## RAMAN AND GAS CHROMATOGRAPHIC ANALYSIS OF $H_2(1/4)$ AND DIMER $[H_2(1/4)]_2$

Abstract: Molecular hydrino  $H_2(1/4)$  and the corresponding dimer  $[H_2(1/4)]_2$ , were observed by Raman spectroscopy by predicted features being (i) a single J = 0 to J = 1 rotational transition of  $H_2(1/4)$  involving a matrix enhanced inverse Raman effect (IRE), (ii) a single J = 0 to J = 2rotational transition of  $H_2(1/4)$ , (iii) a series of rotational transitions of  $[H_2(1/4)]_{\gamma}$  combined with a J = 0 to J = 1 rotational transition of both  $H_2(1/4)$  members of the dimer, (iii) a series of rotational transitions of  $[H_2(1/4)]_2$ , combined with a J = 0 to J = 2 rotational transition of one  $H_2(1/4)$  member of the dimer, and (iv) a split peak corresponding to the J = 0 to J = 2 rotational transition of one  $H_2(1/4)$  member combined with a J=0 to J=1 rotational transition of the other  $H_2(1/4)$  member of the dimer wherein the splitting was due to a rotational transition of  $\left[H_2\left(1/4\right)\right]_2$ . Both  $H_2\left(1/4\right)$  and  $\left[H_2\left(1/4\right)\right]_2$  were further observed by gas chromatography that showed a gas from hydrino producing reactions with a faster migration rate than that of any known gas considering that hydrogen and helium have the fastest prior known migration rates and corresponding shortest retention times. Moreover, this so-called fast peak predicted for molecular hydrino was shown to be manifest as a plurality of peaks corresponding to spin isomers observed by electron paramagnetic resonance spectroscopy (EPR). Depending on the sample conditions, a prior unknown peak was also observed at twice the migration time of the fast or molecular hydrino peak and just after the hydrogen peak that was assigned to the corresponding dimer  $|H_2(1/4)|_2$ .

(The equations of the molecular hydrino rotational energy calculations herein of the form (#.#) and the referenced sections not contained within this section correspond to those of MILLS GUT, https://brilliantlightpower.com/book-download-and-streaming/). In the case of hydrogen-type molecules and molecular dimers, the moment of inertia may be measured using rotational energy spectroscopy such as Raman spectroscopy, and using the known nuclear masses, the moment of inertia gives the nuclear separation. Specifically, for a diatomic molecule having atoms of masses  $m_1$  and  $m_2$ , the moment of inertia is (Eq. (12.66)):

$$I = \mu r^2 \tag{1}$$

where  $\mu$  is the reduced mass given by (Eq. (12.67)):

$$\mu = \frac{m_1 m_2}{m_1 + m_2} \tag{2}$$

and where r is the distance between the centers of the atoms, the internuclear distance. The parameter r also regards the molecular separation distance of hydrogen-type dimers. The

rotational energy levels follow from Eq. (1.71) and are given by (Eq. (12.68)):

$$E_{rotational \ orbital} = \frac{\hbar^2}{2I} J(J+1) \tag{3}$$

where J is an integer. The radiation of a multipole of order  $(\ell, m_{\ell})$  carries  $m\hbar$  units of the z component of angular momentum per photon of energy  $\hbar\omega$ . Thus, the z component of the angular momentum of the corresponding excited rotational state is (Eq. (12.69)):

$$L_z = m\hbar \tag{4}$$

Thus, the selection rule for dipole and quadrupole rotational transitions are (Eq. (12.70)):

$$\Delta J = \pm 1 \tag{5}$$

And

$$\Delta J = \pm 2 \tag{6}$$

For example, the energy difference  $\Delta E$  for a transition of a molecular rotational state with a quantum number J to one with a quantum number of J + 1 is (Eq. (12.71))::

$$\Delta E = E_{J+1} - E_J = \frac{\hbar^2}{I} \left[ J + 1 \right] \tag{7}$$

The calculated internuclear distances and rotational energies for  $H_2$ ,  $D_2$ ,  $H_2^+$ , and  $D_2^+$  are given in Table 11.1 with the experimental values wherein the agreement is excellent. Similarly, the calculated parameters for molecular hydrogen dimers  $[H_2]_2$  and  $[D_2]_2$  given in the Geometrical Parameters and Energies Due to the Intermolecular van Der Waals Cohesive Energies of H<sub>2</sub> Dimer, Solid H<sub>2</sub>, H<sub>2</sub>(1/p) Dimer, and Solid H<sub>2</sub>(1/p) section and the End-Over-End Rotation of Hydrogen-Type Molecular Dimers section also match the experimental observations.

From the measured  $\Delta E$  (Eq. (7)), the moment of inertia can be determined, and then the separation may be determined using Eqs. (1) and (2). For individual molecules, the moment of inertia regards the nuclei separated by the internuclear axis wherein rotation is about a perpendicular axis. For end-over-end rotation, the dimer moment of inertia is given by summation of the moments of inertia for the two sets of nuclei, each equidistant from the center of rotation along the x-axis. In either case, the moment of inertia can be converted into the internuclear and molecular separations along the internuclear or intermolecular axis using the known masses of the nuclei. The rotational energies of hydrogen type molecules are given by (Eq. (12.77)):

$$\Delta E_{J \to J+1} = E_{J+1} - E_J = \frac{p^2 \hbar^2}{0.5m_p (7.411X10^{-11}m)^2} [J+1]$$

$$= p^2 [J+1] 0.01511 \ eV = [J+1] p^2 121.89 \ cm^{-1}$$
(8)

wherein the integer-squared dependence is due to the reciprocal integer dependence of the internuclear distance given by (Eq. (12.76)):

