Laser Chem.
Volume 1(5), May 1983, pp. 195–209
0278-6273/83/0105-0195$12.00/0

Printed in Great Britain

New Continuous Laser Emissions in Te_2

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(Received October 15, 1982; in final form December 3, 1982)

New continuous laser emissions of different Te_2 isotopes on many lines in the spectral range of 4600 Å to 7800 Å have been obtained upon excitation with the 4067 Å and 4131 Å lines of a krypton ion laser. Thresholds of several mW and output efficiencies of 5% have been achieved. By means of Fourier transform spectrometry accurate fluorescence wavenumbers have been determined, leading to an exact assignment of the observed laser lines.

INTRODUCTION

Optically pumped dimer laser investigations performed for some time have succeeded in many new systems covering extended spectral regions.[1] There is presently some interest in the development of new systems that can be excited with available pulsed or cw UV lasers and have emissions in the blue-green or even throughout the whole visible spectral range. Especially S_2 dimers have gained some attention as a laser system with possible high conversion efficiencies and output powers when being excited with powerful excimer lasers.[2] This system also offers some chances for discharge assisted or eventually even pure discharge pumped operation.[3] However, as a pure optically pumped system, S_2 has some disadvantages as high temperatures are necessary to break the normally present polymers into

dimers.[1] Further interesting candidates for the considered spectral range are the other group VI dimers as Se_2 and Te_2 which require much lower temperatures. Continuous laser oscillation of Se_2 excited at 3511 Å with an argon ion laser and with laser emission in the range of 3870 Å to 7090 Å has been reported recently.[4] Laser operation of Te_2 excited with some blue argon laser lines and emission in the yellow-red spectral range has been obtained earlier.[5] In this case excitation starts from higher vibrational levels. In order to obtain low operating temperatures and the broadest possible laser emission range, excitation should start from the lowest vibrational level. In the case of Te_2 this can be achieved by using the violet krypton laser lines at 4067 Å and 4131 Å.[6]

In this paper we report on first continuous laser oscillation of $^{130}Te_2$ and several other Te_2 isotopes upon excitation with these krypton laser lines. By means of Fourier transform spectrometry the involved pump and laser transitions have been identified and accurate transition wavenumbers of the fluorescence lines corresponding to the observed laser lines are given.

SPECTROSCOPIC DATA

Fluorescence spectra of $^{130}Te_2$ and $^{128}Te_2$ isotopic molecules, excited by fixed-frequency argon and krypton ion lasers, have been recorded by high resolution Fourier transform spectrometry and their analysis has been reported recently.[6] In this paper the collisional aspect has only been treated from a spectroscopic point of view. These detailed studies have been especially helpful for the optically pumped laser investigations described here.

Figure 1 gives the potential curves of the X, B and b states. The equilibrium internuclear distance r_e = 2.824 Å of the B state is markedly greater than those of the X and b states (2.5574 Å and 2.5862 Å, respectively). This circumstance is particularly favorable to observe many transitions in the fluorescence series, spanning broad spectral ranges. B–X transitions thereby yield emissions throughout the visible spectral region, while the existence of the b state, 9600 cm^{-1} above the $X\ 0_g^+$ state, leads to intense fluorescence series in the infrared between 6000 cm^{-1} and 15000 cm^{-1}.[7] The equilibrium molecular constants of $^{130}Te_2$ have been determined with great accuracy[6] and these data also allow the calculation of the electronic transitions of the combination of isotopes in natural or enriched

