Optically Pumped Atomic Thulium Lasers

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Abstract—**A thulium vapor column is irradiated with the fourth harmonic of a Nd:YAG laser at 266 nm. The near coincidence with a dipole allowed transition from ground state results in efficient optical pumping yielding 87 simultaneous superfluorescent laser transitions from 300 to 900 nm. Many of the emissions are due to inversions to the ground state of both neutral and singly ionized Tm atoms.**

THIS letter reports what we believe to be the first observations of laser action in optically pumped atomic thulium vapor. A Tm vapor column was irradiated with intense radiation at 266 nm, the fourth harmonic of a Nd:YAG laser; the presence of nearly resonant single photon transitions from the ground state of the neutral and singly ionized species resulted in efficient optical pumping to excited states by the 266 nm light. Superfluorescent laser emission was observed on 87 simultaneously occurring transitions from the region of 300 to 900 nm. Many of these new laser transitions correspond to inversions to the ground state of both the neutral and singly ionized atoms.

Because of its rich electronic spectrum, Tm is an interesting candidate for investigations of new laser transitions. The near coincidence of the fourth harmonic of a Nd:YAG laser at 266 nm with the neutral Tm ($4f^{13}6s^2$ - $4f^{13}6s7p$) transition and the singly ionized Tm$^+$ ($4f^{13}6s$ - $4f^{12}5d6s$) transition suggests that very high levels of atomic excitation as well as efficient multiphoton ionization will lead to a variety of population inversions. A schematic diagram of a few of the pertinent Tm energy levels is shown in Fig. 1. Following single photon excitation to the Tm ($4f^{13}6s7p$) level at 37576 cm^{-1}, many cascade transitions exhibit superfluorescent laser emission along the various electric dipole allowed paths from this state. In addition, photoionization of the $4f^{13}6s7p$ state by absorption of an additional 266 nm photon may occur. The ground state Tm$^+$ ion may likewise be optically pumped in the wings of the nearly resonant Tm$^+$ ($4f^{12}5d6s$) state. Cascade superfluorescence from the excited ion is thereby seen. It is apparent from the complex electronic structure of the Tm atom that a multitude of possible laser emissions can result, and in this experiment one observes 87 simultaneously occurring new laser transitions from 300 to 900 nm. No attempt was made to catalog possible emissions occurring at longer wavelengths, although many such lasers were undoubtedly present.

The experimental arrangement for this work was particularly simple. A commercial Nd:YAG laser (Quanta-Ray model DCR-1A) was frequency quadrupled using successive KDP crystals to generate 40 mJ at 266 nm with a pulse length of approximately 4 ns. This radiation was focused to an area of ~10^{-3} cm^2 over 20 cm in the central hot zone of the Tm metal vapor cell. The Tm cell was constructed in such a way as to permit extended use at our operating temperature of 1700°C, corresponding to a Tm ground state number density of about 5×10^{18} atoms/cm^3, as determined by a resonance line curve of growth measurement [1]. An alumina tube with a tantalum liner served as the cell body. Cooled suprasil end windows were sealed to each end of the alumina tube with O-rings. Up to 400 torr of argon buffer was used in the cell to prevent condensation of the Tm metal onto the windows. The central region was heated using a tube oven constructed of platinum-rhodium braided high-temperature windings. Radiation emitted from the cell ends was imaged into a 0.85 m monochrometer and recorded.

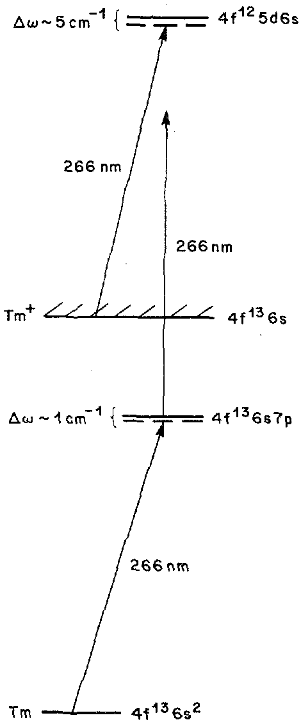

Fig. 1. Schematic energy level diagram for Tm indicating the excitation path for the neutral atom and subsequent photoionization and excitation of the single ion by 266 nm photons.

Table I is a compilation of the observed laser transitions. All of the emissions were narrow and exhibited a linewidth of less than 0.1 Å, the spectrometer resolution. Where identification was possible, the lasing species and the most probable initial and final states involved are indicated. In view of the literally tens of thousands of tabulated Tm transitions [2], in many cases identification of the levels involved is difficult if not impossible using the present arrangement. The approximate strength for each line is indicated by "*S*" for strong, "*M*" for moderate, and "*W*" for weak. Since these emissions were all

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TABLE I
OBSERVED SUPERFLUORESCENT LASER EMISSIONS FROM 300 TO 900 nm (ALL WAVELENGTHS GIVEN IN AIR). KEY TO EMISSION STRENGTHS: "*S*" FOR STRONG, "*M*" FOR MODERATE, "*W*" FOR WEAK. A DASH (—) INDICATES AN UNKNOWN VALUE.

