



Technical Note

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Boulder Laboratories

DIELECTRIC CONSTANT OF
LIQUID PARAHYDROGEN

BY R. J. CORRUCCINI



U. S. DEPARTMENT OF COMMERCE
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ABSTRACT

It is shown that the available data on the dielectric constant of hydrogen conform to the Clausius-Mossotti equation within the probable experimental errors. The published data cover temperatures from the triple point ($\sim 14^\circ\text{K}$) to well above room temperature and a thousandfold range of densities. Using an average value of the specific polarization, tables of the dielectric constant of the liquid have been computed for temperatures from the triple point (13.803°K) to 32°K (58°R) and pressures from saturation to 340 atmospheres.

DIELECTRIC CONSTANT OF LIQUID PARAHYDROGEN

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Introduction Capacitance measurements are used in liquid hydrogen technology to determine liquid level and apparent density. As will be shown, the dielectric constant is represented very well as a function of density by the well known Clausius-Mossotti equation. With the recent publication of extensive experimental data on the density and specific volume of parahydrogen¹ it has now become possible to compute the dielectric constant for a wide range of conditions. This note presents tables of dielectric constant as a function of pressure up to 340 atm (5000 psia) and as a function of temperature from the triple point to near the critical point. The basis of these calculations will now be presented.

The Clausius-Mossotti equation can be written

$$\frac{\epsilon - 1}{\epsilon + 2} v = p$$

in which

- ϵ is the dielectric constant
- v is the specific volume
- p is the specific polarization, a property of the substance having the same dimensions as v .

To the extent that p is a constant for a given substance, the substance may be said to obey the Clausius-Mossotti equation. The equation is derived from classical electrostatic theory on the assumptions that the molecules are non-polar and spherical, there are no short-range interactions between them, and their spatial distribution is isotropic^{2, 3}. Hydrogen satisfies only the first assumption. However, errors due to the other assumptions tend to vanish as the density increases⁴ and are negligible for non-polar gases at ordinary pressures. More complete theories have been developed^{4, 17, 18} that are valid up to moderate densities, but none that are generally accurate at densities characteristic of liquids.

Thus, in applying the equation to liquid hydrogen, deviations certainly are to be expected due to violation of all of the assumptions previously stated except the first. However, it is found that all of the available data can be represented within the probable experimental

errors by a single value of the polarization, even though they cover a thousandfold range of densities. Now the available experimental data will be examined.

Liquid hydrogen. Of five known references, three are from Leiden. A reading of the most recent of the three (Werner and Keesom, 1925⁵) and examination of the scatter of the various ϵ and p values shows clearly that the earlier two (by Breit and Onnes, 1924 and Wolfke and Onnes, 1924) now should be given no weight. Measurements by Guillien⁶ appear to be comparable in precision and accuracy to those of Werner and Keesom. Both of these papers cover the range, 14-20.4°K, and the authors conclude that their data show no consistent variations in p that could be interpreted as deviations from the Clausius-Mossotti equation. To these should be added a value at the boiling point by van Itterbeek and Spaepen⁷ of apparently comparable reliability. Mean values of p obtained in these investigations are given in table 1.

In addition it is possible to compute the dielectric constant from refractive index data using the relation, $\epsilon = n_{\infty}^2$, where n_{∞} is the refractive index extrapolated to very long wavelengths. Johns and Wilhelm⁸ have measured the refractive index of liquid hydrogen from 14° to 20.4°K at wavelengths in the visible region. Guillien⁶ has calculated the value, $n_{\infty}^2 = 1.2288 \pm .0003$ at 20.4°, from their data. The corresponding value of p calculated using the specific volume of normal hydrogen⁹ is shown in table 1.

Solid hydrogen. The data of Werner and Keesom⁵ disagree markedly with those of Guillien.⁶ From Guillien's work and similar experiences at this laboratory in measuring the dielectric constant of liquid and solid nitrogen,¹⁰ it is clear that discrepancies of this kind readily occur due to the difficulty of ensuring that the solid, which has to be cast in place in the measuring condenser, is free from voids. In order to obtain a meaningful test of the Clausius-Mossotti equation using a solid, either extreme care must be taken with the preparation of the sample, or else its effective density must be determined in place by an independent method.