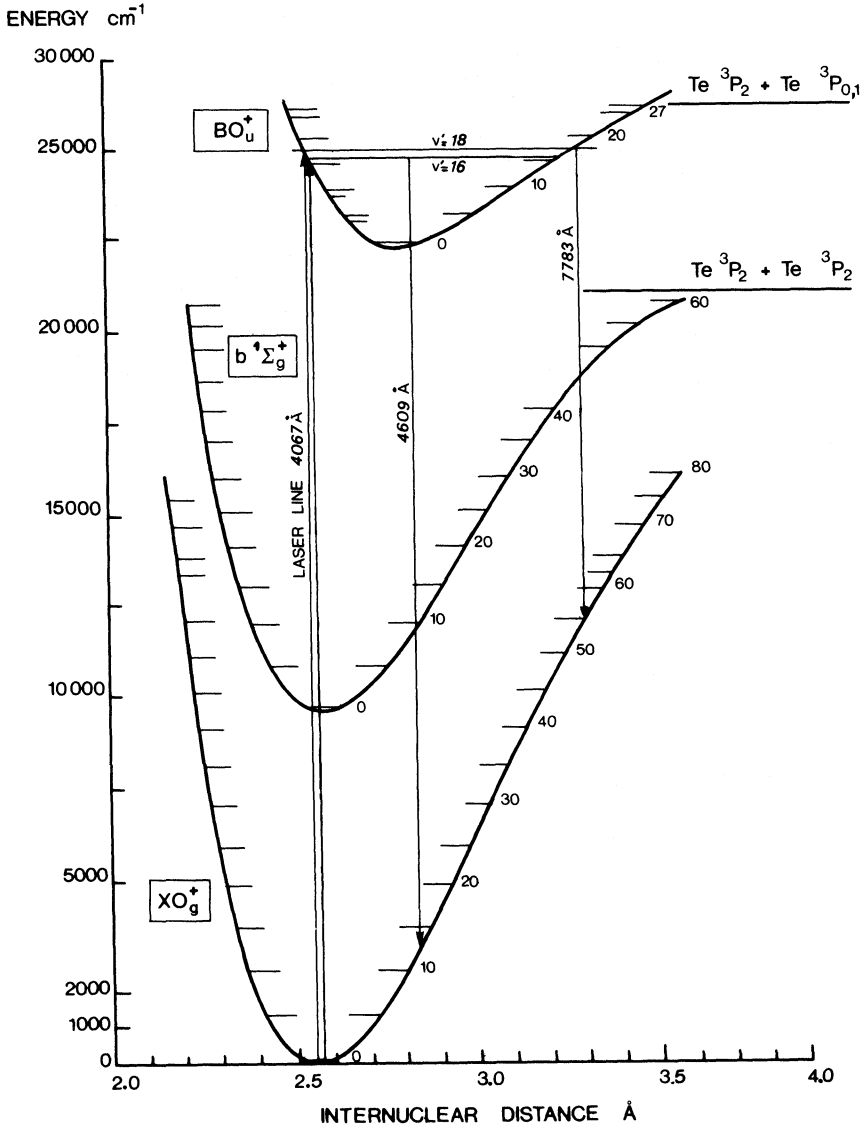

FIGURE 1 Energy level diagram of Te_2 (after Ref. 6) with pump and observed laser transitions for $^{130}Te_2$.

tellurium with an uncertainty of a few mk (0.001 cm^{-1}) and thereby the prediction of further pump and laser lines.

Some of the numerous fluorescence series studied originate in excited levels which are pumped from the most populated $v'' = 0$ level of the fundamental state $X\ 0_g^+$. These series consist mainly of P, R doublets with spacings ranging between 6 cm^{-1} and 20 cm^{-1}. The strongest of them are surrounded by satellites corresponding to $\Delta J = \pm 2, \pm 4 \ldots$, which are populated by rotational relaxation. Excitation and fluorescence transitions for different Te_2 isotopes, analyzed by Fourier transform spectrometry upon excitation with the 4067 Å and 4131 Å lines of the krypton laser, are summarized in Table I. Low resolution fluorescence spectra of $^{130}Te_2$ and natural Te_2 are given in Figures 2 and 3.

LASER INVESTIGATIONS

For the optically pumped laser experiments a small all-quartz cell described elsewhere[4] was filled with natural or isotopically pure tellurium and heated to temperatures around 700°C and 570°C for cell and reservoir respectively. A low-loss linear concentric resonator with mirrors of equal radii (260

TABLE I

Excitation and fluorescence transitions observed for different Te_2 isotopes and analyzed by Fourier transform spectrometry

