

Investigation of Hollow Cathode Ge II and Te II Lasers

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Abstract. Eighteen laser transitions were observed in Te and Ge hollow cathode discharges, four of which (6294, 5649, and 5488 Å of Te II and 5893 of Ge II) are reported for the first time. Charge exchange, Penning ionization, and radiative cascade processes are suggested as being responsible for the population of the upper laser levels.

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Laser action in Te II was first described by Bell et al. in 1966 [1]. In a pulsed ring discharge with Ne as buffer gas, the 5576, 5708, and 6350 Å lines were observed. In 1968 Webb reported other laser transitions (5454, 5640, 5936, 6246, and 7039 Å) in a pulsed discharge with Ne as buffer gas, two of which (6246 and 7039 Å) were also observed with He as buffer gas [2]. Using a 2 m long longitudinal discharge tube and external heating to produce the necessary Te vapor pressure, Silfast and Klein observed 31 laser transitions with both He and Ne as buffer gas [3].

Laser action in Ge II was first reported by Silfast et al. in 1966 [4]. The 5178 and 5131 Å transitions were observed in a pulsed discharge in germanium vapor with He or Ne as buffer gas. In 1974 Green and Webb predicted nine potential laser transitions [5]. Their considerations were based on measurements of relative cross-sections for the excitation of various Ge II levels by charge exchange processes in the Ne afterglow.

Here we report the observation of 15 laser transitions in Te and 3 in Ge hollow cathode discharges, of these three Te II laser lines (6294, 5649, and 5488 Å) and one Ge II laser line (5893 Å) are believed to be reported for the first time. Charge exchange, Penning ionization and radiative cascade processes are suggested as being responsible for the population of the upper laser levels.

1. Experimental Set-Up and Results

The experimental set-up was the same as that described in a previous paper [6]. The necessary Te and Ge atom densities were produced in pulsed hollow cathode discharges via sputtering of the cathode material by buffer gas ions. The slot of the hollow cathode was 50 cm long and had a cross-section of $2 \times 6 \text{ mm}^2$ (width \times depth). The power supply provided current pulses of up to 50 A peak and with durations between 20 μs and several ms. The discharge current density was several orders of magnitude larger than that from simple positive column discharges between plane electrodes at the same pressure [7]. As a consequence, higher densities of metastable atoms and ground-state ions of the buffer gas were obtained. This was favorable for populating the upper laser level via the charge exchange process [8]



and Penning ionization [8]



where B , B^* , and B^+ represent the ground state, metastable state and the ground state of the ion of the buffer gas, respectively, and A , $(A^+)^*$ the ground state and an excited state of ionic Te and Ge, respectively. Of the fifteen laser transitions of Te II observed, three (5020, 4843, and 5708 Å) were excited in a mixture of

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Table 1. The laser transitions observed in hollow cathode Te II and Ge II lasers. The tabulated wavelengths and transition assignments of Te II and Ge II were given by [9] and [14] respectively

Wavelength [Å]		Transition assignment		Species	Buffer gas	Threshold current ^b [A]
Measured	Tabulated	Upper level	Lower level			
5708	5708.120	103 _{7/2} ⁰	85 _{5/2} ¹	Te II	He + Ar	6
5020	5020.39	^c	^c	Te II	He + Ar	30
4843	4842.895	122 _{5/2} ⁰	102 _{3/2}	Te II	He + Ar	36
9377	9378.5 ^d	99 _{3/2} ⁰	88 _{5/2}	Te II	Ne	18
7039	7039.13	97 _{1/2} ⁰	83 _{1/2}	Te II	Ne	12
6649	6648.58	97 _{1/2} ⁰	82 _{3/2}	Te II	Ne	21
6294 ^a	6293.619	97 _{1/2} ⁰	81 _{3/2}	Te II	Ne	21
6246	6245.450	99 _{3/2} ⁰	83 _{1/2}	Te II	Ne	21
6230	6230.728	105 _{3/2} ⁰	88 _{3/2}	Te II	Ne	25
5936	5936.146	99 _{3/2} ⁰	82 _{3/2}	Te II	Ne	15
5756	5755.852	100 _{5/2} ⁰	82 _{3/2}	Te II	Ne	4
5649 ^a	5649.259	96 _{3/2} ⁰	78 _{1/2}	Te II	Ne	15
5488 ^a	5487.951	100 _{5/2} ⁰	81 _{3/2}	Te II	Ne	35
5479	5479.080	105 _{3/2} ⁰	86 _{3/2}	Te II	Ne	18
5450	5449.838	103 _{3/2} ⁰	85 _{5/2}	Te II	Ne	21
5131	5131.7517	4f ² F _{3/2} ⁰	4d ² D _{3/2}	Ge II	He + Ar	15
5178	5178.6475	4f ² F _{7/2} ⁰	4d ² D _{5/2}	Ge II	He + Ar	10
5893 ^a	5893.3886	5p ² P _{3/2} ⁰	5s ² S _{1/2}	Ge II	He + Ar	10