Gaseous hydrogen. There have been many direct measurements of the dielectric constant of the gas; most of them near ambient temperature and pressure. However, Maryott and Buckley¹¹ in providing a "best" value at 20°C and 1 atmosphere have

based it entirely on optical and microwave measurements of the refractive index using the relation, $\epsilon = n_{\infty}^2$. Maryott and Buckley dismiss the direct measurements of ϵ with this statement: "In a majority of cases, values of the dielectric constant measured at radio frequencies do not appear to be of sufficient accuracy to provide useful information for reference purposes." A value of p calculated from Maryott and Buckley's recommended dielectric constant using the specific volume of the real gas⁹ is presented in table 1.

Of special interest to this inquiry are those few studies of the dielectric constant that cover wide ranges of temperature, pressure and density. The extremes of temperature are represented by the work of van Itterbeek and Spaepen¹² (20° - 291°K) and Forro¹³ (295° - 571°K). Each of these authors obtained constant values of p within their experimental limits of accuracy. A more rigorous test is provided by the data of Michels, Sanders and Schipper¹⁴ at 25° and 100°C and at pressures from 1 to 1425 atm. The maximum density achieved equals that of saturated liquid at 26°K. In spite of the great range of densities covered by Michels et al, the polarization showed no discernible trend with density or temperature. Their average value is included in table 1.

Summary of experimental results. The following table summarizes the experimental results that are regarded as especially significant. As indicated, some of the tolerances attached to the mean values of polarization are based on estimates of accuracy by the original authors. Others are guesses by the present author. In most cases, the stated tolerance is approximately equal to the mean deviation of several determinations. None of the investigators referred to the matter of ortho-para composition, and so it is assumed that the samples were normal hydrogen in all cases.

Inasmuch as none of the researches showed internal evidence of dependence of p upon the experimental conditions, the variations in p among the various investigations shown in table 1 cannot be construed as evidence for deviations from the Clausius-Mossotti equation. Parts of the discrepancies may be attributed to the use of different data for the specific volumes and to accidental variations in ortho-para composition. However, the discrepancies are too large to be fully accounted for in these ways, and in large part must be charged to undisclosed systematic errors in the experiments.

Table 1
Specific Polarization of Hydrogen

Reference	State	Temperature °K	Approx. Range of Specific Volume cm ³ /g.	p cm ³ /g.
Werner & Keesom ⁵ 1925	Liq.	14 - 20.4	13-14	1.010 ±.002**
Guillien ⁶ 1940	Liq.	14 - 20.4	13-14	0.9984±.002**
Van Itterbeek & Spaepen ⁷ 1942	Liq.	20.4	14	0.988 ±.003**
Johns & Wilhelm ^{8,6} 1937	Liq.	20.4	14	*0.9986±.001**
Maryott & Buckley ¹¹ 1953	Gas	~293	12,000	*1.0104±.001
Michels, et al ¹⁴ 1953	Gas	298, 373	16-11,000	1.001 ±.002

*From measurements of refractive index.

**Based on estimate of accuracy by original authors

The degree of constancy of polarization that has been demonstrated for hydrogen is rather exceptional and raises the question of how well the Clausius-Mossotti equation might fit other cryogenic fluids. Böttcher²(p. 209) attributes this result to hydrogen having an exceptionally small value of the imperfection parameter, α/a^3 , where a is the average molecular radius, and the "polarizability", α , is given by $P = Mp = 4/3 \pi N\alpha$, in which N is Avogadro's number, and M is the molecular weight. Conversely, the imperfection parameter is larger for carbon dioxide, and, accordingly, the polarization of this gas has been found to vary with pressure by about 2% in 150 atm. Values of these quantities for some cryogenic substances are shown in table 2. The trend of α/a^3 suggests that nitrogen and oxygen will show relative deviations from the Clausius-Mossotti equation intermediate between those of hydrogen and carbon dioxide, while helium should show even less deviation than hydrogen.

Table 2
Imperfection Parameters

Substance	P	$10^{24}\alpha$	* α	α/a^3
	cm ³ /mol	cm ³	Å	
He	0.51	0.20	1.28	0.10
H ₂	2.02	0.79	1.47	0.25
N ₂	4.3	1.71	1.85	0.27
O ₂	4.0	1.59	1.76	0.29
CO ₂	7.3	2.89	2.1	0.31

*Based on a collection¹⁵ of values determined from second virial coefficients and viscosities. Those determined from second virial coefficients were given greater weight.