$^{130}Te_2$	Excitation			Fluorescence	Intensity[a]
Kr^{2+} 4067 Å	$B\ 0_u^+ - X\ 0_g^+$	16-0	R(36)	$B\ 0_u^+ - X\ 0_g^+$	s*
				$B\ 0_u^+ - X\ 1_g^+$	w
				$B\ 0_u^+ - b\ ^1\Sigma_g^+$	s
		18-0	R(172)	$B\ 0_u^+ - X\ 0_g^+$	s*
				$B\ 0_u^+ - X\ 1_g^+$	w
				$B\ 0_u^+ - b\ ^1\Sigma_g^+$	s
$^{128}Te_2$					
Kr^{2+} 4131 Å	$B\ 0_u^+ - X\ 0_g^+$	14-0	R(116)	$B\ 0_u^+ - X\ 0_g^+$	w
				$B\ 0_u^+ - b\ ^1\Sigma_g^+$	w

[a]w: weak, s: strong, with respect to total emission.
*Fluorescence transition showing laser effect.

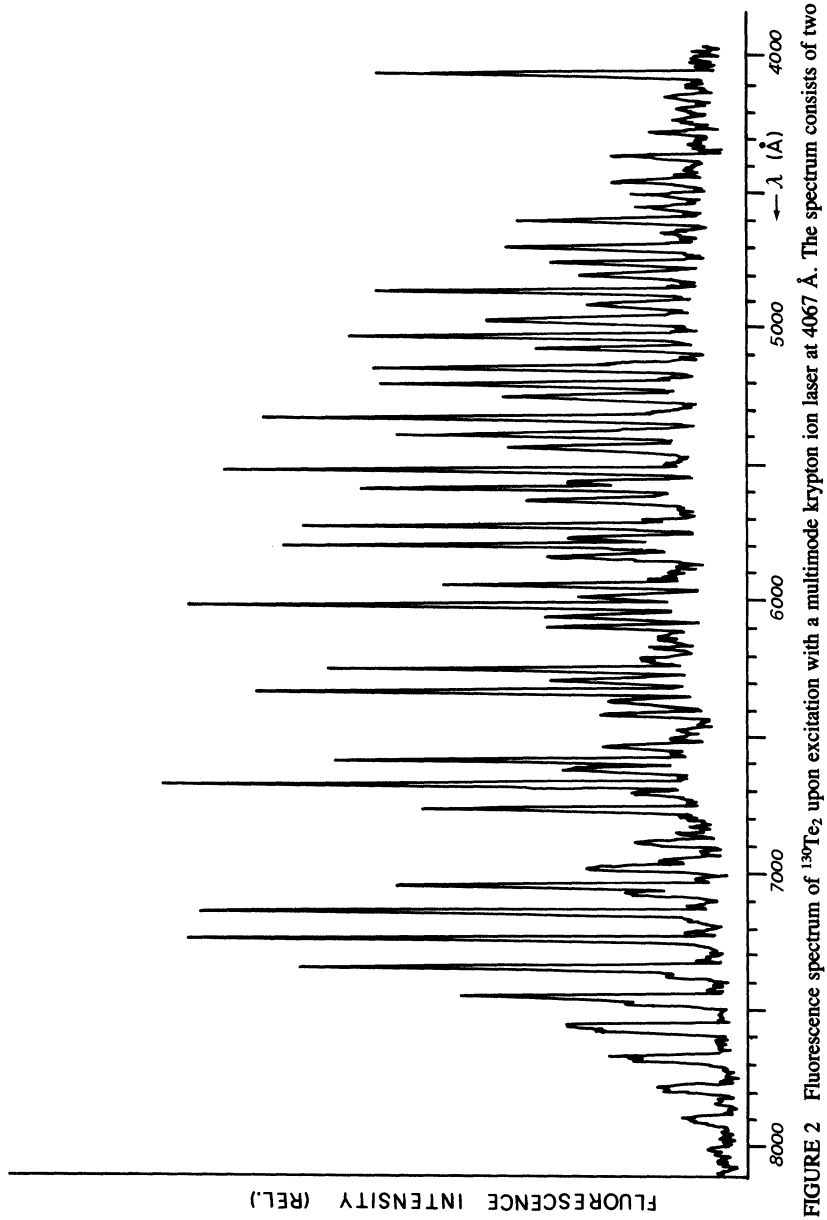

FIGURE 2 Fluorescence spectrum of $^{130}Te_2$ upon excitation with a multimode krypton ion laser at 4067 Å. The spectrum consists of two progressions (see Table II).

