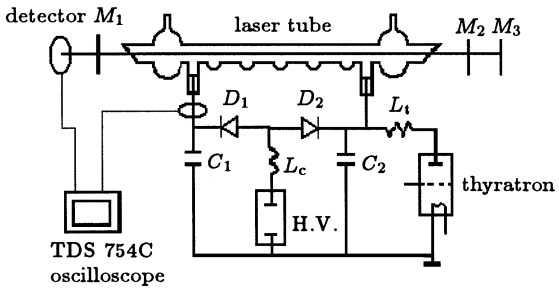


Fig.2. Schematic diagram of the experimental arrangement.

Table 1. The reflectivity of cavity mirrors at corresponding laser lines.

	Ca^+ laser cavity				Sr^+ laser cavity		
	M_1	M_2	M_3		M_1	M_2	M_3
Laser line/nm	Reflectivity/%			Laser line/nm	Reflectivity/%		
370.6/373.7	85	99.8	–	416.2/430.5	70	99.8	–
854.2/866.2	10	10	99	1033/1091	10	10	99

4. Optimization of the excitation circuit

Based on our previous work on various pulsed discharge circuits used for metal vapour lasers,[15] we found that the Blumlein circuit shown in Fig.2 can simultaneously satisfy the requirements for both self-terminating and recombination lasers. For obtaining a set of optimal combination of parameters of the discharge circuit, we set up a computer model to simulate the electric parameters of Blumlein circuit. The equivalent discharge circuit is shown in Fig.3. Thus, a complete description of the state of the discharge circuit is made by solving four coupled first-order ordinary differential equations:

$$\mathrm{d}V_1/\mathrm{d}t = -I_1/C_1, \tag{4}$$

$$\mathrm{d}V_2/\mathrm{d}t = -(I_2 - I_1)/C_2, \tag{5}$$

$$\mathrm{d}I_1/\mathrm{d}t = (V_1 - V_2 - [R_\mathrm{p} I_1])/L_1, \tag{6}$$

$$\mathrm{d}I_2/\mathrm{d}t = (V_2 - [R_\mathrm{th} I_2])/L_2, \tag{7}$$

with

$$R_\mathrm{th} = 10^6 \exp(-t/\tau_\mathrm{th}) + R_\mathrm{th0}, \tag{8}$$

and

$$R_\mathrm{p} = 10^3 \exp(-t/\tau_\mathrm{p}) + R_\mathrm{p0}, \tag{9}$$

where R_th and R_p represent the thyratron resistance with τ_th=5 ns and the plasma resistance with τ_p=25

ns, respectively. V_1 and V_2 are the charging voltage of the storage capacitors C_1 and C_2. L_1 and L_2 are lumped loop inductances. R_{th0} =5 Ω and R_{p0} =35 Ω represent the stable resistances of the thyratron and the plasma during discharge period. Under the condition of V_1 = V_2 =11kV and C_1=1 nF, the simulated discharge current waveforms varying with the capacitor C_2 are shown in Fig.4. It can be seen that the value of C_2 has certain effect on the fall edge of the discharge current. When the C_2 capacitance is in the range from 1.5nF to 2nF, the discharge current both with the sharp rise edge and fall edge can simultaneously satisfy the requirement of self-terminating laser and recombination laser. Through comparison of the simulated current pulse with the experimental one (the I pulse in Fig.5) in the same capacitances of C_1=1nF and C_2=2nF, one can see that the simulated result is in good agreement with the experiment.

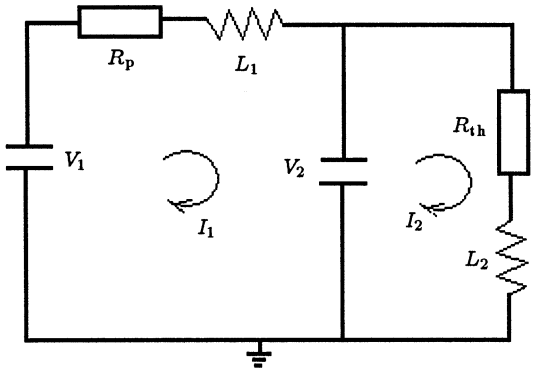


Fig.3. Equivalent discharge circuit of Blumlein circuit.

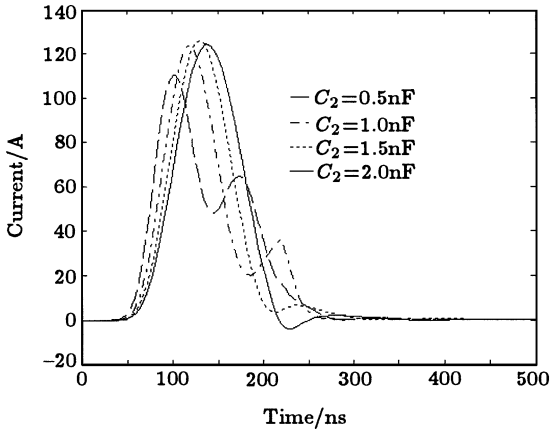


Fig.4. Laser tube current pulses varying with the C_2 capacitance under condition of V_1 = V_2=11kV and C_1=1nF.