Tables of dielectric constant. A value of p of 1.00 cm³/g (0.0160 ft³/lb) is seen to represent the experimental results listed in table 1 within about $\pm 1\%$. This value has been used to calculate tables 3-7 using the Clausius-Mossotti formula,

$$\epsilon = \frac{v + 2p}{v - p}$$

and recently published provisional values¹ of the specific volume of parahydrogen. The latter are estimated to be accurate within 0.2%¹⁶ except near the critical point, where the equation of state used to represent the data introduced systematic errors. The deviations due to the equation of state were eliminated from the few values of specific volume near the critical point before calculating the dielectric constant. Consequently, the error in ϵ is almost entirely due to the one percent uncertainty in p . The error in $(\epsilon - 1)$ from this source is also about one percent, while the error in ϵ is estimated to range from 0.15% at the lowest density to 0.25% at the highest. It should be noted that the densities at the higher pressures (below the stepped lines in tables 3 and 6) exceed the maximum density at which the dielectric constant has been experimentally measured by as much as 15%. The validity of the adopted value of p at these higher densities has not been established.

P atm	20	21	22	23	24	25	26	27	28	29	30	31	32
1	1.2297												
2	1.2302	1.2260	1.2216										
3	1.2306	1.2265	1.2221	1.2174	1.2122								
4	1.2311	1.2270	1.2227	1.2180	1.2129	1.2073	1.2010						
5	1.2315	1.2275	1.2233	1.2186	1.2136	1.2081	1.2020	1.1950					
6	1.2320	1.2280	1.2238	1.2192	1.2143	1.2089	1.2029	1.1962	1.1883				
7	1.2324	1.2285	1.2243	1.2198	1.2150	1.2097	1.2039	1.1973	1.1897	1.1805			
8	1.2329	1.2290	1.2249	1.2204	1.2157	1.2105	1.2048	1.1984	1.1911	1.1824			
9	1.2333	1.2295	1.2254	1.2210	1.2163	1.2112	1.2056	1.1994	1.1924	1.1842	1.1734		
10	1.2337	1.2300	1.2259	1.2216	1.2169	1.2119	1.2065	1.2004	1.1936	1.1857	1.1758	1.1621	
15	1.2358	1.2322	1.2284	1.2243	1.2200	1.2153	1.2103	1.2049	1.1990	1.1924	1.1847	1.1758	1.1645
20	1.2378	1.2343	1.2307	1.2268	1.2227	1.2184	1.2137	1.2088	1.2034	1.1976	1.1913	1.1839	1.1757