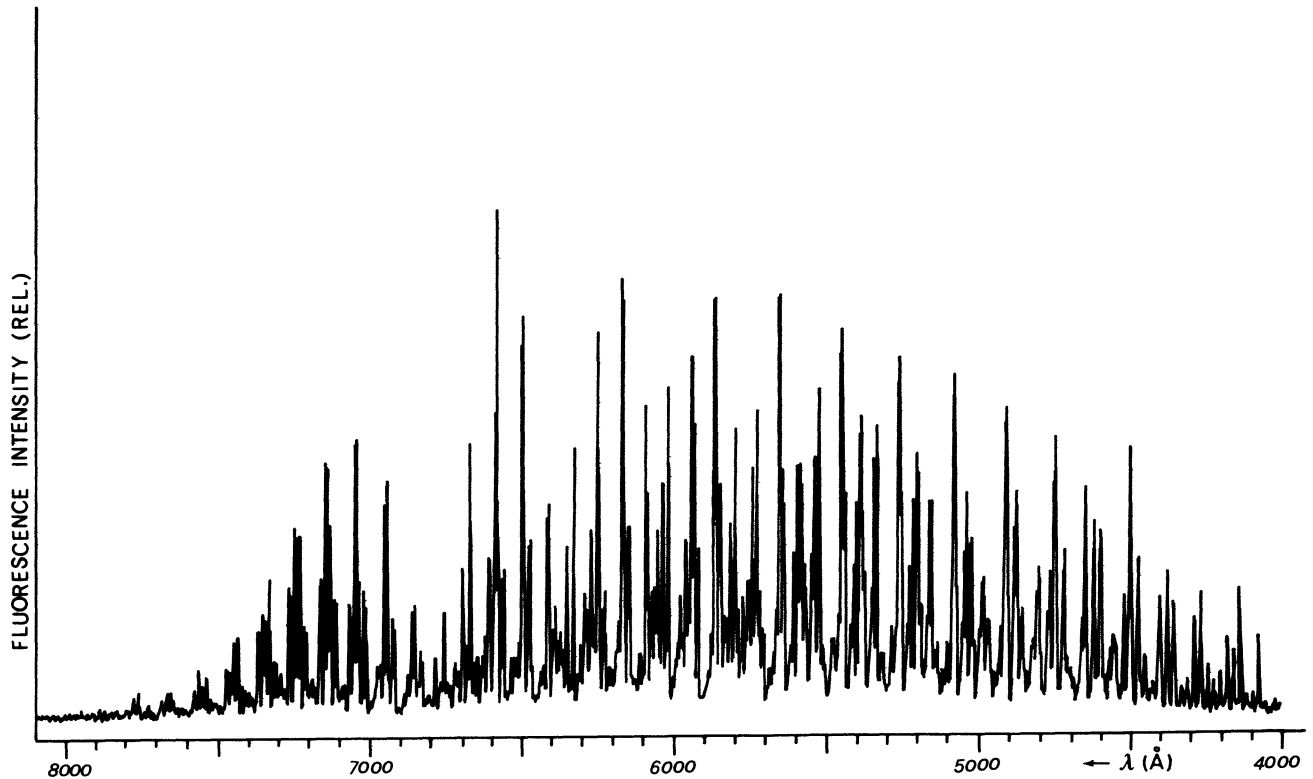

FIGURE 3 Fluorescence spectrum of natural Te_2 upon excitation with a multiline krypton ion laser at 4067 Å and 4131 Å. The excitation transitions and fluorescence progressions for the individual isotopes are given in Table III.

mm) at a distance of roughly 460 mm was initially used to obtain laser oscillation. The pump was a 15-watt krypton laser at 4067 Å or 4131 Å, or at both wavelengths (multiline operation). Four sets of mirrors with different coatings in the range of about 4000 Å to 8000 Å have been used for the laser experiments. Investigations to obtain laser oscillation in the mentioned infrared spectral range have not been performed so far. For $^{130}Te_2$ continuous laser oscillation excited by the 4067 Å line has been obtained on most of the fluorescence doublets in the spectral range of 4600 Å to 7800 Å (Figure 2). The laser lines, belonging to two fluorescence series, are given in Table II together with additional spectroscopic data. For wavelengths below 5000 Å, laser oscillation is weak and critically depends on the operating conditions. With one set of mirrors, multiline oscillation on more than 20 lines has been observed. With the 4131 Å excitation line, laser oscillation has been obtained as well but much weaker than in the case of the 4067 Å line.

