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THE ABSORPTION SPECTRUM OF HYDRATED AMERICIUM CHLORIDE

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May 2, 1952

Berkeley, California

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The study of the absorption spectra of the lanthanide and actinide elements is an important step in the correlation of these two groups since the spectra arise from transitions within the 4f and 5f electronic shells. Previous work in this laboratory has shown a similarity in the solution absorption spectra of americium and europium.[1] Freed and Leitz observed 17 lines in the spectrum of hydrated americium chloride.[2] The data reported here are more complete since a larger sample and a spectrograph of greater dispersion were used.

Approximately one mg. of americium (separated and purified by Dr. B. B. Cunningham) was available for growing crystals. The americium solution (6 N HCl) was extracted with ether to remove iron which interferes even in trace amounts. Sodium and ammonium chloride were removed to prevent dilution of the americium chloride crystals in an unknown manner.

Slow evaporation, the first crystal growing method tried, proved unsatisfactory since the product was an opaque powder completely useless for spectroscopy. The failure to grow clear crystals undoubtedly resulted from the decomposition of water by the intense alpha radiation (approximately $7 \cdot 10^9$ alphas $min.^{-1}$). To minimize the radiation chemistry effect, the solutions were evaporated rapidly under a heat lamp. The products obtained were conglomerate rather than single crystals, but were sufficiently clear

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for optical work. These preparations gradually grew opaque as a result of alpha decomposition of the water of crystallization, but they could be used for about two days.

The sample was mounted on a thin glass window which was then fastened to a brass container which had another window on the opposite side. This sample mounting was used for all measurements. Figure 1 shows the 77° K. optical arrangement; the Dewar flask is removed for the room temperature measurements. A Baird three meter grating spectrograph was used, and the spectrum was measured from 3600 A to 8500 A at both temperatures. The wave lengths, vacuum wave numbers, and relative intensities appear in the following table.

The most striking effect observed at low temperature is the greatly increased resolution. No densitometer measurements were made, but visual comparisons of similar exposures at the two temperatures revealed no marked alteration of relative intensities. This fact indicates that the population of the levels is not changed appreciably by lowering the temperature to 77° K.

A study was made of the frequency distribution of wave-number differences in an attempt to determine the energy difference between the $^7F_0$ ground state and possible low-lying excited levels. A number of constant intervals were observed varying from 20 cm^{-1} to 140 cm^{-1}, which probably correspond to vibrational levels reported for hydrated europium chloride.[3] Others were observed at 176, 288, 322 or 353, and 450 cm^{-1}. Since we have no temperature dependence data, no assignments can be made, and no distinction can be made between electronic and vibrational levels.

Table of Wavelengths, Intensities and Wave Numbers for $AmCl_3 \cdot xH_2O$ at Room Temperature and Liquid Nitrogen Temperature

	R.T.				N_2T		
No.	λ(A)	I	$\tilde{\nu}(cm^{-1})$	No.	λ (A)	I	$\tilde{\nu}(cm^{-1})$
1	8133	10	12,292	1	8180	10wh	12,222
				2	8159	10s	12,253
				3	8123	8h	12,307
				4	8108	8h	12,330
				5	8078	5	12,376
				6	8064	5	12,397
				7	8051	5	12,417
2	7993	8	12,508	8	8011	7	12,479
				9	7964	6	12,553
3	5168	10	19,346	10	5169	10	19,341
4	5163	10	19,364	11	5164	10	19,359
5	5154	7	19,397	12	5152	10	19,405
6	5139	5h	19,454	13	5137	10	19,461
				14	5131	4	19,484
				15	5119	8	19,529
				16	5110	8	19,564
7	5104	5h	19,587	17	5104	4	19,587
				18	5097	6	19,614
8	5084	8h	19,664	19	5090	5	19,641
				20	5074	5	19,703
				21	5067	8	19,730
9	5060	10h	19,757	22	5060	10	19,757
10	5047	10h	19,808	23	5051	10	19,793
				24	5045	10	19,816
				25	5038	10	19,844
				26	5010	5	19,955
11	4991	4	20,030	27	4996	4	20,010
				28	4988	4	20,043
12	4974	3	20,098	29	4972	3	20,107
13	4726	3	21,154				
14	4695	8s	21,293	30	4693	6	21,302
15	4646	6	21,517	31	4650	8	21,499
16	4639	6	21,550	32	4644	8	21,527
17	4596	5	21,752	33	4595	5	21,757
				34	4589	5	21,785
				35	4583	4	21,814
				36	4569	4	21,881
18	4555	10	21,948	37	4555	10	21,948
19	4434	5	22,547	38	4432	6	22,557
20	4315	5h	23,168	39	4318	7	23,152
21	4301	5h	23,244	40	4302	7	23,238
22	4285	5h	23,331	41	4292	5	23,293
				42	4283	7	23,342
				43	4270	5	23,413
23	4237	4	23,595	44	4239	4	23,534
24	4211	4	23,741				
25	4110	4	24,324				
26	4056	4	24,648				
27	4051	4	24,678				
				45	4032	5	24,795
				46	4027	5	24,825

