

UC Berkeley

UC Berkeley Previously Published Works

Title

Mid-IR laser action in the H₃ Rydberg molecule and some possible astrophysical implications

Permalink

<https://escholarship.org/uc/item/2gp424dh>

Journal

AIP Conference Proceedings, 1642(1)

ISSN

0094-243X

ISBN

9780735412828

Author

Saykally, Richard J

Publication Date

2015-01-22

DOI

10.1063/1.4906707

Peer reviewed

Mid-IR laser action in the H 3 Rydberg molecule and some possible astrophysical implications

Richard J. Saykally

Citation: [AIP Conference Proceedings](#) **1642**, 413 (2015); doi: 10.1063/1.4906707

View online: <http://dx.doi.org/10.1063/1.4906707>

View Table of Contents: <http://scitation.aip.org/content/aip/proceeding/aipcp/1642?ver=pdfcov>

Published by the [AIP Publishing](#)

Articles you may be interested in

[Precision spectroscopy comes to the mid-IR](#)

Phys. Today **68**, 16 (2015); 10.1063/PT.3.2866

[High brightness from unstable resonator mid-IR semiconductor lasers](#)

J. Appl. Phys. **107**, 123113 (2010); 10.1063/1.3435208

[Improved thermal management of mid-IR quantum cascade lasers](#)

J. Appl. Phys. **103**, 043103 (2008); 10.1063/1.2840136

[Mid-IR interband cascade lasers for remote chemical sensing applications](#)

AIP Conf. Proc. **420**, 603 (1998); 10.1063/1.54852

[Precision spectroscopy comes to the mid-IR](#)

Phys. Today ; 10.1063/PT.5.7180

Mid-IR Laser Action in the H₃ Rydberg Molecule and Some Possible Astrophysical Implications

Richard J. Saykally

Department of Chemistry, University of California, Berkeley, California 94720-1460

Abstract. Mid-IR lasing has been observed in supersonic plasmas and assigned to d to p transitions in tri-atomic hydrogen Rydberg states. Possible astrophysical implications are discussed.

Keywords: triatomic hydrogen, laser, first stars

PACS: 30, 90

While searching for IR spectra of molecular ions in pulsed planar supersonically expanding plasmas with cavity ringdown spectroscopy[1,2] in the mid-1990s, we accidentally observed numerous laser transitions near 7 microns originating in the ringdown supercavity itself. Many of these, observed in water/rare gas mixtures, were assigned to highly vibrationally excited water molecules[3]. We proposed a population inversion mechanism based on generating excited water molecules “from the top down” by dissociative electron attachment to hydronium ions. This has since been supported by detailed studies of ION–electron recombination [4].

Subsequently, numerous lasing transitions were observed in the same spectral region in mixtures of hydrogen and rare gases. At the time, we were unable to assign these to a specific carrier. However, following recent discussions with Professor Chris Green, assignment to transitions among rovibronic states of the Rydberg molecule H₃, generated by electron recombination with the H₃⁺ ion, seemed feasible. Specifically, their detailed calculations[5] showed that the d-states($\ell=2$) are relatively long-lived, while the p-states($\ell=1$) predissociate rapidly, yielding an “excimer-like” population inversion mechanism. Moreover, these d- to p-state transitions occur exactly in the region where lasing is observed, as shown in Figure 1. The flowing afterglow experiments of Glosik et al[6] suggest a 3-body “collision assisted recombination” mechanism, rather than a simple 2-body recombination process, because of the relatively high($> 10^{14}$ cm⁻³) gas density obtaining in the supersonic discharge source.