Natural tellurium pumped by both 4067 Å and 4131 Å radiations gives rise to laser oscillation as well. The fluorescence spectrum (Figure 3) is extremely dense due to the contribution of different isotopic molecules. Natural Te consists of 8 isotopes with the following abundances[8]: ^{130}Te: 34.48%; ^{128}Te: 31.79%; ^{126}Te: 18.71%; ^{125}Te: 6.99%; ^{124}Te: 4.61%; ^{123}Te: 0.87%; ^{122}Te: 2.46%; ^{120}Te: 0.089%. With the help of Fourier transform spectrograms it has been possible to establish that four different molecules—$^{130}Te_2$, $^{128}Te_2$, $^{130}Te^{128}Te$, and $^{128}Te^{126}Te$—give rise to the laser effect (Table III). Some high resolution spectra, recorded in the second order of a 3.4 m grating spectrograph, showing simultaneous oscillation of different Te_2 isotopes are given in Figure 4. It is observed that the doublet lines are mainly of inequal intensity. For natural Te_2, laser oscillation has only been investigated in the spectral range of about 5200 Å to 7500 Å without any further optimization (Table IV); but it is believed that oscillation at shorter wavelengths and with higher output powers will also be possible. For $^{130}Te_2$ additional laser data are summarized in Table IV. The search for possible laser lines has been carried out using a linear two-element resonator with low output coupling. In this case thresholds of a few milliwatts can typically be obtained. For output power investigations and single line operation a linear-folded resonator with a plan end-mirror and an internal prism or interference filter has been used. For the given pump power, a maximum output coupling of about 30% was possible, which corresponds to a gain coefficient of roughly 0.04 cm^{-1}. The Te_2

TABLE II

$^{130}Te_2$ fluorescence transitions and observed laser lines

Excitation transitions $X\ 0_g^+$ ($v'' = 0, J'' = 36$) → $B\ 0_u^+$ ($v' = 16, J' = 37$) Kr^{2+} 4067 Å
$X\ 0_g^+$ ($v'' = 0, J'' = 172$) → $B\ 0_u^+$ ($v' = 18, J' = 173$)

	Laser emission $B\ 0_u^+$ ($v' = 16, J' = 37$) → $X\ 0_g^+$ ($v'', J'' = 36, 38$)				Laser emission $B\ 0_u^+$ ($v' = 18, J' = 173$) → $X\ 0_g^+$ ($v'', J'' = 172, 174$)			
	Fluorescence transitions[a] ν (cm^{-1})		Laser lines[b]		Fluorescence transitions[a] ν (cm^{-1})		Laser lines[b]	
v''	36(R)	38(P)	Observed intensity	Mirror range	172(R)	174(P)	Observed intensity	Mirror range
10	22 167.293	22 161.498						
11	21 931.945	21 926.167			21 963.711	21 937.157		
12	697.657	691.893	vw		732.346	705.867		
13	464.435	458.691	vw		502.061	475.656		
14	232.287	226.558	vw		272.855	246.522		
15	001.210	20 995.498	vw		044.731	018.469		
16	20 771.209	765.509	vw		20 817.688	20 791.499		
17	542.286	536.602	vw		591.734	565.616		
18	314.441	308.775	vw		366.865	340.823		
19	087.681	082.030	vw		143.091	117.122		
20	19 862.006	19 856.371	vw		(19 920.411)	(19 894.517)		
21	637.420	631.802			698.831	673.009		
22	413.927	408.323	vw		478.353	452.604		
23	191.528	185.940	w		258.979	233.305		
24	18 970.228	18 964.656			040.715	015.115	vw	
25	750.028	744.473	w		18 823.563	18 798.038		
26	530.935	525.395	w		607.526	582.077		
27	312.949	307.426			392.610	367.236	w	

