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Notes and Lines

New Infrared Laser Lines in Argon, Krypton, and Xenon

E. L. BROWN, M. A. GUNDERSEN, AND P. F. WILLIAMS

Abstract—Fourteen new infrared laser transitions in Ar, Kr, and Xe ranging from 3.725 to 17.233 μm are reported. All of the Kr laser transitions and all but one of the Ar and Xe transitions have upper levels in the s or d state: the majority of the transitions satisfy the condition $\Delta K = \Delta J$. Identification of a previously observed, unidentified laser line at 5.804 μm in Ar is given.

Nobel gas lasers are of interest for many applications because they exhibit long-term frequency stability, a wide range of wavelengths, and useful power levels. In pulsed systems high peak power has been observed over a wide range of wavelengths [1], [2], [6]. Because of the interest in new infrared (IR) lasers and the lack of published data in the IR under these conditions, a survey of Ar, Kr, and Xe was conducted. A 3 m longitudinal discharge laser capable of operation at pressures down to 5 μm and at current densities up to approximately 175 A/cm² was used. Nineteen new transitions were observed, ranging in wavelength from 3.631 to 17.233 μm . All but one of the transitions were identified as neutral species, although it had been anticipated that new transitions at high current densities would be ionized species. The strongest transitions were observed in Kr at 4.15 μm and Xe at 7.8 and 8.4 μm .

A longitudinal discharge laser was used for this work [2]. The plasma tube was made from Pyrex tubing with an inside diameter of 19 mm and the electrical discharge was divided into two 1.5 m sections for better stability. Each end of the laser contained an indium cathode [3] and the center electrodes were grounded anodes constructed from molybdenum. The ends of the plasma tube were sealed with KCl Brewster windows which passed radiation with wavelengths from 0.1 to 17 μm , although the optical quality of the windows hindered oscillation at wavelengths below roughly 1 μm . The optical cavity consisted of a 20 m radius mirror and a flat containing a 3 mm diameter pinhole to couple out radiation. Both mirror surfaces were silver overcoated with thorium oxyfluoride. 0.2 μF capacitors were charged to a voltage of 16–20 kV and discharged through the plasma tube with a spark gap. The current rise time was roughly 1 μs , and peak current densities up to 176 A/cm² were obtained.

Rough wavelength measurements were made with various filters and more accurate measurements were made using a half meter double spectrometer with 600 grooves/mm gratings blazed at 16 μm . The laser radiation was detected by a HgCdTe detector with sensitivity extending to 18 μm . With this apparatus, measurements accurate to 0.003 μm could be made between 2 and 18 μm .

EXPERIMENTAL RESULTS

Table I gives a listing of the new laser transitions observed in each of the noble gases [7]. The light pulse occurred typically 100 ns after the beginning of the current pulse and had a pulse length of 200–400 ns depending on the pressure and gas used.

In order to make term assignments, a computer program was

TABLE I
INFRARED TRANSITIONS OBSERVED IN Ar, Kr, AND Xe LASERS

	Measured Wavelength (Microns) ± 0.003	Calculated Wavelength μ	Term Assignment	Relative Line Strength ^a	Pressure (Microns)
Ar	5.804 ^b	5.8037	4d[14] ₂ -5p[14] ₂	w	20
	6.812	6.8129	4d[4] ₁ -5p[14] ₂	w	250
	7.956	7.955	5d[14] ₁ -6p[14] ₁	m	20
	11.042	11.0415	7p[4] ₀ -7s[14] ₁	w	20
	13.475			m	250
Kr	8.115	8.1151	8s[14] ₁ -7p[24] ₂	w	50
	9.637	9.639	6d[14] ₁ -7p[14] ₁	w	50
	10.937	10.9337	5d[4] ₁ -6p[4] ₁	m	50
	17.070	17.0709	4d'[24] ₃ -6p[24] ₃	m	50
	17.233	17.2328	4d'[24] ₃ -6p[24] ₂	m	50
Xe	3.725	3.7265	5d'[24] ₂ -7p[24] ₃	w	150
	7.767	7.7665	6d[14] ₁ -7p[14] ₁	s	150
	7.782	7.7813	5d'[14] ₁ -8p[14] ₁	s	150
	8.404	8.4042	6d[14] ₁ -7p[4] ₀	s	150
	11.582	11.5821	9p[4] ₀ -9s[14] ₁	m	20

^a Pulse energies for the strong lines were of the order .1–1 mJ.

^b reported in Ref. 5.

written to calculate the energy differences between known energy levels [4], [7] satisfying the jl coupling selection rules ($\Delta l = +1$ and $\Delta J = 0, \pm 1$). The measured wavelengths were then compared to those calculated. The procedure was followed not only for neutral species but also for the known ionized species. All but one of the observed wavelengths were identified with neutral transitions. No term assignment could be given to the 13.475 μm line observed in Ar, even after including all the known ionized transitions.