25	1.2396	1.2363	1.2328	1.2291	1.2253	1.2211	1.2168	1.2122	1.2073	1.2021	1.1964	1.1903	1.1832
30	1.2414	1.2382	1.2349	1.2313	1.2276	1.2237	1.2196	1.2153	1.2107	1.2059	1.2008	1.1952	1.1891
35	1.2431	1.2400	1.2368	1.2334	1.2298	1.2261	1.2222	1.2181	1.2138	1.2093	1.2046	1.1995	1.1942
40	1.2448	1.2418	1.2386	1.2354	1.2319	1.2284	1.2246	1.2208	1.2167	1.2124	1.2080	1.2033	1.1984
45	1.2464	1.2434	1.2404	1.2372	1.2339	1.2305	1.2269	1.2232	1.2193	1.2153	1.2111	1.2067	1.2021
50	1.2479	1.2450	1.2421	1.2390	1.2358	1.2325	1.2291	1.2255	1.2218	1.2179	1.2139	1.2098	1.2055
60	1.2508	1.2481	1.2453	1.2424	1.2394	1.2363	1.2331	1.2297	1.2263	1.2227	1.2191	1.2153	1.2114
70	1.2535	1.2510	1.2483	1.2455	1.2427	1.2397	1.2367	1.2336	1.2303	1.2270	1.2236	1.2201	1.2165
80	1.2561	1.2536	1.2511	1.2484	1.2457	1.2429	1.2400	1.2371	1.2340	1.2309	1.2277	1.2244	1.2211
90	1.2585	1.2561	1.2537	1.2512	1.2486	1.2459	1.2431	1.2403	1.2374	1.2345	1.2315	1.2284	1.2252
100	1.2608	1.2586	1.2562	1.2538	1.2513	1.2487	1.2461	1.2434	1.2406	1.2378	1.2349	1.2320	1.2290
120	1.2652	1.2631	1.2609	1.2586	1.2563	1.2539	1.2514	1.2489	1.2464	1.2438	1.2412	1.2385	1.2357
140	1.2693	1.2672	1.2651	1.2630	1.2608	1.2586	1.2563	1.2540	1.2516	1.2492	1.2467	1.2442	1.2417
160	1.2730	1.2711	1.2691	1.2671	1.2650	1.2629	1.2607	1.2585	1.2563	1.2540	1.2517	1.2494	1.2470
180	1.2766	1.2747	1.2728	1.2709	1.2689	1.2669	1.2649	1.2628	1.2606	1.2585	1.2563	1.2541	1.2518
200	1.2799	1.2781	1.2763	1.2745	1.2726	1.2707	1.2687	1.2667	1.2647	1.2626	1.2605	1.2584	1.2563
220	1.2831	1.2814	1.2796	1.2779	1.2760	1.2742	1.2723	1.2704	1.2685	1.2665	1.2645	1.2625	1.2605
240		1.2845	1.2828	1.2811	1.2793	1.2775	1.2757	1.2739	1.2720	1.2701	1.2682	1.2663	1.2643
260		1.2874	1.2858	1.2841	1.2824	1.2807	1.2790	1.2772	1.2754	1.2736	1.2717	1.2699	1.2680
280		1.2886	1.2870	1.2853	1.2837	1.2821	1.2803	1.2786	1.2768	1.2751	1.2733	1.2714	
300		1.2914	1.2898	1.2882	1.2866	1.2850	1.2833	1.2817	1.2800	1.2782	1.2765	1.2747	
320			1.2925	1.2910	1.2894	1.2878	1.2862	1.2846	1.2829	1.2813	1.2796	1.2779	
340			1.2951	1.2936	1.2921	1.2905	1.2890	1.2874	1.2858	1.2842	1.2825	1.2809	