$$2c' = \frac{0.7411}{p} \,\mathring{A} \tag{9}$$

For example, the rotational energy of  $H_2(1/4)$  is four squared or 16 times that of  $H_2$  due to the internuclear distance being one fourth that of  $H_2$  (Eq. (9)). Similarly, the bond distance between molecular centers of  $[H_2(1/4)]_2$  (Eq. (16.202)) is very close to one fourth that of  $[H_2]_2$  (Eq. (16.181)) wherein the bonding in both cases is due to the corresponding van der Waals forces. The corresponding rotation energy that is dependent on the  $H_2(1/4)$  internuclear distance of  $0.35010a_0$  and the dimer molecular separation of  $1.94251a_0$  is (Eq. (16.262)):

$$\Delta E \left( \left[ H_2 \left( 1/4 \right) \right]_2 \right) = E_{J+1} - E_J = \frac{\hbar^2}{I \left( \left[ H_2 \left( 1/4 \right) \right]_2 \right)} \left[ J+1 \right]$$

$$= \frac{\hbar^2}{\frac{m_p}{2} \left[ \left( 1.94251a_0 \right)^2 + \left( 1.94251a_0 + 0.35010a_0 \right)^2 \right]}$$

$$= \left[ J+1 \right] 44.45 \ cm^{-1}$$
(10)

Conversely to determining separations from the rotational energy, the Raman-spectroscopydetermined  $\Delta E$  given by Eqs. (8) and (10) can be compared to the observed rotational energy to confirm the separation distances of molecular hydrino and molecular hydrino dimers.

Raman samples comprising solid web-like fibers were prepared by wire detonation of an ultrahigh purity Fe wire in a rectangular cuboid Plexiglas chamber having a length of 46 cm and a width and height of 12.7 cm. A 10.2 cm long, 0.25 mm diameter Fe metal wire (99.995%, Alfa Aesar #10937-G1) was mounted between two Mo poles with Mo nuts at a distance of 9 cm from the chamber floor, a 15 kV capacitor (Westinghouse model 5PH349001AAA, 55  $\mu$ F) was charged to about 4.5 kV corresponding to 557 J by a 35 kV DC power supply, and a 12 V switch with a triggered spark gap switch (Information Unlimited, model-Trigatron10, 3 kJ) was used to close the circuit from the capacitor to the metal wire inside of the chamber to detonate the wire. The detonation chamber contained air comprising 20 Torr of water vapor controlled by a humidifier and a water vapor sensor. The water vapor served as a source of HOH catalyst and atomic H to form molecular hydrino  $H_2(1/4)$ . The high voltage DC power supply was turned off before closing the trigger switch. The peak voltage of about 4.5 kV was discharged as a damped harmonic oscillator over about 300  $\mu$ s at a peak current of 5 kA. Web-like fibers formed in about 3-10 minutes after the wire detonation. Analytical samples were collected from the chamber floor and walls, as well as on a Si wafer placed in the chamber. Raman spectra were recorded on the web material using the Horiba Jobin Yvon LabRAM Aramis Raman spectrometer with a HeCd 325 nm laser in microscope mode with a magnification of 40X or with a 785 nm laser.

In addition, FeOOH (99+%, Alfa Aesar#A1626730) was used as a hydrino solid fuel to form solid Raman samples. FeOOH can form HOH catalyst and atomic hydrogen upon chemical decomposition by heating and was confirmed to produce excess heat. Similar hydrino solid fuels showed analytical signatures of the predicted hydrino product  $H_2(1/4)$  [R. Mills, J. Lotoski, W. Good, J. He, "Solid Fuels that Form HOH Catalyst," Int'l J. Hydrogen Energy, Vol. 39 (2014), pp. 11930–11944 DOI: 10.1016/j.ijhydene.2014.05.170.]. Ball milling has an effect equivalent to heating to drive chemical reactions [T. Markmaitree, R. Ren, L. L. Shaw, "Enhancement of lithium amide to lithium imide transition via mechanical activation", J. Phys. Chem. B., Vol. 110, No. 41, pp. 20710-20718]; so, FeOOH was ball milled in a stainless steel chamber using stainless steel balls for 10 hours. Raman spectra were recorded on the solid product using the 785 nm laser.

The Raman spectra obtained using a Horiba Jobin Yvon LabRam ARAMIS spectrometer with a 785 nm laser on solid web-like fibers prepared by wire detonation of an ultrahigh purity Fe wire in air maintained with 20 Torr of water vapor are shown in FIGURES 1-4. As shown in the 1490 cm<sup>-1</sup> to 2050 cm<sup>-1</sup> Raman spectral region (FIGURE 1), a broad negative peak was observed at 1947 cm<sup>-1</sup> with broad positive peaks on the red and blue wavenumber sides of the negative peak spanning the 1865 cm<sup>-1</sup> to 2010 cm<sup>-1</sup> region (145 cm<sup>-1</sup> span). The negative peak is assigned to the excitation of the J = 0 to J = 1 rotational transition of  $H_2(1/4)$  having a theoretical energy in vacuum of 1950 cm<sup>-1</sup> (Eq. (8) with p = 4). This is not an allowed Raman transition, but the effect of the polarization of the Fe<sub>2</sub>O<sub>3</sub> matrix may lift the selection rule. Consider that the recorded data of a Stokes spectrum comprises the scattered radiation having a continuum of energies of the incident 1.58 eV phonon minus that transferred to the sample excitations, and the lost energy is displayed. The rotational energy of the forbidden J = 0 to J = 1 transition is 0.24 eV, so a direct scattering will result in emission at 1.58 eV - 0.24 eV = 1.34 eV. The observed negative peak assigned to  $H_2(1/4)P_0(1)$  at about 0.24 eV requires a diminution of the corresponding scattered 1.24 eV light. There must has a high background of continuum 0.24 eV  $\pm 145/2$  cm<sup>-1</sup> avaited layed