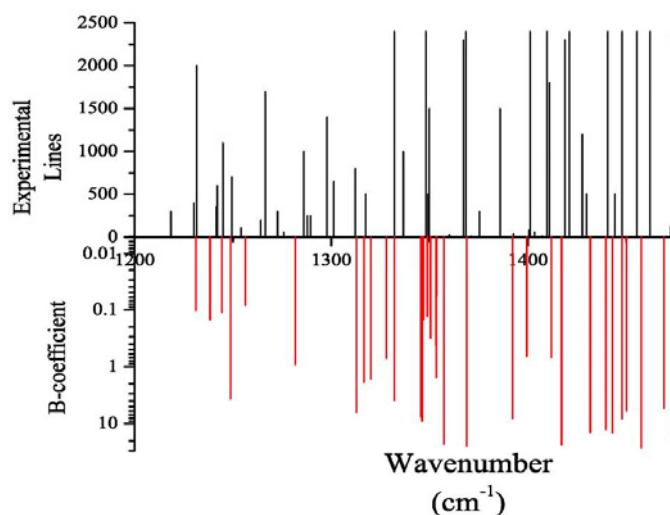


FIGURE 1. Comparison of experimental results with calculated nd \rightarrow n'p transitions of H₃. The experimental laser strength is in arbitrary units with linear scale, while the present theoretical stimulated emission (B)-coefficients are in the units of 1022 (m/JS²) on a logarithmic scale. The calculated line positions have a precision of about 13 cm⁻¹.

The Berkeley spectrometer used to produce and measure IR laser transitions generated in supersonic plasmas is described in Reference 3. Laser emission lines were produced from an ultrahigh finesse optical supercavity containing a supersonically expanding plasma in the spectral range of 930 - 4,370 cm^{-1} , with the majority occurring near 7 micrometers (1430 cm^{-1}). The supersonic slit discharge generates a weakly ionized plasma with neutral gas density expanding as the reciprocal of distance from the slit nozzle, such that the pressure in the region of observation is in the range of 0.1 to 10 Torr (ca. $10^{15} - 10^{17} \text{ cm}^{-3}$). Fifty-seven laser transitions were observed in both H_2 -He and H_2 -Ne gas mixtures. Twenty-nine laser transitions found in H_2 -He were also observed in H_2O -He or H_2O -Ne discharges. Our terahertz spectroscopy studies [7] of similar supersonic plasmas and associated optical processes involving dilute H_2O /rare gas mixtures, wherein a set of stimulated emission transitions were found to be produced only in such mixtures, indicated a molecular ion rotational temperature near 100K and a vibrational temperature near 2000K at the pressures (110 Torr) prevailing in the active region of the discharge. It was found that the laser intensity was highly dependent on partial pressure of H_2 in the expanding gas mixture, maximizing the intensity of the strongest lines at a S/N ratio >2000 when the mixing ratio was $< 1\%$. No lasing was observed when pure H_2 gas was used for the expansion, although no broad spectral searches were conducted.

SOME POSSIBLE ASTROPHYSICAL IMPLICATIONS

The observation of intense infrared stimulated emission transitions in triatomic molecular hydrogen may have some interesting applications in several astrophysical contexts. The experimental conditions under which these have thus far been observed (densities $>10^{14} \text{ cm}^{-3}$, low H_2/RG ratios) do not seem directly relevant to interstellar clouds, or indeed to cosmic processes in general, primarily because of the low hydrogen fractions that optimized the laser intensities. However, we repeat that these lasers were discovered accidentally while seeking other ends, and the conditions and spectral coverage were not examined very extensively. It may well turn out that these or similar laser transitions can be observed under other conditions and in other spectral regions. The results presented here are clearly suggestive, not definitive and are designed to motivate further studies (and funding thereof!). Nevertheless, we speculate regarding some contexts wherein these H_3 lasers could prove to be important.