28	096.074	090.567	m				
29	17 880.317	17 874.826	m		17 966.153	17 940.930	m
30	665.682	660.201			754.621	729.474	m
31	452.167	446.708	vs				
32	239.781	234.339	vs		334.971	309.978	vs
33	028.528	023.103			126.862	101.946	vs
34	16 818.412	16 813.003	m		16 919.902	16 895.067	
35	609.437	604.044	vs		714.100	689.340	w
36	401.607	396.232			509.458	484.776	m
37	194.928	189.570			305.981	281.378	
38	15 989.405	15 984.063	s		103.675	079.151	
39	785.043	779.718	vs		15 902.546	15 878.101	m
40	581.846	576.538			702.599	678.234	w
41	379.821	374.530				479.557	
42	178.972	173.699	vs		306.276	282.073	m
43	14 979.305	14 974.049	vs		109.912	085.790	vs
44	780.828	775.589	m		14 914.756	14 890.716	w
45	583.546	578.325				696.857	
46	387.465	382.261			528.092	504.217	w
47	192.592	187.406	vs		336.599	312.806	m
48	13 998.933	13 993.765	vs		146.340	122.632	m
49	806.495	801.345	vs		13 957.325	13 933.701	
50	615.286	610.153	vs		769.561	746.021	
51	425.314	420.200	vs		583.053	559.599	w
52	236.584	231.488	vs		397.814	374.447	s
53	049.105	044.027	s		213.852	190.571	vs
54	12 862.885	12 857.826	m		031.174	007.981	s
55	677.933	672.893			12 849.790	12 826.685	s

[a]Transition wavenumbers determined by Fourier transform spectrometry. Values corresponding to $v'' < 11$ are missing, due to the limited spectral range of the apparatus used.

[b]vw: very weak, w: weak, m: mean, s: strong, vs: very strong.

TABLE III

Fluorescence transitions and observed laser lines in natural Te_2

$^{130}Te_2$ Kr^{2+} 4067 Å

Excitation $X\ 0_g^+$ ($v'' = 0$, $J'' = 36$) → $B\ 0_u^+$ ($v' = 16$, $J' = 37$)

Laser emission $B\ 0_u^+$ ($v' = 16$, $J' = 37$) → $X\ 0_g^+$ (v'', $J'' = 36, 38$)

v''	J''	$\nu_{vac.}(cm^{-1})$[a]	Intensity[b]
31	36(R)	17 452.167	vw
	38(P)		
32	36(R)	17 239.781	vw
	38(P)	17 234.339	
35	36(R)	16 609.437	w
	38(P)	16 604.044	
38	36(R)	15 989.405	vw
	38(P)	15 984.063	
39	36(R)	15 785.043	w
	38(P)	15 779.778	
43	36(R)	14 979.305	vw
	38(P)	14 974.049	
48	36(R)	13 998.937	vw
	38(P)	13 993.765	
49	36(R)	13 806.495	m
	38(P)	13 801.345	
50	36(R)	13 615.286	w
	38(P)	13 610.153	
51	36(R)	13 425.314	vw
	38(P)	13 420.200	

$^{130}Te^{128}Te$ Kr^{2+} 4131 Å

Excitation $X\ 0_g^+$ ($v'' = 0$, $J'' = 110$) → $B\ 0_u^+$ ($v' = 14$, $J' = 109$)

Laser emission $B\ 0_u^+$ ($v' = 14$, $J' = 109$) → $X\ 0_g^+$ (v'', $J'' = 108, 110$)

v''	J''	$\nu_{vac.}(cm^{-1})$[a]	Intensity[b]
25	108(R)	18 392.991	m
	110(P)	18 376.690	
28	108(R)	17 740.253	s
	110(P)	17 724.094	
31	108(R)	17 097.681	s
	110(P)	17 081.668	
32	108(R)	16 885.769	m
	110(P)	16 869.805	

TABLE III *(continued)*

v''	J''	$\nu_{vac.}(cm^{-1})$[a]	Intensity[b]
35	108(R)	16 256.931	vs
	110(P)	16 241.114	
36	108(R)	16 049.634	w
	110(P)	16 033.866	
39	108(R)	15 434.757	s
	110(P)	15 419.140	
40	108(R)	15 232.155	w
	110(P)	15 216.588	
41	108(R)	15 030.740	vw
	110(P)	15 015.224	
44	108(R)	14 433.680	m
	110(P)	14 418.319	
45	108(R)	14 237.076	vs
	110(P)	14 221.767	
46	108(R)	14 041.691	vs
	110(P)	14 026.434	
47	108(R)	13 847.534	m
	110(P)	13 832.329	