1.34 eV light. There must be a high background of continuum 0.24 eV  $\pm 145/2$  cm<sup>-1</sup> excited levels wherein the hydrino rotation is excited indirectly by the background. Then, the concerted background excitation and the J = 0 to J = 1 transition is stimulated by the laser and possibly enhanced by the matrix or surface wherein the forbidden J = 0 to J = 1 transition becomes allowed. These inverse-Raman effect (IRE) and surface-enhanced IRE mechanisms cause a diminution of the scattered 1.34 eV radiation since the background excitation and coupled hydrino excitation results in emission at  $1.58 \ eV - \left[ 0.24 \ eV \left( background \right) + 0.24 \ eV \left( J = 0 \ to \ J = 1 \ transition \right) \right] = 1.1 \ eV \ radiation$ . The negative peak may be observed at slightly higher energy due to the losses in energy transfer to excite the  $H_2(1/4)$  rotation. FIGURE 1. The Raman spectrum (1490 cm<sup>-1</sup> to 2050 cm<sup>-1</sup>) obtained using a Horiba Jobin Yvon LabRam ARAMIS spectrometer with a 785 nm laser on solid web-like fibers prepared by wire detonation of an ultrahigh purity Fe wire in air maintained with 20 Torr of water vapor showing a single J = 0 to J = 1 rotational transition of  $H_2(1/4)$  involving a matrix enhanced inverse Raman effect (IRE).



As shown in the 3420 cm<sup>-1</sup> to 4850 cm<sup>-1</sup> Raman spectral region (FIGURE 2), a rotational series of peaks was observed. The energies of the peaks are given in TABLE 1. The maximum separation is 44 cm<sup>-1</sup> which matches the predicted rotational energy of  $\left[H_2(1/4)\right]_2$  given by Eq. (10). The peak separation decreases with energy as expected due to centrifugal distortion. An intensity and peak width discontinuity is observed at 3850 cm<sup>-1</sup>, that matches the double transition J = 0 to J = 1 for both of the  $H_2(1/4)$  molecules of the dimer. Another transition of the same energy that satisfies the selection rule of Eq. (6) for both molecules is  $[H_2(1/4)]_2 S_0(3) + S_0(0)$ . However, this transition would require one member of the dimer being in a high-energy rotational state. Using Eqs. (8), and (10), the position at 3850 cm<sup>-1</sup> matches the predicted energy of 3900.5 cm<sup>-1</sup> minus the dimer rotational energy of 44 cm<sup>-1</sup>. Thus, the position of the discontinuity of peaks is assigned to the transition  $\left[H_2(1/4)\right]_2 P_0(1) + P_0(1)$ . The blue side corresponding to the angular momentum quantum number change of  $\Delta i = +1$  is assigned to the transitions  $\left[H_2(1/4)\right]_2 P_0(1) + P_0(1) + \Sigma l(\Delta j = +1)$ . And, the red side corresponding to the angular momentum number change of  $\Delta j = -1$ is assigned to quantum the transitions  $\left[H_2(1/4)\right]_2 P_0(1) + P_0(1) + \Sigma l(\Delta j = -1)$ . Due to the extraordinary high energies, the transitions cannot be assigned to any prior known compound. The series of peaks assigned to  $\left[H_2(1/4)\right]_2$ rotation was confirmed to originate from the sample by treating the Fe<sub>2</sub>O<sub>3</sub>:  $H_2(1/4)$  sample with HCl. As shown in FIGURE 3, all of the Raman peaks and features were eliminated by the acid treatment.

FIGURE 2. The Raman spectrum (3420 cm<sup>-1</sup> to 4850 cm<sup>-1</sup>) obtained using a Horiba Jobin Yvon LabRam ARAMIS spectrometer with a 785 nm laser on solid web-like fibers (Fe web) prepared by wire detonation of an ultrahigh purity Fe wire in air maintained with 20 Torr of water vapor showing a series of rotational transitions of  $\left[H_2(1/4)\right]_2$  combined with a J = 0 to J = 1 rotational transition of both  $H_2(1/4)$  members of the dimer.



FIGURE 3. The Raman spectrum (3420 cm<sup>-1</sup> to 4850 cm<sup>-1</sup>) obtained using a Horiba Jobin Yvon LabRam ARAMIS spectrometer with a 785 nm laser showing that all of the Raman peaks of FIGURE 2 were eliminated by the acid treatment of the Fe<sub>2</sub>O<sub>3</sub>:  $H_2(1/4)$  sample with HCl.



TABLE 1. The position, energies, and energy separations of the 785 nm Raman spectral peaks in the 3420 cm<sup>-1</sup> to 4850 cm<sup>-1</sup> region assigned to the transitions  $[H_2(1/4)]_2 P_0(1) + P_0(1) + \Sigma l(\Delta j = \pm 1)$  (FIGURE 2).

Peak Number	Energy (cm <sup>-1</sup> )	Energy (eV)	$\Delta$ Energy	$\Delta$ Energy (eV)
1	3887	0.482	<u>(cm)</u> <u>43</u>	0.00533
2	3930	0.487	44	0.00546
3	3974	0.493	43	0.00533
4	4017	0.498	41	0.00508
5	4058	0.503	40	0.00496
6	4098	0.508	43	0.00533
7	4141	0.513	40	0.00496
8	4181	0.518	40	0.00496
9	4221	0.523	39	0.00484
10	4260	0.528	40	0.00496
11	4300	0.533	39	0.00484
12	4339	0.538	38	0.00471
13	4377	0.543	36	0.00446
14	4413	0.547	36	0.00446
15	4449	0.552	36	0.00446
16	4485	0.556	35	0.00434
17	4520	0.560	32	0.00397
18	4552	0.564	30	0.00372
19	4582	0.568	33	0.00409
20	4615	0.572	30	0.00372
21	4645	0.576	27	0.00335
22	4672	0.579	26	0.00322
23	4698	0.582	25	0.00310
24	4723	0.586	23	0.00285
25	4746	0.588	20	0.00248
26	4766	0.591	16	0.00198
27	4782	0.593	21	0.00260
28	4803	0.595	14	0.00174
29	4817	0.597		