^a New laser transition

^b Measured in pulsed mode with a pulse width of 0.1 ms

^c Transition assignment unknown [9]

^d Wavelength and transition assignment given by [3]

He (12 mbar) and a small amount of Ar (0.5 mbar), the other twelve lines were excited in pure Ne (6 mbar) as buffer gas (Table 1). None of the fifteen laser transitions could be excited either with He or with Ne as buffer gas. Figure 1 shows the relevant energy levels of Te II, the Te II laser transitions being marked by solid lines. The assignment of the Te II spectrum is still incomplete, most of the excited states have only been classified in terms of their J values and energies; the laser transition at 5020 Å has not been classified at all [9] and is therefore missing in Fig. 1.

It is noteworthy that all the observed laser transitions of Te II except for 5020 and 5708 Å originate from levels the energies of which are close to the ground states of the buffer gas ions (Ne⁺ or He⁺). For the 5020 Å transition the assignment is unknown and the 5708 Å transition originates from a level lying far below the He⁺ ground state. The three new laser transitions originate from levels having odd parity like the other eleven laser transitions [3].

The emission spectra of the Te hollow cathode were measured with both He and Ne as buffer gas. In the case of Ne as buffer gas, the intensities of transitions which originate from levels lying near the ground state of Ne⁺ were enhanced; in the case where He is used as buffer gas the transitions starting from levels near the He⁺ ground state were stronger. The current de-

pendences of the spontaneous intensities of the laser transitions were measured, all showing a linear dependence.

All laser lines of Ge II were observed with mixtures of He (12 mbar) and Ar as buffer gas. The optimum partial pressure of Ar was 0.5 mbar for the laser lines at 5178 and 5131 Å, and 0.1 mbar for the 5893 Å line. The behaviours of the spontaneous and stimulated emission at 5178, 5131, and 5893 Å were similar to each other. That is: (A) With increasing current density the intensities of the spontaneous emission at 5178, 5131, and 5893 Å increased at first in a nearly linear manner, then at about 0.4 A/cm² the slope changed and the increase was slower than before. (B) The time behaviors of both the laser output power and the spontaneous emission intensity at 5178, 5131, and 5893 Å showed a peak 10 μs after termination of the discharge current. (It is noteworthy that the behaviour of another Ge II line at 4814 Å and of the He I line at 4471 Å was very similar to that of the 5178, 5131, and 5893 Å lines of Ge II.)

The strong lines of the spontaneous emission spectrum of the germanium hollow cathode are shown in a level scheme (Fig. 2). They mainly originated from the levels lying near the He metastable states and none of them originated from the levels lying near the He⁺ ground state.