$^{128}Te^{126}Te$ Kr^{2+} 4131 Å

Excitation $X\ 0_g^+$ ($v'' = 0$, $J'' = 116$) → $B\ 0_u^+$ ($v' = 14$, $J' = 115$)

Laser emission $B\ 0_u^+$ ($v' = 14$, $J' = 115$) → $X\ 0_g^+$ (v'', $J'' = 114, 116$)

v''	J''	$\nu_{vac.}(cm^{-1})$[a]	Intensity[b]
28	114(R)	17 698.670	s
	116(P)	17 681.377	
31	114(R)	17 052.377	s
	116(P)	17 035.240	
32	114(R)	16 839.264	m
	116(P)		
35	114(R)	16 206.943	w
	116(P)	16 190.018	
39	114(R)	15 380.414	m
	116(P)	15 363.705	
44	114(R)	14 374.375	w
	116(P)	14 357.944	
45	114(R)	14 176.846	m
	116(P)	14 160.470	
46	114(R)	13 980.558	m
	116(P)	13 964.239	

TABLE III (*continued*)

$^{128}Te_2$ Kr^{2+} 4067 Å

Excitation $X\ 0_g^+$ ($v'' = 0$, $J'' = 56$) → $B\ 0_u^+$ ($v' = 16$, $J' = 57$)

Laser emission $B\ 0_u^+$ ($v' = 16$, $J' = 57$) → $X\ 0_g^+$ (v'', $J'' = 56, 58$)

v''	J''	$\nu_{vac.}(cm^{-1})$[a]	Intensity[b]
28	56(R)	18 054.1	vw
	58(P)	18 045.6	
31	56(R)	17 406.6	vw
	58(P)	17 398.1	
35	56(R)	16 559.4	vw
	58(P)	16 551.0	
38	56(R)	15 936.2	vw
	58(P)	15 927.8	
39	56(R)	15 730.8	vw
	58(P)	15 722.4	
43	56(R)	14 921.1	vw
	58(P)	14 912.7	
48	56(R)	13 936.0	w
	58(P)	13 927.7	
49	56(R)	13 742.7	m
	58(P)	13 734.4	
50	56(R)	13 550.6	w
	58(P)	13 542.3	

[a]Transition wavenumbers determined by Fourier transform spectrometry.
[b]vw: very weak, w: weak, m: mean, s: strong, vs: very strong.

system can be operated with multimode and single mode excitation as well. Lowest thresholds and highest efficiencies are obtained with single mode excitation tuned exactly to resonance; however, as the single mode power of the krypton laser was only a smaller fraction of the multimode power, with multimode excitation higher output powers were obtained.

The Te_2 laser has also been operated with a ring resonator. For most of the lines unidirectional oscillation is obtained, as is typical for these coherently excited Raman-type dimer lasers.[1] However, for some lines weak oscillation in the backward direction is also observed. Investigations of this backward oscillation have not been performed so far, but they may give some interesting information about relaxation processes and the gain contribution due to population inversion.[1]