As shown in the 6000 cm<sup>-1</sup> to 7600 cm<sup>-1</sup> Raman spectral region (FIGURE 4), a strong single Raman transition was observed at 6366 cm<sup>-1</sup> which is assigned to the allowed  $\left[H_2(1/4)\right]_2 S_0(2)$ 

transition of  $H_2(1/4)$  corresponding to the state change J = 0 to J = 2 (Eqs. (6), (3) and (8)). The second order 785 nm laser line is observed at 6375 cm<sup>-1</sup>. The 6366 cm<sup>-1</sup> peak was confirmed to be from the sample by ultraviolet irradiation. The 6366 cm<sup>-1</sup>, 785 nm Raman peak was also observed in other compounds comprising  $H_2(1/4)$  such as the white polymeric compound formed by dissolving Ga<sub>2</sub>O<sub>3</sub> collected from a hydrino reaction run in the SunCell® in aqueous KOH, allowing fibers to grow, and float to the surface where they were collected by filtration.  $H_2(1/4)$ was identified by X-ray photoelectron spectroscopy, electron paramagnetic resonance spectroscopy, electron beam emission spectroscopy, and Raman spectroscopy [131]. The latter spectrum is shown in FIGURE 5, and the Raman spectrum recorded after ultraviolet irradiation is shown in FIGURE 6. The peak assigned to  $H_2(1/4)$  was eliminated by the UV irradiation.

FIGURE 4. The 785 nm, Fe-web Raman spectrum (6000 cm<sup>-1</sup> to 7600 cm<sup>-1</sup>) showing a single J = 0 to J = 2 rotational transition of  $H_2(1/4)$  and a series of rotational transitions of  $\left[H_2(1/4)\right]_2$  combined with a J = 0 to J = 2 rotational transition of one  $H_2(1/4)$  member of the dimer.



FIGURE 5. The Raman spectrum (6360 cm<sup>-1</sup> to 6390 cm<sup>-1</sup>) obtained using a Horiba Jobin Yvon LabRam ARAMIS spectrometer with a 785 nm laser on the white polymeric compound (WPC) formed by dissolving Ga<sub>2</sub>O<sub>3</sub> collected from a hydrino reaction run in the SunCell® in aqueous KOH, allowing fibers to grow, and float to the surface where they were collected by filtration.



FIGURE 6. The 785 nm-laser Raman spectrum (6360 cm<sup>-1</sup> to 6390 cm<sup>-1</sup>) recorded the WPC after ultraviolet irradiation showing the elimination of the  $\left[H_2(1/4)\right]_2 S_0(2)$  peak at 6366 cm<sup>-1</sup>.



A sharp spectral discontinuity was observed starting at 6410 cm<sup>-1</sup>, 44 cm<sup>-1</sup> higher energy than the energy of the  $[H_2(1/4)]_2 S_0(2)$  transition, that matches the rotation energy of  $[H_2(1/4)]_2$  given by Eq. (10). The dimer rotational series of peaks was also observed for the angular momentum quantum number change of  $\Delta j = +1$ . The discontinuity and series of peaks is

assigned to  $\left[H_2(1/4)\right]_2 S_0(2) + \Sigma l(\Delta j = +1)$  corresponding to the combined rotations of  $\left[H_2(1/4)\right]_2$  and the  $\left[H_2(1/4)\right]_2 S_0(2)$  transition of  $H_2(1/4)$ .

The Raman spectra obtained using a Horiba Jobin Yvon LabRam ARAMIS spectrometer with a 785 nm laser on solid FeOOH powder prepared by ball milling for 10 hours are shown in FIGURES 7-9. As shown in the 3500 cm<sup>-1</sup> to 8000 cm<sup>-1</sup> Raman spectral region (FIGURE 7), the transitions  $[H_2(1/4)]_2 P_0(1) + P_0(1),$ assigned to series peaks the of  $\left[H_{2}(1/4)\right]_{2}P_{0}(1)+P_{0}(1)+\Sigma l\left(\Delta j=-1\right), \left[H_{2}(1/4)\right]_{2}P_{0}(1)+P_{0}(1)+\Sigma l\left(\Delta j=+1\right), \left[H_{2}(1/4)\right]_{2}S_{0}(2)+\frac{1}{2}S_{0$ , and  $\left[H_2(1/4)\right]_2 S_0(2) + \Sigma l(\Delta j = +1)$  were also observed in the Fe<sub>2</sub>O<sub>3</sub>:  $H_2(1/4)$  product of FeOOH ball milling. The 3420 cm<sup>-1</sup> to 4850 cm<sup>-1</sup> Raman spectral region (FIGURE 8) shows the the  $\left[H_2(1/4)\right]_2 P_0(1) + P_0(1), \quad \left[H_2(1/4)\right]_2 P_0(1) + P_0(1) + \Sigma l(\Delta j = -1),$ details of and  $\left[H_2(1/4)\right]_2 P_0(1) + P_0(1) + \Sigma l(\Delta j = +1)$  transitions. And, the 6000 cm<sup>-1</sup> to 7600 cm<sup>-1</sup> Raman spectral region (FIGURE 9) shows the details of the  $[H_2(1/4)]_2 S_0(2)$ and  $\left[H_2(1/4)\right]_2 S_0(2) + \Sigma l(\Delta j = +1)$  transitions. In the latter case, the sharp spectral discontinuity was observed starting at 6410 cm<sup>-1</sup>, 44 cm<sup>-1</sup> higher energy than the energy of the  $\left[H_2(1/4)\right]_2 S_0(2)$ transition at 6366 cm<sup>-1</sup>. The peak positions and energies are shown in TABLE 2. The maximum separation is 44 cm<sup>-1</sup> which matches the predicted rotational energy given by Eq. (10). The peak separation decreases with energy as expected due to centrifugal distortion.