Term assignments obtained in this manner are given in Table I using Racah jl notation with the upper levels given first. The energy levels are denoted by $n[K]j$ with an unprimed l value indicating a $^2P_{3/2}$ core state and a primed l value indicating a $^2P_{1/2}$ core. Transitions appear to occur in groups sharing initial levels.

A term assignment for the previously observed but unidentified 5.804 μm line in Ar [5] is given in the table. Two $d'-p$ transitions in Kr and two transitions in Xe have been listed in the Table that involve a change in the parent core configuration. Although these transitions are forbidden by selection rules for jl coupling, similar transitions have been previously reported [1]. In addition, because the density of states increases for the high lying levels, for an equal distribution of electron energies these levels have a lower probability of attaining a sufficient population to produce an inversion. Therefore, transitions with upper levels above about 8s, 7p, 6d are neglected.

All of the Kr transitions observed and all but one of the Ar and Xe transitions have upper levels in the s or d state. Previous observations [1] suggest that inversion between the $s-p$ and $d-p$ states can be produced by electron impact excitation. When the primary excitation mechanism is due to electron impact with electron energies well above the threshold, excita-

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tion can occur predominately from the ground state p -shell to s and d states that are strongly connected to the ground states by optical transitions. From a calculation of line strengths, it may be shown that the strongest transitions satisfy $\Delta K = \Delta J$ [2]. All of the transitions in Xe, all but one transition in Ar, and half of the transitions in Kr satisfy this condition.

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Optically Pumped CW Submillimeter Emission Lines from Methyl Mercaptan CH_3SH

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Abstract—Twenty-three new emission lines of CH_3SH have been observed and their polarization with respect to the pumping radiation determined, but no assignments have yet been made.

Methyl mercaptan, CH_3SH , is the sulphur analogue of methanol, a molecule which is shown to have a large number of CW FIR emission lines [1]–[6]. Many of the emission lines of methanol have short wavelengths due partly to the internal rotation effects and partly to the large rotational A constant. The infrared spectrum of methyl mercaptan shows two fundamentals which absorb in the region of operation of the CO_2 laser; the $\nu_6(A')$ and $\nu_{11}(A'')$ methyl wagging vibrations at 1072 and $956\ \text{cm}^{-1}$, respectively [7]. It therefore seemed to be a good candidate for production of FIR radiation at relatively short wavelengths. Absorption of the CO_2 laser radiation was monitored optoacoustically, and although CH_3SH seems to absorb with about an order of magnitude less intensity than CH_3OH , twenty-three FIR emission lines were discovered on a total of fifteen pumping lines.

The CO_2 pump laser was constructed from a 2 m long 12 mm ID discharge tube with a resonator comprised of a 150 line/mm gold coated grating and a 20 percent transmission, 10 m radius of curvature CdTe output coupling mirror. The FIR

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TABLE I
SUMMARY OF EXPERIMENTAL RESULTS

Laser line	Wavelengths (microns)	Polarization relative to pump polarization	CO_2 Threshold power (Watts)	F.I.R. power
9 μR (34)	116		8.5	VW
9 μR (30)	161		9	W
9 μR (28)	351	⊥	4	S
9 μR (18)	185	⊥	2.5	S ^c
	205		5.5	W
9 μP (12)	403	⊥	6	M
9 μP (16)	341	⊥	10	VW
	384		6	S
9 μP (18)	127	⊥	10	VW
9 μP (22)	379		3	S
9 μP (24)	128		6.5	VW
9 μP (30)	298		5	M
9 μP (38)	319		4	S
	262		8	W
9 μP (44)	224		10	VW
	316	a	a	VW
	456		3.5	M
10 μR (34)	117		7	VW
	147	a	a	VW
	234	⊥	8	VW
	370		7	M
10 μP (24)	124	b	8	W
10 μP (16)	324	⊥	8	W

cavity consisted of two internal gold coated mirrors of 10 cm diameter separated by almost 2 m. Both mirrors had a radius of curvature of 2.5 m; the input coupling hole was 1.5 mm diameter and the output coupling hole was 2 mm diameter. The output mirror was mounted on a translator with a motor driven micrometer screw to allow the cavity length to be continuously varied and the resulting cavity scans were used for measurement of the FIR wavelengths. The pumping radiation was chopped for synchronous detection and focussed through the input coupling hole using a mirror with a radius of curvature of 50 cm. A Golay cell was used to detect the FIR emission.

The observed emission lines and their relative polarizations are listed in Table I. They were all observed with optimum pressures of 50–100 mtorr as measured by a Pirani gauge. The wavelengths reported are estimated to be accurate to 2 percent. Note that the greatest number of lines reported are pumped in the 9 μm region and that many of these are the strongest emissions with the lowest threshold power. This probably reflects the greater intensity of the ν_6 band.

It would be satisfying to assign these transitions, but unfortunately the most recent publication of the CH_3SH spectrum in the 9–10 μm region contains only very rough values of the band origins and does not even have a rotational analysis [7]. It thus appears that an assignment or even partial identification of these transitions is not yet possible with the present state of knowledge of the ν_6 and ν_{11} fundamentals of CH_3SH .

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