Note: Values below the stepped line represent an extrapolation of p with density.

Table 4. Dielectric Constant of Saturated Liquid Parahydrogen vs. Temperature ($^{\circ}\text{K}$)

T, $^{\circ}\text{K}$	ϵ	T, $^{\circ}\text{K}$	ϵ
13.803 *	1.2503	23	1.2168
14	1.2497	24	1.2119
15	1.2467	25	1.2067
16	1.2436	26	1.2011
17	1.2403	27	1.1949
18	1.2369	28	1.1880
19	1.2333	29	1.1802
20	1.2295	30	1.1710
20.268**	1.2285	31	1.1598
21	1.2255	32	1.1444
22	1.2213		

* Triple point

** Normal boiling point

Table 5. Dielectric Constant of Saturated Liquid Parahydrogen vs. Pressure (atm)

P, atm.	ϵ	P, atm.	ϵ
1	1.2285	6	1.1865
2	1.2175	7	1.1794
3	1.2087	8	1.1720
4	1.2010	9	1.1641
5	1.1937	10	1.1553

Table 6 Dielectric Constant of Liquid Parahydrogen vs. Temperature (°R) and Pressure (psia)

P psia	T, °R													
	36	38	40	42	44	46	48	50	52	54	56	58		
15	1.2294													
20	1.2296	1.2249												
30	1.2299	1.2253	1.2203											
40	1.2302	1.2257	1.2207	1.2152										
50	1.2305	1.2260	1.2211	1.2157	1.2097									
60	1.2308	1.2263	1.2215	1.2161	1.2102	1.2036								
70	1.2311	1.2267	1.2218	1.2166	1.2108	1.2043	1.1969							
80	1.2314	1.2271	1.2222	1.2170	1.2113	1.2050	1.1977							
90	1.2318	1.2274	1.2226	1.2175	1.2118	1.2056	1.1984	1.1901						
100	1.2321	1.2277	1.2229	1.2179	1.2123	1.2062	1.1992	1.1910	1.1811					
120	1.2327	1.2284	1.2237	1.2187	1.2132	1.2073	1.2005	1.1928	1.1836	1.1710				
140	1.2332	1.2290	1.2244	1.2195	1.2142	1.2083	1.2018	1.1945	1.1858	1.1745				
160	1.2338	1.2297	1.2251	1.2203	1.2151	1.2093	1.2030	1.1960	1.1877	1.1774	1.1635			
180	1.2344	1.2303	1.2259	1.2211	1.2159	1.2103	1.2043	1.1974	1.1896	1.1800	1.1678			
200	1.2349	1.2309	1.2265	1.2218	1.2168	1.2114	1.2055	1.1988	1.1913	1.1824	1.1714			
250	1.2363	1.2323	1.2281	1.2237	1.2189	1.2137	1.2081	1.2019	1.1951	1.1873	1.1782	1.1671		
300	1.2376	1.2338	1.2297	1.2254	1.2208	1.2158	1.2105	1.2048	1.1984	1.1915	1.1835	1.1742		
350	1.2389	1.2352	1.2312	1.2271	1.2226	1.2178	1.2128	1.2073	1.2014	1.1951	1.1879	1.1797		
400	1.2401	1.2365	1.2327	1.2286	1.2243	1.2197	1.2149	1.2097	1.2042	1.1982	1.1917	1.1843		
450	1.2413	1.2377	1.2340	1.2301	1.2259	1.2215	1.2169	1.2119	1.2067	1.2010	1.1950	1.1882		
500	1.2425	1.2390	1.2354	1.2315	1.2275	1.2233	1.2188	1.2140	1.2090	1.2036	1.1979	1.1918		
600	1.2448	1.2414	1.2380	1.2343	1.2304	1.2264	1.2222	1.2178	1.2131	1.2082	1.2031	1.1977		
700	1.2469	1.2437	1.2404	1.2368	1.2331	1.2293	1.2254	1.2212	1.2169	1.2124	1.2076	1.2026		
800	1.2489	1.2458	1.2426	1.2392	1.2358	1.2321	1.2284	1.2244	1.2203	1.2160	1.2116	1.2070		
900	1.2508	1.2478	1.2448	1.2415	1.2382	1.2346	1.2311	1.2273	1.2235	1.2194	1.2152	1.2109		
1000	1.2526	1.2497	1.2468	1.2437	1.2405	1.2370	1.2336	1.2300	1.2263	1.2225	1.2186	1.2145		
1250	1.2570	1.2544	1.2516	1.2487	1.2456	1.2425	1.2394	1.2361	1.2328	1.2293	1.2259	1.2222		
1500	1.2610	1.2585	1.2559	1.2532	1.2504	1.2476	1.2446	1.2416	1.2385	1.2353	1.2321	1.2288		
1750	1.2648	1.2623	1.2599	1.2573	1.2546	1.2520	1.2492	1.2464	1.2435	1.2406	1.2376	1.2345		
2000	1.2682	1.2659	1.2636	1.2612	1.2586	1.2561	1.2534	1.2508	1.2481	1.2454	1.2425	1.2396		
2500	1.2744	1.2724	1.2703	1.2680	1.2658	1.2635	1.2612	1.2588	1.2563	1.2537	1.2512	1.2487		
3000	1.2802	1.2783	1.2763	1.2743	1.2721	1.2700	1.2679	1.2656	1.2633	1.2610	1.2588	1.2564		
3500	1.2837	1.2837	1.2817	1.2799	1.2779	1.2759	1.2738	1.2718	1.2697	1.2676	1.2653	1.2632		
4000		1.2867	1.2851	1.2832	1.2812	1.2793	1.2775	1.2755	1.2735	1.2713	1.2693			
4500			1.2899	1.2881	1.2862	1.2844	1.2826	1.2807	1.2789	1.2769	1.2749			
5000			1.2943	1.2925	1.2909	1.2892	1.2874	1.2855	1.2839	1.2820	1.2802			

Note: Values below the stepped line represent an extrapolation of p with density.

Table 7.