FIGURE 7. The Raman spectrum (3500 cm<sup>-1</sup> to 8000 cm<sup>-1</sup>) obtained using a Horiba Jobin Yvon LabRam ARAMIS spectrometer with a 785 nm laser on solid FeOOH powder prepared by ball milling for 10 hours showing that the series of peaks assigned to the transitions  $\begin{bmatrix} H_2(1/4) \end{bmatrix}_2 P_0(1) + P_0(1)$ ,  $\begin{bmatrix} H_2(1/4) \end{bmatrix}_2 P_0(1) + P_0(1) + \Sigma l(\Delta j = -1)$ ,  $\begin{bmatrix} H_2(1/4) \end{bmatrix}_2 P_0(1) + P_0(1) + \Sigma l(\Delta j = +1)$ ,  $\begin{bmatrix} H_2(1/4) \end{bmatrix}_2 S_0(2)$ , and  $\begin{bmatrix} H_2(1/4) \end{bmatrix}_2 S_0(2) + \Sigma l(\Delta j = +1)$ were also observed in the Fe<sub>2</sub>O<sub>3</sub>:  $H_2(1/4)$  product of FeOOH ball milling.



FIGURE 8. The Raman spectrum ) (3420 cm<sup>-1</sup> to 4850 cm<sup>-1</sup>) obtained on solid Fe<sub>2</sub>O<sub>3</sub>:  $H_2(1/4)$  showing the details of the  $[H_2(1/4)]_2 P_0(1) + P_0(1)$ ,  $[H_2(1/4)]_2 P_0(1) + P_0(1) + \Sigma l(\Delta j = -1)$ , and  $[H_2(1/4)]_2 P_0(1) + P_0(1) + \Sigma l(\Delta j = +1)$  transitions.



FIGURE 9. The Raman spectrum (6000 cm<sup>-1</sup> to 7600 cm<sup>-1</sup>) obtained on solid Fe<sub>2</sub>O<sub>3</sub>:  $H_2(1/4)$  showing the details of the  $[H_2(1/4)]_2 S_0(2)$  and  $[H_2(1/4)]_2 S_0(2) + \Sigma l(\Delta j = +1)$  transitions.



TABLE 2. The position, energies, and energy separations of the 785 nm Raman spectra peaks in the 6000 cm<sup>-1</sup> to 7600 cm<sup>-1</sup> region assigned to the transitions  $\left[H_2(1/4)\right]_2 S_0(2) + \Sigma l(\Delta j + 1)$  (FIGURE 9).

Peak Number	Energy (cm <sup>-1</sup> )	Energy (eV)	$\Delta$ Energy (cm <sup>-1</sup> )	$\Delta$ Energy (eV)
1	6418	0.796	44	0.00546
2	6462	0.801	45	0.00558
3	6507	0.807	41	0.00508
4	6548	0.812	29	0.00360
5	6577	0.815	29	0.00360
6	6606	0.819	26	0.00322
7	6632	0.822	28	0.00347
8	6660	0.826	29	0.00360
9	6689	0.829	30	0.00372
10	6719	0.833	28	0.00347
11	6747	0.836	32	0.00397
12	6779	0.840	30	0.00372
13	6809	0.844	31	0.00384
14	6840	0.848	33	0.00409
15	6873	0.852	30	0.00372
16	6903	0.856	27	0.00335
17	6930	0.859	26	0.00322
18	6956	0.862	27	0.00335

19	6983	0.866	30	0.00372
20	7013	0.869	27	0.00335
21	7040	0.873	27	0.00335
22	7067	0.876	24	0.00298
23	7091	0.879	28	0.00347
24	7119	0.883	25	0.00310
25	7144	0.886	25	0.00310
26	7169	0.889	24	0.00298
27	7193	0.892	23	0.00285
28	7216	0.895	24	0.00298
29	7240	0.898	24	0.00298
30	7264	0.901	21	0.00260
31	7285	0.903	23	0.00285
32	7308	0.906	20	0.00248
33	7328	0.909	19	0.00236
34	7347	0.911	19	0.00236
35	7366	0.913	17	0.00211
36	7383	0.915	17	0.00211
37	7400	0.917	15	0.00186
38	7415	0.919	13	0.00161
39	7428	0.921	12	0.00149
40	7440	0.922		

The Raman spectra obtained using a Horiba Jobin Yvon LabRam ARAMIS spectrometer with a 325 nm laser on solid web-like fibers prepared by wire detonation of an ultrahigh purity Fe wire in 20 Torr water vapor-air atmosphere and on FeOOH powder prepared by ball milling for 10 hours are shown in FIGURES 10 and 11, respectively. In both cases, a series of 1000 cm<sup>-1</sup> separated peaks were observed in the 17,000 to 22,000 cm<sup>-1</sup> region. The peaks are similar to the second order ro-vibration fluorescence emission of  $H_2(1/4)$  observed in the Raman spectrum (FIGURE 12) recoded on NaOH:KCl (1:1 wt%) getter of the  $H_2(1/4)$  product gas from high current ignition of hydrated silver shots wherein the spectrum was recorded using the Horiba Jobin Yvon LabRam ARAMIS spectrometer with a 325 nm laser [R. Mills, Y. Lu, R. Frazer, "Power Determination and Hydrino Product Characterization of Ultra-low Field Ignition of Hydrated Silver Shots", Chinese Journal of Physics, Vol. 56, (2018), pp. 1667-1717.]. The corresponding first order spectrum in the ultraviolet comprised a series of 0.24 eV evenly spaced peaks centered at 260 nm called the 260 nm e-beam band recoded with a EUV spectrometer on emission from high-energy electron beam bombardment of solid getter materials that trapped  $H_2(1/4)$  [R. Mills, X Yu, Y. Lu, G Chu, J. He, J. Lotoski, "Catalyst induced hydrino transition (CIHT) electrochemical cell," (2012), Int. J. Energy Res., (2013), DOI: 10.1002/er.3142]. The 2-fold reduction in the peak spacing in the spectra of FIGURES 10 and 11 indicates that the source of the spectra is the second order ro-vibrational emission of the dimer  $\left[H_2(1/4)\right]_2$  wherein the reduced mass in Eqs. (1-3) and (8) increases by a factor of 2. The corresponding vibration energy that is proportional to the reciprocal square root of the reduced mass (Eq. (11.148)) is predicted to be 0.707 times that of  $H_2(1/4)$ .