Observed Wavelength (nm)	Ionic Species	Upper Laser Level (cm^{-1})	Lower Laser Level (cm^{-1})	Relative Emission Strength
305.1	I	-	-	W
307.9	I	32446	0	W
336.5	II	29967	237	W
340.2	II	38361	8957	W
349.7	I	28564	0	W
356.6	I	28051	0	W
356.8	I	28024	0	W
361.0	II	27702	0	W
370.4	II	35754	8770	W
371.8	I	26889	0	W
372.2	I	44307	17454	M
372.9	II	35754	8957	W
376.3	II	26575	0	W
385.8	II	34871	8957	W
411.8	I	41012	16742	M
412.2	I	44001	19735	W
415.1	I	41841	17752	W
436.0	I	22930	0	W
436.2	I	40533	17613	W
441.5	I	39386	16742	W
488.1	I	39479	18990	W
506.1	I	19754	0	S
506.2	I	19748	0	S
511.5	I	19548	0	M
530.7	I	18837	0	M
563.4	I	-	-	S
567.6	I	17614	0	S
576.6	I	17343	0	M
580.9	I	40517	23309	M
597.1	I	16742	0	S
601.9	-	-	-	M
611.9	I	33793	17454	M
617.0	I	35054	18853	M
640.4	I	35363	19748	S
646.1	I	32217	16742	S
651.9	I	37138	21799	W
658.9	I	40128	24957	W
662.0	I	23873	8771	M
665.8	I	32359	17343	S
672.3	I	38751	23882	M
678.2	I	32359	17614	S
684.5	I	32359	17753	S
684.8	I	32217	17614	S
691.4	I	37768	23309	M
695.6	I	37711	23335	M
698.8	I	-	-	M
699.6	I	41728	27440	M
703.8	I	33961	19753	M
704.3	II	-	-	M
733.9	I	35363	21737	M
734.2	I	37557	23941	M
739.2	I	32359	18837	W
740.6	I	-	-	W
740.8	II	-	-	W
740.9	I	33240	19748	W
743.6	II	-	-	W
743.9	I	-	-	M
744.8	I	38123	24701	M
748.1	I	35363	21997	S
752.2	I	41743	28448	W
752.6	I	43408	30125	W
753.2	I	-	-	W
755.9	I	-	-	S
757.7	I	41646	28448	W
766.9	I	39741	26701	W
776.8	I	37576	24701	W
780.4	I	-	-	S
784.2	II	35682	22929	W
784.5	I	39444	26701	W
784.9	I	-	-	M
792.9	I	32359	19754	S
793.1	I	32359	19748	S
798.3	I	37138	24611	S
802.0	I	32217	19748	S
817.0	I	33396	21161	W
818.0	I	39259	27037	W
818.6	I	39709	27491	W
821.5	I	37657	25488	M
823.3	I	-	-	W
838.0	I	37138	25207	M
848.0	I	-	-	M
848.2	I	-	-	M
853.3	I	42667	30947	W
866.0	II	-	-	W
873.0	I	39479	28024	W
880.3	I	32479	21120	W
883.6	I	32479	21161	M

superfluorescent and no external cavity resonator was used, it is entirely possible that the relative intensity of these lasers could be greatly manipulated or the strength of a particular transition enhanced by the use of an optical resonator. The total output energy at all wavelengths was measured to be about 5 mJ per pulse or 13 percent conversion of the 266 nm pump radiation.

It is apparent from the transition identifications in Table I that many of the observed lasers correspond to inversions to the ground state of neutral Tm and, in a few cases, singly ionized Tm^+. Assuming an oscillator strength of $f = 0.1$ for the Tm $(4f^{13}6s^2 - 4f^{13}6s7p)$ transition, one would expect a single photon pump rate of $W = 3.7 \times 10^{13}\ s^{-1}$ from the ground state at the 266 nm power density of 1×10^{10} W/cm^2. Rapid photoionization or cascade decay out of the Tm $4f^{13}6s7p$ state may then empty this upper pump level, resulting in a marked depletion of the Tm ground state population and the creation of highly excited Tm vapor. Under these conditions population inversions to the ground state may result [3]. This process is the archetypal three-level laser system ala the ruby laser and is possible because of the strong resonance pumping [4]. Superfluorescence on the resonance line of strontium using two-photon pumping has recently been demonstrated using this principle [5]. Once Tm^+ ground state ions have been produced by photoionization of the Tm $(4f^{13}6s7p)$ state, a similar process may occur in the wings of the Tm^+ $(4f^{13}6s - 4f^{12}5d6s)$ transition, accounting for the various ionic inversions observed. The outputs listed in Table I were observed in both the copropogating and counterpropogating directions relative to the pump beam, indicating that the transitions were indeed due to population inversions and not due to parametric four-wave mixing processes. The temporal behavior of the emissions followed the 266 nm pulse and a 4-10 ns duration output pulse was typical.

Threshold behavior for the multitude of laser transitions developed approximately as follows. Laser transitions occurring in neutral Tm exhibited a pumping threshold of about 3×10^8 W/cm^2, while the threshold for stimulated emission in the ion was approximately 4×10^9 W/cm^2, or a factor of 13 higher. The higher threshold for the ionic lasers is intuitively correct based on the model of multiphoton ionization followed by single photon excitation from the ionic ground state. The strength of the emissions showed no marked dependence on buffer gas pressure up to Ar pressures of 400 torr, although very high buffer gas pressures could lead to significant collisional redistribution of the excited state populations and subsequently, different laser behavior.

This work has demonstrated 87 new laser transitions in TM vapor through relatively simple optical excitation. Many other rare-earth elements possess similar single photon absorption coincidences with harmonics of Nd:YAG or excimer laser wavelengths, and based on this work one would expect a tremendous variety of new laser transitions to result.

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