5. Results and discussion

It is well known that both the self-terminating laser and the recombination laser excited by a longitudinal pulsed discharge require a buffer gas to convey the electric energy and to maintain stable discharge. Different kinds of buffer gases have great influence on the laser performance. Generally, all inert gases, especially neon and helium, can be used as buffer gas for the self-terminating laser, while for the recombination laser, only helium is confirmed to be an optimal buffer gas in order to cool the electron temperature as fast as possible during the recombination period. Therefore, helium was employed as a buffer gas for obtaining alternative lasing of the two different mechanisms in Ca and Sr ionic lasers by pulsed discharge. Based on the above theoretical analysis in Section 2 and the parametric optimization of the asymmetric Blumlein circuit with C_1=1nF and C_2=2nF, we made a further experimental investigation on a calcium ionic recombination laser and observed a weak optical pulse appearing at the rise edge of the current pulse as well as an intensive recombination laser pulse at the end of current pulse in detecting the photoelectric pulse waveforms. The weaker optical signal was identified to be the two near-infrared self-terminating laser lines of singly ionized calcium at 854.2nm and 866.2nm by an infrared grating interferometer. Furthermore, with the improvement of the optical cavity described in Section 3, the weaker signal becomes much more intense and was confirmed as a laser pulse; meanwhile the intensity of the recombination laser pulse remains stable under the same set of operation conditions. Thus, the alternate laser oscillations of the above two different mechanisms in single ions of calcium were successfully realized. Subsequently, we carried out another experiment on strontium ionic laser and obtained a similar result to that of the calcium ionic laser. The typical photoelectric pulse waveforms of alternate oscillations of self-terminating laser and recombination laser in single ions of calcium or strontium are shown in Fig.5. In the figure, I represents the current pulse of laser tube, L_s and L_r are laser pulses of self-terminating and recombination lasers, respectively. In doing the measurement, we carefully corrected the delay time between the electric signal and the optical signal in the detecting system, and simultaneously displayed them on a Tektronix TDS 754C oscilloscope. It is clear that the self-terminating laser pulse L_s appears at the rise edge of the current pulse, while the recombination laser pulse L_r occurs during the plasma recombination period after the end of discharge. The duration of L_s (∼50ns) is much shorter than that of L_r (∼450ns), the delay time between them is about 220 ns. So there is enough time for the resonant level (the common level

of the two class lasers, 4p $^2P^0_{1/2,3/2}$ for Ca^+ and 5p $^2P^0_{1/2,3/2}$ for Sr^+) to be depopulated by rapid spontaneous emission and electron collision. Thus the formation of the population inversion between the upper and the lower levels of recombination laser will not be influenced by the self-terminating laser action.

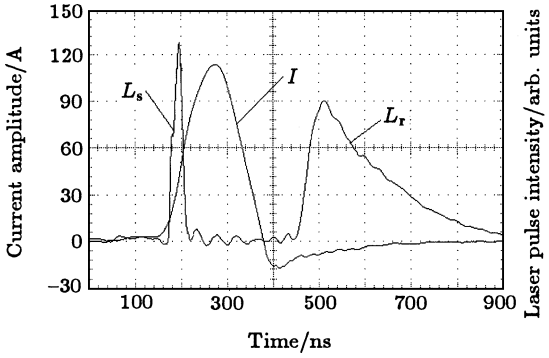


Fig.5. Typical photoelectric waveforms of alternate laser oscillations with different mechanisms in calcium ionic laser.

The dependence of output power on the working parameters was initially studied after obtaining the alternate oscillations of the two classes of lasers in Ca^+ and Sr^+. Table 2 lists the maximum total power (P_t), individual output power (P_r for recombination laser and P_s for self-terminating laser) and corresponding operation parameters. Because the compound optical cavity and the higher helium pressure are favorable to the recombination laser action, the output power of the recombination laser is about two times that of the self-terminating laser. If the working conditions, especially the optical cavity and the discharge circuit, were further improved, we can predict that the total laser output power will be increased to several hundreds of milliwatts or even to an order of a watt, and the desirable ratio of the P_r to P_s will be attained with varying operation parameters. Thus, this class of laser can provide an opportunity to obtain the discrete tunable laser with multiple lines in a wider spectral band.

Table 2. The optimal operation parameters and corresponding average output power of calcium and strontium ionic lasers.

Lasants	Helium pressure P_{He}/kPa	Charging voltage U_c/kV	PRF/kHz	P_t/mW	P_r/mW	P_s/mW
Single Ca ion	18.5	10.2	5.5	128	82	40
Single Sr ion	20.5	10.0	5.2	320	205	115

6. Conclusion

Based on the theoretical analysis of their laser mechanisms and energy level structures, the univalent calcium and strontium ions are proved to be ideal lasants both for self-terminating laser and recombination laser. The alternate oscillations of their self-terminating and recombination lasers were successfully obtained in experiment with longitudinal pulsed discharge by improving the optical cavity and the excitation circuit. It is shown that this class of laser can provide a new way to realize the discrete tunable laser in a wider spectral range from ultraviolet to infrared band and has practical applications in many fields such as microelectronics, genetic engineering and infrared technology.

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