FIGURE 10. The Raman spectrum obtained using a Horiba Jobin Yvon LabRam ARAMIS spectrometer with a 325 nm laser on solid web-like fibers prepared by wire detonation of an ultrahigh purity Fe wire in 20 Torr water vapor-air atmosphere showing the matrix-shifted second order ro-vibration fluorescence emission of  $\left[H_2(1/4)\right]_2$ .



FIGURE 11. The Raman spectrum obtained using a Horiba Jobin Yvon LabRam ARAMIS spectrometer with a 325 nm laser on FeOOH powder prepared by ball milling for 10 hours showing the matrix-shifted second order ro-vibration fluorescence emission of  $\left[H_2(1/4)\right]_2$ .



FIGURE 12. The second order ro-vibration fluorescence emission of  $H_2(1/4)$  observed in the Raman spectrum recoded on NaOH:KCl (1:1 wt%) getter of the  $H_2(1/4)$  product gas from high current ignition of hydrated silver shots wherein the spectrum was recorded using the Horiba Jobin Yvon LabRam ARAMIS spectrometer with a 325 nm laser.



The  $H_2(1/4)$  theoretical rotational transition energies  $E_{rot}H_2(1/4)$  at the matrix shifted  $v=1 \rightarrow v=0$  vibrational energy given previously [R. Mills, Y. Lu, R. Frazer, "Power Determination and Hydrino Product Characterization of Ultra-low Field Ignition of Hydrated Silver Shots", Chinese Journal of Physics, Vol. 56, (2018), pp. 1667-1717] were calculated using the 260 nm peak formula that was converted to the second order energies by multiplication by a factor of 0.5:

$$E_{rot}H_2(1/4) = \left[30769\ cm^{-1} - 0.5((0,\pm1)1950(J+1)\ cm^{-1}\ +\ 2(30769\ cm^{-1} - 13,236\ cm^{-1}))\right] (11)$$

wherein 325 nm corresponds to an energy of 30769 cm<sup>-1</sup> and the theoretical peak energies were subtracted from the Raman 325 nm laser energy to convert the second order fluorescence spectrum

into a Raman spectrum, the lattice-shifted vibrational energy was given by the energy of the Q(0) peak (13,236 cm<sup>-1</sup>), and 0, ±1 correspond to Q, P, and R branches. The corresponding fluorescence or photoluminescence energy  $E_{rot}^{PL}H_2(1/4)$  follows from Eq. (11):

$$E_{rot}^{PL}H_2(1/4) = \left[ (0,\pm 1)1950(J+1) \ cm^{-1} + 2(30769 \ cm^{-1} - 13,236 \ cm^{-1}) \right]$$
(12)

Comparison of the theoretical transition energies (Eq. (11)) and transition assignments with the observed Raman peaks are given in TABLE 3. The first order fluorescence energies (Eq. (12)) are also given in TABLE 3.

Conversely, the spectra of FIGURES 10 and 11 were converted into the experimental first order of ro-vibrational energies  $E_{rot} \left[ H_2(1/4) \right]_2$  of  $\left[ H_2(1/4) \right]_2$  at the corresponding matrix shifted  $v = 1 \rightarrow v = 0$  vibrational energy using the formula:

$$E_{rot1st order} \left[ H_2 \left( 1/4 \right) \right]_2 = 2 \left[ 30769 \ cm^{-1} - Raman \ peak \ \left( cm^{-1} \right) \right]$$
(13)

The observed peak positions and the results of conversion of the 325 nm Raman spectrum on the Fe web-like fibers (FIGURE 10) into the equivalent of a first order fluorescence spectrum (Eq. (13)) are given in TABLE 4. Comparing the results in TABLE 4 to those of  $H_2(1/4)$  in TABLE 3 demonstrates that the average energy spacing of the former peaks is almost exactly <sup>1</sup>/<sub>2</sub> that of the latter, and the energy of the Q(0) peak of the spectrum of TABLE 4 is shifted by a factor of  $\frac{1}{\sqrt{2}}$ 

relative to that of TABLE 3. The shifts that match theoretical predictions support the assignment of the spectra of FIGURES 10 and 11 to the second order fluorescence of  $\left[H_2(1/4)\right]_2$ . The close agreement between theory and the spectra further confirms the  $\left[H_2(1/4)\right]_2$  assignment of the spectra recorded with the 785 nm laser (FIGURES 1-4, 7-9).

TABLE 3. Comparison of the theoretical transition energies (Eq. (11)) and transition assignments with the observed Raman peaks, the first order fluorescent or photoluminescence (PL) energies (Eq. (12)), and the energy and wavenumber separation of contiguous peaks. [R. Mills, Y. Lu, R. Frazer, "Power Determination and Hydrino Product Characterization of Ultra-low Field Ignition of Hydrated Silver Shots", Chinese Journal of Physics, Vol. 56, (2018), pp. 1667-1717].