Dielectric Constant of
Saturated Liquid Parahydrogen
vs. Pressure (psia)

P psia	T °R	ε
10	34.260	1.2329
15	36.603	1.2279
20	38.436	1.2238
30	41.291	1.2168
40	43.529	1.2108
50	45.400	1.2053
60	47.022	1.2001
70	48.464	1.1951
80	49.768	1.1902
90	50.962	1.1854
100	52.067	1.1806
120	54.062	1.1705
140	55.832	1.1594
160	57.424	1.1461
180	58.860	1.1257

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U. S. DEPARTMENT OF COMMERCE
Luther H. Hodges, *Secretary*

NATIONAL BUREAU OF STANDARDS
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THE NATIONAL BUREAU OF STANDARDS

The scope of activities of the National Bureau of Standards at its major laboratories in Washington, D.C., and Boulder, Colorado, is suggested in the following listing of the divisions and sections engaged in technical work. In general, each section carries out specialized research, development, and engineering in the field indicated by its title. A brief description of the activities, and of the resultant publications, appears on the inside of the front cover.

WASHINGTON, D.C.

Electricity. Resistance and Reactance. Electrochemistry. Electrical Instruments. Magnetic Measurements. Dielectrics. High Voltage.

Metrology. Photometry and Colorimetry. Refractometry. Photographic Research. Length. Engineering Metrology. Mass and Scale. Volumetry and Densimetry.

Heat. Temperature Physics. Heat Measurements. Cryogenic Physics. Equation of State. Statistical Physics.

Radiation Physics. X-ray. Radioactivity. Radiation Theory. High Energy Radiation. Radiological Equipment. Nucleonic Instrumentation. Neutron Physics.

Analytical and Inorganic Chemistry. Pure Substances. Spectrochemistry. Solution Chemistry. Standard Reference Materials. Applied Analytical Research.

Mechanics. Sound. Pressure and Vacuum. Fluid Mechanics. Engineering Mechanics. Rheology. Combustion Controls.

Organic and Fibrous Materials. Rubber. Textiles. Paper. Leather. Testing and Specifications. Polymer Structure. Plastics. Dental Research.

Metallurgy. Thermal Metallurgy. Chemical Metallurgy. Mechanical Metallurgy. Corrosion. Metal Physics. Electrolysis and Metal Deposition.

Mineral Products. Engineering Ceramics. Glass. Refractories. Enameled Metals. Crystal Growth. Physical Properties. Constitution and Microstructure.

Building Research. Structural Engineering. Fire Research. Mechanical Systems. Organic Building Materials. Codes and Safety Standards. Heat Transfer. Inorganic Building Materials.

Applied Mathematics. Numerical Analysis. Computation. Statistical Engineering. Mathematical Physics. Operations Research.

Data Processing Systems. Components and Techniques. Computer Technology. Measurements Automation. Engineering Applications. Systems Analysis.

Atomic Physics. Spectroscopy. Infrared Spectroscopy. Solid State Physics. Electron Physics. Atomic Physics.

Instrumentation. Engineering Electronics. Electron Devices. Electronic Instrumentation. Mechanical Instruments. Basic Instrumentation.

Physical Chemistry. Thermochemistry. Surface Chemistry. Organic Chemistry. Molecular Spectroscopy. Molecular Kinetics. Mass Spectrometry.

Office of Weights and Measures.

BOULDER, COLO.

Cryogenic Engineering. Cryogenic Equipment. Cryogenic Processes. Properties of Materials. Cryogenic Technical Services.

Ionosphere Research and Propagation. Low Frequency and Very Low Frequency Research. Ionosphere Research. Prediction Services. Sun-Earth Relationships. Field Engineering. Radio Warning Services. Vertical Soundings Research.

Radio Propagation Engineering. Data Reduction Instrumentation. Radio Noise. Tropospheric Measurements. Tropospheric Analysis. Propagation-Terrain Effects. Radio-Meteorology. Lower Atmosphere Physics.

Radio Standards. High Frequency Electrical Standards. Radio Broadcast Service. Radio and Microwave Materials. Atomic Frequency and Time Interval Standards. Electronic Calibration Center. Millimeter-Wave Research. Microwave Circuit Standards.

Radio Systems. Applied Electromagnetic Theory. High Frequency and Very High Frequency Research. Modulation Research. Antenna Research. Navigation Systems.

Upper Atmosphere and Space Physics. Upper Atmosphere and Plasma Physics. Ionosphere and Exosphere Scatter. Airglow and Aurora. Ionospheric Radio Astronomy.