No.	λ(A)	I	$\tilde{\nu}$ (cm^{-1})	No.	λ(A)	I	$\tilde{\nu}$(cm^{-1})
28	4023	8	24,850	47	4025	5	24,838
29	4006	6	24,956				
30	3960	10s	25,245	48	3963	6	25,226
				49	3961	6	25,239
31	3952	5	25,297	50	3953	4	25,290
				51	3948	4	25,322
32	3811	5s	26,232	52	3813	4	26,219
				53	3811	6	26,232
33	3801	5h	26,301	54	3801	10	26,301
				55	3797	10	26,329
				56	3787	7	26,399
34	3784	4	26,420	57	3784	9	26,420
35	3765	4	26,553	58	3767	5	26,539
				59	3764	5	26,560
				60	3751	4	26,652
				61	3733	5	26,780
36	3729	4	26,809	62	3728	4	26,816
				63	3722	5	26,860
				64	3704	4	26,990
37	3694	4	27,063				
				65	3667	4	27,262
38	3656	4	27,345	66	3662	4	27,300
				67	3644	4	27,435
				68	3631	5	27,533
39	3624	4	27,686	69	3626	6	27,571
				70	3619	6	27,624

s = sharp

w = wide

h = hazy

The failure to obtain a temperature effect presents two possibilities. First, assuming a Boltzman distribution, the lowest excited level must be less than the 300 cm^{-1} level reported for europium;[3] or second, a temperature of 77° K was not attained. The latter is a very real possibility since americium generates roughly 0.0015 cal min^{-1} mg^{-1}. Obviously this heat is not lost to the surroundings instantaneously and, even after a steady state heat flow to the surroundings is established, there will be a temperature gradient in the sample which will be a function of the physical dimensions and properties of the sample and surroundings. The possibility of anomalous temperature effects must not be overlooked in the interpretation of spectroscopic and magnetic data for radioactive elements.

We wish to thank Dr. B. B. Cunningham for the loan of the americium.

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[1] B. J. Stover, J. G. Conway, and B. B. Cunningham, J. Am. Chem. Soc. 73, 491 (1951).

[2] S. Freed and F. J. Leitz, J. Chem. Phys. 17, 540 (1949).

[3] F. H. Spedding, C. C. Moss, and R. C. Waller, J. Chem. Phys. 8, 908 (1940).

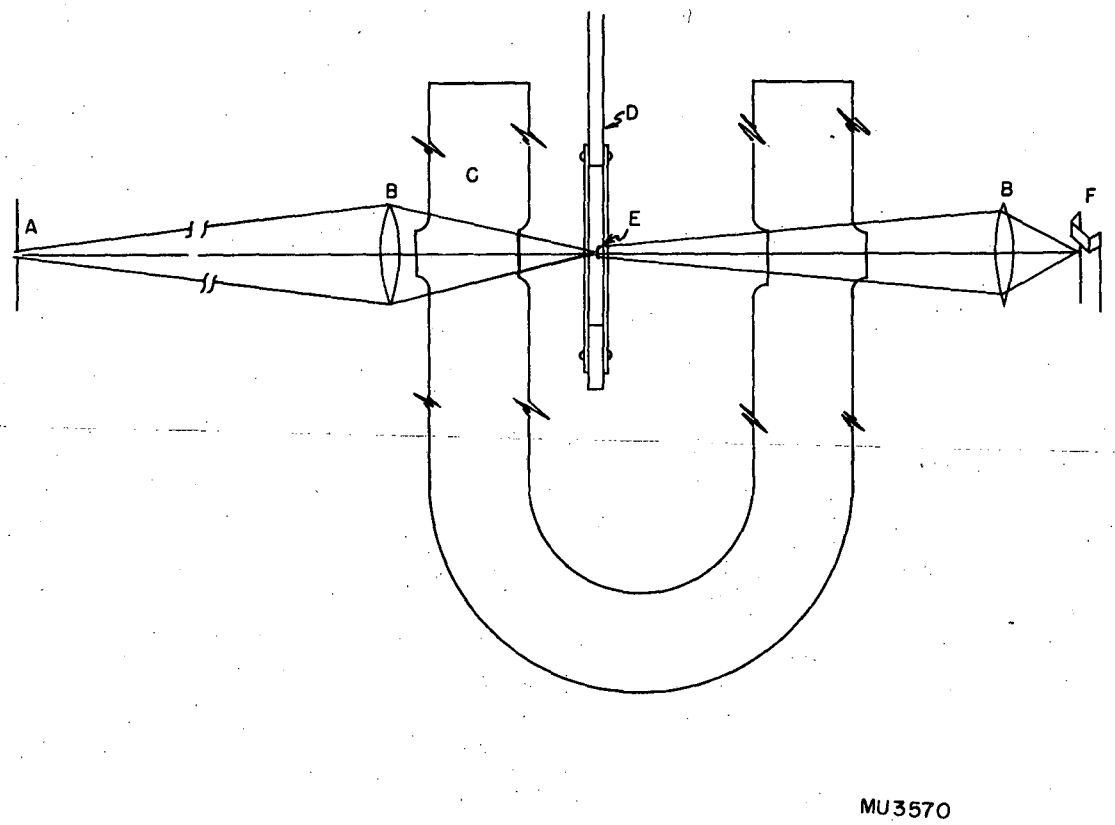

Fig. 1

Optical arrangement for obtaining the absorption spectra of $AmCl_3 \cdot H_2O$ at room temperature and liquid nitrogen temperature

a. Slit
b. Condensing lens
c. Dewar flask with optically flat windows
d. Sample holder
e. Position of sample
f. Tungsten filament lamp