Assignment	Observed	Calculated	Calculated	Calculated	$\Delta$ Energy	$\Delta$ Energy
	Energy	2nd Order	1st Order	1st Order	(cm⁻¹)	(eV)
	(cm⁻¹)	PL Energy	PL Energy	PL Energy		
		(cm⁻¹)	(cm⁻¹)	(eV)		
P(4)	17130	17136	27266	3.380	1950	0.242
P(3)	16157	16161	29216	3.622	1950	0.242
P(2)	15183	15186	31166	3.864	1950	0.242
P(1)	14210	14211	33116	4.106	1950	0.242
Q(0)	13236	13236	35066	4.347	1950	0.242
R(0)	12262	12261	37016	4.589	1950	0.242
R(1)	11289	11286	38966	4.831	1950	0.242
R(2)	10315	10311	40916	5.073		
				Average	<b>1950</b>	0.242

TABLE 4. Conversion of the peak positions of the spectra of FIGURES 10 and 11 into second order and then equivalent first order fluorescent or photoluminescence (PL) energies (Eq. (13)), and the energy and wavenumber separation of contiguous peaks.

Assignment	Observed Peak Wavenumber (cm <sup>-1</sup> )	2nd Order PL Energy (cm <sup>-1</sup> )	1st Order PL Energy (cm <sup>-1</sup> )	1st Order PL Energy (eV)	$\Delta$ Energy (cm $^{-1}$ )	∆ Energy (eV)
P(7)	21470	9299	18598	2.306	1084	0.134
P(6)	20928	9841	19682	2.440	760	0.094
P(5)	20548	10221	20442	2.534	806	0.100
P(4)	20145	10624	21248	2.634	800	0.099
P(3)	19745	11024	22048	2.734	850	0.105
P(2)	19320	11449	22898	2.839	906	0.112
P(1)	18867	11902	23804	2.951	906	0.112
Q(0)	18414	12355	24710	3.064	954	0.118
R(0)	17937	12832	25664	3.182	1168	0.145
R(1)	17353	13416	26832	3.327		
				Average	915	0.113

To further confirm the ro-vibrational assignment of  $\left[H_2(1/4)\right]_2$ , the Horiba Jobin Yvon LabRam ARAMIS spectrometer with a 442 nm laser was used to perform Raman spectroscopy on a sample from an CIHT electrochemical cell reported previously [R. Mills, X Yu, Y. Lu, G Chu, J. He, J. Lotoski, "Catalyst induced hydrino transition (CIHT) electrochemical cell," (2012), Int. J. Energy Res., (2013), DOI: 10.1002/er.3142] that supported the reaction to form hydrinos. The CIHT electrochemical cell comprised (i) a molten LiOH-LiBr eutectic salt mixture that served as a OH<sup>-</sup> conductor, (ii) a hydrogen-supplying anode that oxidized OH<sup>-</sup> to OH that subsequently reacted with H to form HOH catalyst wherein the H supplied at the anode also reacted with HOH catalyst to form  $H_2(1/4)$ , and (iii) a cathode that was supplied with H<sub>2</sub>O and O<sub>2</sub> that were reduced to OH<sup>-</sup>. During operation, gases from the electrochemical cell passed through a KOH getter. The sample showed a very intense 260 nm e-beam band recoded with a EUV spectrometer on emission from high-energy electron beam bombardment of the solid KOH getter sample. With the presence of  $H_2(1/4)$  confirmed, Raman analysis was further performed. As shown in Figure 13, a broad feature centered at 4233 cm<sup>-1</sup> was assigned to a series of rotational transitions of  $\left[H_2(1/4)\right]_2$ combined with a J = 0 to J = 1 rotational transition of both  $H_2(1/4)$  members of the dimer. The corresponding designation in Figure 13 is  $[H_2(1/4)]_2 P_0(1) + P_0(1) + \Sigma l(\Delta j = \pm 1)$ . The J = 0 to J = 2 rotational transition of  $H_2(1/4)$  was observed at 6288 cm<sup>-1</sup>. The corresponding designation in Figure 13 is  $[H_2(1/4)]_2 S_0(2)$ . The J=0 to J=2 rotational transition of one  $H_2(1/4)$ member combined with a J = 0 to J = 1 rotational transition of the other  $H_2(1/4)$  member of the dimer was observed as a split peak at 8256 cm<sup>-1</sup> and 8226 cm<sup>-1</sup> wherein the separation was assigned to a rotational transition of  $\left[H_2(1/4)\right]_2$ . The corresponding designation in Figure 13 is  $\left[H_{2}(1/4)\right]_{2}S_{0}(3) + P_{0}(1) + l(\Delta j = +1).$ 

FIGURE 13. The Raman spectrum obtained using a Horiba Jobin Yvon LabRam ARAMIS spectrometer with a 442 nm laser on a KOH getter that absorbed  $H_2(1/4)$  gas from a CIHT cell showing the  $[H_2(1/4)]_2 P_0(1) + P_0(1) + \Sigma l(\Delta j = \pm 1), [H_2(1/4)]_2 S_0(2),$  and the  $[H_2(1/4)]_2 S_0(3) + P_0(1) + l(\Delta j = +1)$  transitions.