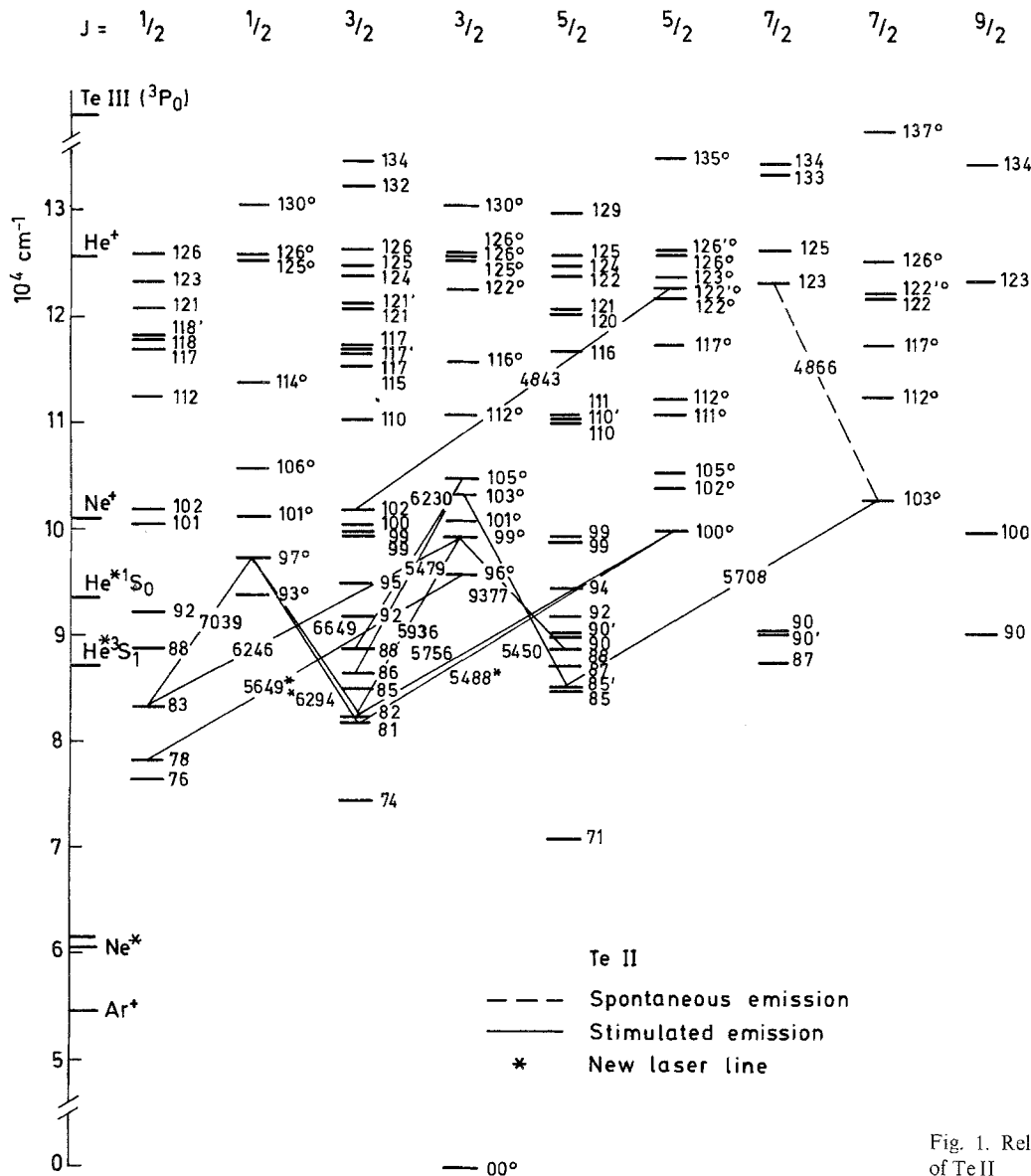


Fig. 1. Relevant energy level diagram of Te II

2. Discussion

We suggest that the charge exchange process (1) is responsible for the population of the upper laser levels of all the twelve Te II laser transitions with Ne as buffer gas and the laser transition of Te II at 4843 Å with He as buffer gas. The reasons are as follows: (A) Energy defects of these thirteen laser transitions are $|\Delta E| \leq 0.63 \text{ eV}$; generally charge exchange processes are very effective when the energy defect is of the order of 0.1–2 eV [8]. (B) The enhancement of the intensities of the spontaneous transitions which originate from the energy levels lying near the ground state of Ne⁺ or He⁺ implies that charge exchange processes are important in the Te hollow cathode discharge. (C) The linear current dependences of the intensities of the

transitions which are responsible for population inversion are also evidence of the existence of the charge exchange process. A similar phenomenon in a Cu hollow cathode laser has been explained by Hoog et al. [15], i.e. the buffer gas ions B⁺ are created by both electron impact ionization and cumulative ionization, these two processes depending linearly on the current. Since the major loss process of B⁺ is the charge exchange process and the copper density also varies linearly as the current, the steady-state buffer gas ion density should be independent of the discharge current and, according to reaction (1), the spontaneous emission is expected to show linear behaviour. (D) None of the fifteen laser transitions could be excited when either He or Ne was used. This also supports the suggestion that the charge exchange process is responsible for the pump-