Detection of the First Stars and Galaxies Formed in the Universe

The first stars are believed to have formed a few hundred million years after the Big Bang, from gas accreted in dark matter halos. In order for stellar densities to be achieved, the accreting gas must cool by converting translational energy to radiation, which is accomplished in secondary stars largely via far-IR emission from collisionally excited fine-structure levels of carbon and oxygen atoms. In the first stars, only hydrogen and helium (and traces of lithium) were available, and it is believed that far-IR radiation via collisionally excited quadruple-allowed rotational transitions in trace amounts H_2 was the primary cooling mechanism. This very inefficient cooling requires that the first stars were extremely massive ($> 10^5$ solar masses). When these first stars turned on, they emitted extremely intense UV light that ionized surrounding gas. Nearby dark matter halos could thereby exhibit other cooling mechanisms that require ionization of the accreting gas. Hence, species such as HeH^+ , HD^+ , and H_3^+ could be important in the formation of the first galaxies. In particular, if H_3^+ were formed in significant densities, recombination with electrons to produce neutral H_3 Rydberg molecules may have been prominent and the production of IR, and possibly far-IR (via high l-states) emission, and perhaps stimulated emission could have occurred. If this were the case, the production of stimulated emission from such massive bodies over large pathlengths could have produced light that may presently be detectable with current instrumentation. Huge redshifts (ca. 25) would be expected, such that the 7 micron lasing reported here would be shifted deep into the far-IR. Hence, careful examination of the 200-500 micron region with modern high resolution telescopes (e.g. HiFi) could prove interesting in this context.

Laser Cooling: A Novel Mechanism for Primordial Star/Galaxy Formation

If indeed H_3^+ forms in primordial star forming regions, then rotational transitions would be excited in collisions with H and He, just as for the neutral H_2 molecule-but with much stronger coupling to translation due to the net charge. Rydberg states formed upon electron-ion recombination would exhibit a correspondingly excited ion core, which would engender new IR emission lines that would effectively radiate away translational energy. Also, the dense Rydberg state manifold itself could couple strongly to translational motions in H and He, via state changing collisions that could effect laser cooling.

Plasma Dynamics in the Atmospheres of Giant Planets

Given that strong IR emission from H_3^+ has been observed from all three local giant planets [10, 11, 12] it might be expected that IR emission from Rydberg states of H_3 could also be observable. If so, this could produce new constraints on the nature of their outer atmospheres.

Other objects that might exhibit H_3 Rydberg lasing (suggested by Prof Chris McKee) are atmospheres of low mass stars that are cool enough to have significant H_2 and that have large flares, shocks in molecular gas that are fast enough to produce significant ionization, and gamma ray bursts or X-ray flashes that impinge on molecular gas, such that the post-burst gas simultaneously has high ionization and a high H_2 content.

ACKNOWLEDGEMENT

This work was supported by the Chemical Physics Program of the AFOSR under Dr. Michael Berman.

REFERENCES

1. J. B. Paul, R. A. Provencal, C. Chapo, E. Michael, A. Pettersson, and R. J. Saykally, ACS Symposium Series 720 on Cavity-ringdown spectroscopy: an ultratrace-absorption measurement technique, (1997).
2. R.N. Casaes, R.A. Provencal, J.B. Paul, and R.J. Saykally, J. Chem. Phys. 116, 6640-6647, (2002).
3. E. A. Michael, C. J. Keoshian, D. R. Wagner, S. K. Anderson, and R. J. Saykally, Chem. Phys. Lett. 338, 277-284, (2001).
4. R. D. Thomas, et al, J. Phys. Chem. A 114, 4843 (2010).
5. R. J. Saykally, et al., J. Phys. Chem. (submitted 10/2010).
6. J. Glosik, I. Korolov, R. Plasil, O. Novotny, T. Kotrik, P. Hlavenka, J. Varju, I. A. Mikhailov, V. Kokoouline, and C. H. Greene, J. Phys. B-At. Mol. Opt. 41, 191001 (2008).
7. E. A. Michael, C. J. Keoshian, S. K. Anderson, and R. J. Saykally, J. Mol. Spectrosc. 208, 219 (2001).
8. V. Bromm and R. B. Larson, Annu. Rev. Astron. Astrophys. 42, 79 (2004).
9. V. Brown, N. Yoshida, L. Heruquist, and C. F. McKee, Nature. 459, 49 (2009).