The dimer  $\left[H_2(1/4)\right]_2$ , was further identified by gas chromatography of gas collected from the SunCell® [113].  $H_2(1/4)$  gas was collected from the SunCell® using a valved microchamber connected to the vacuum line and cooled to 15 K by a cryopump system (Helix Corp., CTI-Cryogenics Model SC compressor; TRI-Research Model T-2000D-IEEE controller; Helix Corp., CTI-Cryogenics model 22 cryodyne). The liquefied gas was warmed to room temperature to achieve 10 Torr chamber pressure and was injected into an HP 5890 Series II gas chromatograph with a capillary column (Agilent molecular sieve 5 Å, (50 m x 0.32 mm, df = 30  $\mu$ m) at 303 K (30 °C), argon carrier gas, and a thermal conductivity detector (TCD) at 60 °C.  $H_2(1/4)$  was observed at 7.17 minutes and hydrogen that co-condensed with  $H_2$  gas was observed at 10.68 minutes (FIGURE 14). The collected gas contained no helium, the peak was negative with a helium carrier gas indicating that the sample gas had a higher thermal conductivity, and the migration rate was faster than that of helium with an argon carrier gas. No known gas has a faster migration rate and high thermal conductivity than  $H_2$  or He which is characteristic of and identifies hydrino since it has a much greater mean free path due to exemplary  $H_2(1/4)$  having 64 times smaller volume and 16 times smaller ballistic cross section. Hydrogen condensed under pressure and temperature conditions that violate the Clausius Clapeyron equation due to the raising of the  $H_2$  liquefaction temperature by co-condensation with  $H_2(1/4)$ .

FIGURE 14. The gas chromatograph of gas collected from the SunCell® by a cryopump showing  $H_2(1/4)$  at 7.17 minutes and hydrogen that co-condensed with  $H_2$  gas was observed at 10.68 minutes. Due to its small size,  $H_2(1/4)$  has a faster migration rate than any gas previosly known. Hydrogen condensed under pressure and temperature conditions that violate the Clausius Clapeyron equation due to the raising of the  $H_2$  liquefaction temperature by co-condensation with  $H_2(1/4)$ .



 $H_2(1/4)$  comprises an unpaired electron, and spin isomer states were observed by electron paramagnetic resonance spectroscopy (EPR). Specially, the  $H_2(1/4)$  EPR spectrum comprises (i) a doublet at the g-factor of 2.0046386 (Eq. (16.218)) with members separated by spin-orbital splitting energy  $E_{S/O}$  (Eq. (16.223)) wherein each member is sub-split by the integer fluxon energy  $\Delta B_{\Phi}$  (Eq. (16.232)), (ii) a singlet at the g-factor of 2.0057983 (Eq. (16.233)), (iii) a doublet at the g-factor of 2.0057983 (Eq. (16.233)) with members separated by spin nuclear splitting energy  $E_{S/N}$  (Eq. (16.238)) wherein the down field and upfield members each have a series of satellite peaks with a member separation of  $E_{S/O}$  (Eq. (16.223)) and each satellite peak has an integer fluxon energy sub-splitting of  $\Delta B_{\Phi}$  (Eqs. (16.248-16.251). Evidence of spin isomers of  $H_2(1/4)$ was sought by gas chromatography. When the collection and sample heating from cryogenic temperature were manipulated, it was observed that the early gas chromatographic peak for  $H_2(1/4)$  split into a plurality of peaks as shown in FIGURES 15-16. These peaks are assigned to spin isomers of  $H_2(1/4)$  akin to the ortho and para spin isomers of  $H_2$  that are observed by gas chromatography under cryogenic conditions.

FIGURE 15. The gas chromatograph of gas collected from the SunCell<sup>®</sup> by a cryopump showing multiple  $H_2(1/4)$  peaks assigned to spin isomers.



FIGURE 16. The gas chromatograph of gas collected from the SunCell<sup>®</sup> by a cryopump showing multiple  $H_2(1/4)$  peaks assigned to spin isomers.



Upon the collected SunCell® gas standing at cryogenic temperature, it was observed that a peak developed that had a migration time slightly greater than that of hydrogen and about twice the retention time of the  $H_2(1/4)$  peak that occurs before the hydrogen peak (FIGURE 17). The peak was assigned to the dimer  $[H_2(1/4)]_2$ . This result provides insight into the mechanism for the formation of molecular hydrino dimers in the Fe web-like fibers and FeOOH. In both cases, iron oxide can serve as a matrix to capture  $H_2(1/4)$  gas. Since  $H_2(1/4)$  is magnetic due to an unpaired electron and the iron oxides such as Fe<sub>2</sub>O<sub>3</sub> are magnetic due to a plurality of unpaired

electrons, cooperative magnetic ordering may facilitate dimer formation even at elevated temperatures in the magnetized lattices as shown by Raman spectroscopy (FIGURES 1-4, 7-9).

FIGURE 17. The gas chromatograph of gas collected from the SunCell® by a cryopump showing a peak just after the  $H_2$  peak assigned to  $\left[H_2(1/4)\right]_2$ .



The  $H_2(1/4)$  spin isomer peaks (FIGURE 18) and the  $[H_2(1/4)]_2$  peak (FIGURES 19-20) were also observed from gallium oxide comprising molecular hydrino, Ga<sub>2</sub>O<sub>3</sub>:H<sub>2</sub>(1/4), collected from the SunCell® and heated to 850°C in a vacuum-tight stainless-steel pressure vessel to cause the release of gas which was introduced into the gas chromatograph with a six-way valve. These results further confirm the EPR results assigned to  $H_2(1/4)$  that were performed on samples purified from the same starting material.

FIGURE 18. The gas chromatograph of hydrino gas evolved from gallium oxide comprising molecular hydrino, Ga<sub>2</sub>O<sub>3</sub>:H<sub>2</sub>(1/4), collected from the SunCell® and heated to 850°C in a vacuum tight stainless-steel pressure vessel to cause the release of gas which was introduced into the gas chromatograph.  $H_2(1/4)$  spin isomer peaks were observed before the  $H_2$  peak.



FIGURE 19. The gas chromatograph of hydrino gas evolved from a Ga<sub>2</sub>O<sub>3</sub>:H<sub>2</sub>(1/4) sample that showed  $H_2(1/4)$  a spin isomer peak before the  $H_2$  peak.



FIGURE 20. The gas chromatograph of hydrino gas evolved from a Ga<sub>2</sub>O<sub>3</sub>:H<sub>2</sub>(1/4) sample showing a peak just after the  $H_2$  peak assigned to  $\left[H_2(1/4)\right]_2$ .