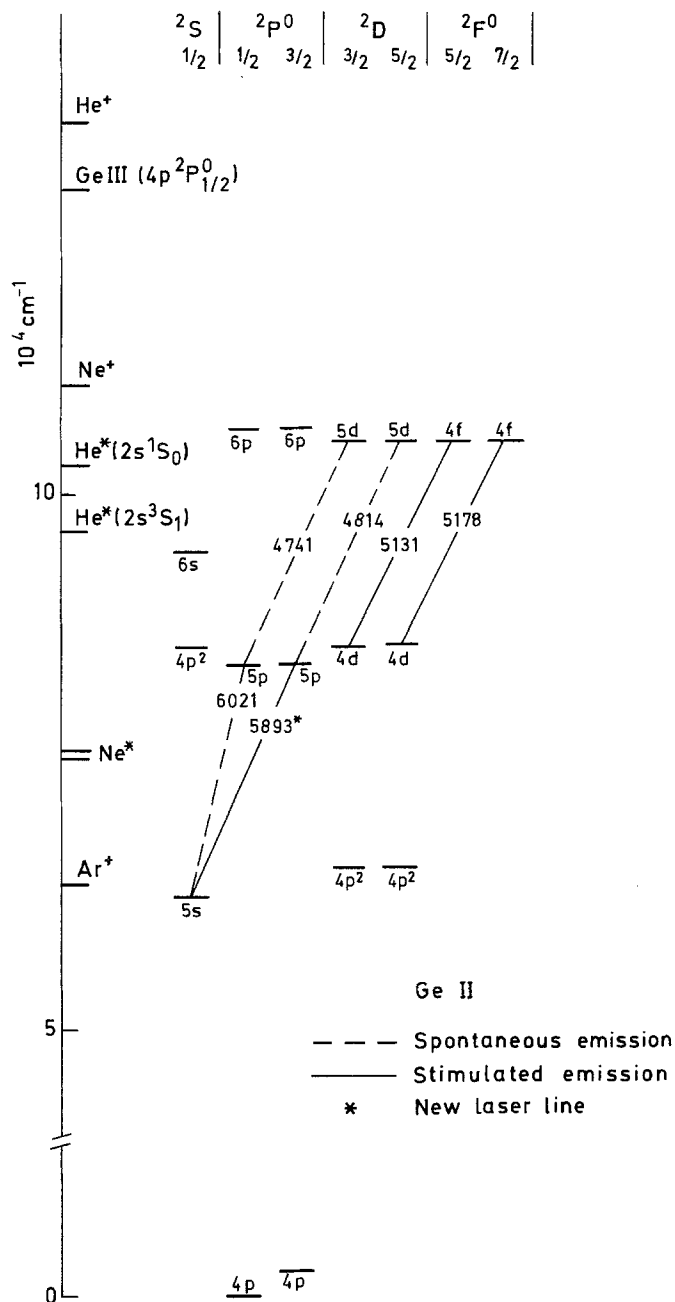


Fig. 2. Relevant energy level diagram of Ge II

ing process. Penning ionization does not appear to have any influence on the population of the upper laser levels of Te II because the energy levels of the metastable states of He* and Ne* are far below the upper laser levels (Fig. 1).

As for the Te II laser transition at 5708 Å with He as buffer gas, the upper laser level $103_{7/2}^0$ is 2.8 eV lower than the He⁺ ground-state level, so it cannot be populated directly by a charge exchange process. It appears that the population process of the laser transition at 5708 Å is stepwise, i.e. a direct charge ex-

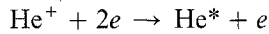
change process followed by a radiative cascade process. The charge exchange process directly populates the $123_{7/2}$ level, and then the radiative cascade process from $123_{7/2}$ to $103_{7/2}^0$ populates the upper laser level $103_{7/2}^0$ (Fig. 1). The evidence is as follows: (A) The transition $123_{7/2} - 103_{7/2}^0$ at 4866 Å is the strongest one in the Te II spontaneous emission spectrum with He as buffer gas, but it is very weak with Ne as buffer gas. (B) For the energy level $123_{7/2}$ the energy defect is 0.25 eV. (C) A linear current dependence of the intensity of the transition at 4866 Å was observed.

All Te II upper laser levels have odd parity, this phenomenon having been explained by Silfast and Klein [3]. The spontaneous spectra we observed shows that the charge exchange process pumps not only the levels having odd parity but also the levels having even parity. For example, the transition $123_{7/2} - 103_{7/2}^0$ (4866 Å) is also obviously enhanced by the charge exchange process. Therefore this phenomenon is not due to the charge exchange process. It seems to be caused by the fact that fast radiative decay to the Te⁺ ground state levels is only available to those lower laser levels having even parity since the Te⁺ ground states which originate from the configuration $5s^2 5P^3$ are all of odd parity.

As for the Ge II laser transitions, we suggest that the population processes are as follows: (A) For the laser transitions at 5178 and 5131 Å, the upper laser levels $4f^2 F_{7/2}^0$ and $4f^2 F_{5/2}^0$ are populated directly by the Penning ionization process (2). (B) For the laser transition at 5893 Å the population process is that Penning ionization directly populates the energy level $5d^2 D_{5/2}$, of which the upper laser level $5p^2 P_{3/2}^0$ is populated by the radiative cascade process $5d^2 D_{5/2} - 5p^2 P_{3/2}^0$ (4814 Å).