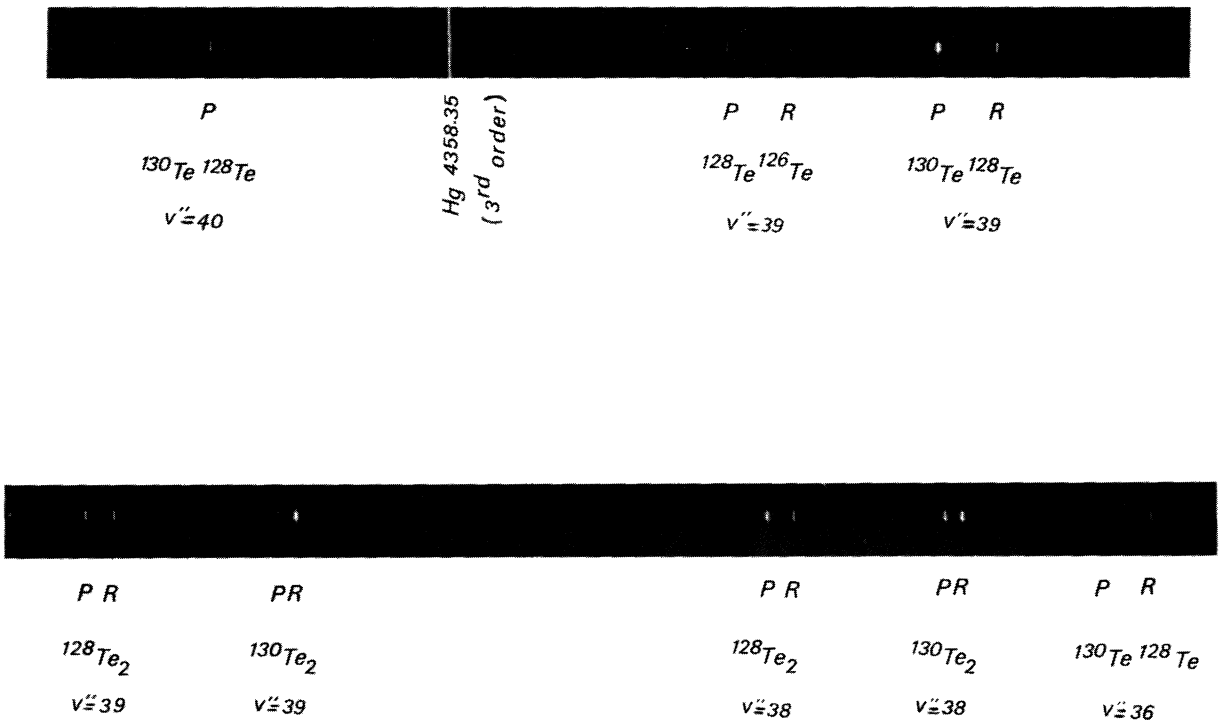

FIGURE 4 High resolution spectrum of laser doublets in the range 6200–6600 Å.

TABLE IV

Summary of Te_2 laser data

	$^{130}Te_2$	Natural Te_2
Pump line	4067 Å	4067 Å 4131 Å
Laser range	4600–7800 Å	5200–7500 Å[a]
Laser lines	~50 doublets	~40 doublets
Threshold	<2 mW; < 6 W cm^{-2}	<10 mW
Pump power (multimode)	up to 600 mW	~1500 mW (both lines)
Output power (multiline)	up to 30 mW (output coupling 4%)	3 mW (output coupling <0.5%)
Output power (single line)	up to 10 mW	—
Gain[b]	~0.04 cm^{-1}	—
Length of vapor zone	7 cm	
Temperature Reservoir/cell	~510°C/700°C	
Dimer Pressure	~2.5 m bar	

[a]For natural Te_2 laser investigations at shorter wavelengths have not been performed.
[b]Determined from maximum output coupling.

CONCLUSION

The technique of fixed-frequency laser-induced fluorescence together with high resolution Fourier transform spectrometry has enabled systematic investigations of optically pumped laser oscillation in natural tellurium and $^{130}Te_2$ dimers. So far hundreds of new laser lines in the spectral range of 4600 Å–7800 Å on the $B\ 0_u^+ \rightarrow X\ 0_g^+$ transition have been obtained. Taking into account Franck-Condon factors and fluorescence spectra, one can anticipate the enlargement of this spectral range in the infrared up to about 1.5 μ on the $B\ 0_u^+ \rightarrow b\Sigma_g^+$ transition. At least, intense fluorescence lines on this transition have been observed.[7]

Furthermore, additional laser lines may be achieved by modifying the composition of natural tellurium, and it should also be possible to extend the laser range slightly towards the blue by proper operation of the laser system. Due to its dense emission spectrum, Te_2 fluorescence and laser

lines may be considered as a reference system comparable to that provided by iodine.[9] Besides, the fluorescence spectra display strong relaxations, which may lead to more closely distributed collision-induced laser lines in cells containing a buffer gas. Under this aspect investigations on the Te_2 system are of further actual interest.

Acknowledgment

This work was supported by C.N.R.S. through International A.T.P.

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