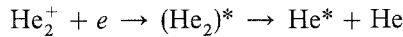
The reasons for the $4f^2 F_{7/2}^0$, $4f^2 F_{5/2}^0$, and $5d^2 D_{5/2}$ levels being populated directly by the Penning ionization process are as follows: (A) The energy defects between the $2s^1 S_0$ level of He I and $4f^2 F_{7/2}^0$, $4f^2 F_{5/2}^0$, and $5d^2 D_{5/2}$ of Ge II are about 0.3 eV, the Penning ionization process usually being possible in the case of energy defects of up to at least 2 eV [11]. (B) Stronger spontaneous emission originating from the levels lying in the region 2 eV below the $2s^1 S_0$ state of He* was observed (Fig. 2). (C) As mentioned above, the slopes of the spontaneous emission intensities at 5178, 5131, 5893, and 4814 Å changed at about 0.4 A/cm². This can be explained by the metastable helium density saturation beginning at a current density of 0.4 A/cm² [7]. (D) It is noteworthy that the Wigner spin rule is obeyed by Penning ionization processes between the $2s^1 S_0$ state of He I and the $4f^2 F_{5/2}^0$ and $5d^2 D_{5/2}$ states of Ge II. (E) As mentioned above, the time behaviors of the spontaneous emission intensities at 5178, 5131, 5893, and 4814 Å of Ge II were very similar to that of

the He I 4471 Å line. They all showed a peak about 10 μs after termination of the discharge current. The peak in the afterglow of a He discharge was observed and explained by Fugol et al. [12] and Pakhomov [13]. After termination of the discharge current the collisional-radiative recombination process



He*: the excited He atom

and the dissociative-recombination reaction



(He₂)*: the unstable excited helium molecule

cause a sharp increase in intensity of some He lines which originate from high-lying states $n=3, 4, 5,$ and $6,$ including the 4471 Å line ($4^3D - 2^3P$). The similarity in time behavior between the Ge II lines (5178, 5131, 5893, and 4814 Å) and the He I line (4471 Å) implies that helium atoms were responsible for the pumping processes of the 5178, 5131, 5893, and 4814 Å transitions of Ge II. This supports our suggestion that the $4f^2F_{7/2}^0,$ $4f^2F_{5/2}^0,$ and $5d^2D_{5/2}$ levels were populated by the Penning ionization process.

It appears that only one metastable state ($2s^1S_0$) out of two metastable states ($2s^1S_0$ and $2s^3S_1$) of the atom contributed to the population processes via Penning ionization although usually the density of $2s^3S_1$ is higher than that of $2s^1S_0$. The reasons are as follows: (A) The $4f^2F_{7/2}^0,$ $4f^2F_{5/2}^0,$ and $5d^2D_{5/2}$ levels lie lower than the $2s^1S_0$ level but higher than the $2s^3S_1$ level (Fig. 2) and one of the requirements for the presence of the Penning ionization process is a positive energy defect [8, 11]. (B) No strong or medium intensity spontaneous emission originating from the levels 0–2 eV below the $2s^3S_1$ level was observed. (C) It is noteworthy that the Penning ionization processes between the $2s^3S_1$ state, of He I and the $4f^2F_{7/2}^0,$ $4f^2F_{5/2}^0,$ and $5d^2D_{5/2}$ states of Ge II violate the Wigner spin rule.

Although the behavior of the 5893 Å line was similar to that of the 5178 and 5131 Å lines in many respects, the optimum Ar partial pressure of the 5893 Å line

(0.1 mbar) was different from that of the 5178 and 5131 Å lines (0.5 mbar). A possible reason is that the Ar can increase the sputtering yield and in turn increase the population of the upper laser level. This is true for all laser lines observed. But for 5893 Å, because the lower level $5s^2S_{1/2}$ happens to lie 0.14 eV lower than the ground state of Ar⁺, the Ar can also increase the population of the lower laser level, thus decreasing the population inversion. This causes the different optimum Ar partial pressure between the two laser lines (5178 and 5131 Å) and the 5893 Å line.

In summary, we have experimentally verified that laser action in Te and Ge hollow cathode discharges can be achieved via charge exchange, Penning ionization and radiative cascade processes. Four new laser lines (6294, 5649, and 5488 Å of Te II and 5893 Å of Ge II) were observed.

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