ters, and G. Weinreich, Phys. Rev. Letters 7, 118 (1961).
[2]G. D. Boyd and J. P. Gordon, Bell System Tech. J. 40, 489 (1961).
[3]G. D. Boyd and H. Kogelnik, Bell System Tech. J. 41, 1347 (1962).
[4]H. Kogelnik and W. W. Rigrod, Proc. I. R. E. 50, 220 (1962).
[5]W. W. Rigrod, Appl. Phys. Letters 2, 51 (1963).
[6]J. A. Giordmaine, Phys. Rev. Letters 8, 19 (1962).
[7]D. A. Kleinman, Phys. Rev. 128, 1761 (1962).
[8]P. D. Maker, R. W. Terhune, M. Nisenoff, and C. M. Savage, Phys. Rev. Letters 8, 21 (1962).
[9]R. C. Miller and A. Savage, Phys. Rev. 128, 2175 (1962).
[10]N. Bloembergen, Symposium on Optical Masers, Polytechnic Institute of Brooklyn, Brooklyn, New York 16 April 1963 (to be published).
[11]R. W. Terhune, P. D. Maker, and C. M. Savage, Phys. Rev. Letters 8, 404 (1962).
[12]J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. 127, 1918 (1962); N. Bloembergen and P. S. Pershan, Phys. Rev. 128, 606 (1962).
[13]R. W. Terhune, P. D. Maker, and C. M. Savage, Appl. Phys. Letters 2, 54 (1963).
[14]W. W. Rigrod and A. J. Rustako, J. Appl. Phys. 34, 967 (1963).
[15]H. A. H. Boot and D. M. Clunie, Nature 197, 173 (1963).
[16]E. H. Byerly, J. Goldsmith, and W. H. McMahan, Proc. I. E. E. E. 51, 360 (1963).

ION PAIR ABSORPTION IN $PrCl_3$

G. H. Dieke and E. Dorman
The John Hopkins University, Baltimore, Maryland
(Received 16 May 1963)

In experiments by Versanyi[1] it was demonstrated that in $PrCl_3$ one photon can excite simultaneously two Pr^{3+} ions to different excited levels, one of which was always $^3P_0$. This was shown by the appearance of fluorescence from $^3P_0$ with monochromatic excitation at frequencies where a single ion could not possibly absorb but which numerically corresponded to the sum of different absorption frequencies of the single ion.

Naturally fluorescence can occur only if there is absorption at the exciting frequency. We have now been able to obtain with suitable crystals of pure $PrCl_3$ under the proper experimental arrangements the absorption lines corresponding to ion pair absorption. The region most suitable for observing these absorption lines is that beyond the $^3P_2$ lines, as we know that there can be no absorption lines of the Pr^{3+} ion between 22 220 and about 50 000 cm^{-1}.

It turns out that in the region between 22 220 and 28 100 cm^{-1} there are about 90 weak but very distinct absorption lines, the frequencies of almost all of which can be written as the sum of two ordinary absorption frequencies. In this region one is sure that there are no single-absorption lines, and occasional impurity lines can be recognized by their greater sharpness.

Table I gives the wave numbers and classifications of the observed lines in a representative interval. The intensities are estimates from microphotometer traces, such as shown in Fig. 1. The

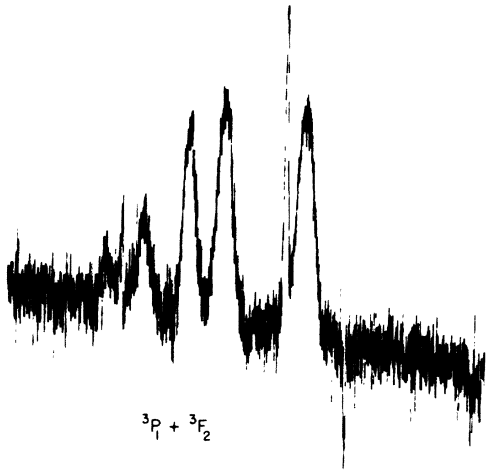

FIG. 1. Ion pair absorption lines of Pr^{3+} near 26 000 cm^{-1}. The sharp line is due to Nd^{3+}.

width of the lines is of the order of 6 cm^{-1}, considerably larger than that of the impurity lines. The lines are the sums of the two energy levels within the limits of experimental errors, which may here be more than 1 cm^{-1} for weak and crowded lines. The table shows that there are occasionally systematic deviations because the energy levels of concentrated $PrCl_3$, having to be derived from very broad lines, are not always accurately known.

Practically all sums of known levels which fall

Table I. Ion pair absorption lines of $PrCl_3$. All lines found in this interval are listed. The symbols have the following meanings: ν, observed wave number; I, estimated intensity; L_1,μ_1,L_2,μ_2, designation of level and Stark components of excited states of first and second ion, respectively; ν_{calc}, calculated wave number.

ν	I	L_1+L_2	μ_1,μ_2	ν_{calc}
25 368.5	30	P_0+F_2	0 ,1	68.7
381.5	5			
397.1	29	P_0+F_2	0 ,2	97.1
406.3	4			
410.8	10	P_0+F_2	0 ,0	10.6
960.0	34	P_1+F_2	1 ,1	60.2
967.1	9s[a]			
971.5	2	P_1+F_2	1 ,0[b]	70.2
977.5	1			
988.4	28	P_1+F_2	1 ,2	88.6
		P_1+F_2	0 ,1	90.14
26 001.7	20	P_1+F_2	1 ,0	02.1
017.3	10	P_1+F_2	0 ,2	18.6
030.3	3	P_1+F_2	0 ,0	32.1
116.2	2s[a]			
193.5	15	I_6+F_2	0 ,1	92.6
221.7	30	I_6+F_2	0 ,2	21.0
235.9	11	I_6+F_2	0 ,0	34.5
265.4	2			
277.2	1			
289.2	4	I_6+F_2	0',1	88.3
305.5	9	I_6+F_2	1 ,1	03.5
315.7	3	I_6+F_2	0',2	16.7
331.6	18	I_6+F_2	1 ,2	31.9
350.1	3			
374.4	13	D_2+G_4	0 ,2	73.8
407.7	8	P_2+H_6	0 ,3	03.9
732.0	9	P_0+F_3	0 ,0	31.9
746.9	18	P_0+F_3	0 ,2	48.7
753.0	7	P_0+F_3	0 ,3	54.3
797.6	10	P_0+F_3	0 ,1	97.8
812.7	2	P_0+F_3	0 ,3'	13.1
850.3	1			
27 099.7	8	P_2+F_2	0,1	00.2
108.4	2	P_2+F_2	0,0[b]	10.3
119.0	20	P_2+F_2	1,1	19.8
128.0	5	P_2+F_2	0,2	28.6
139.5	17	P_2+F_2	2,1	39.5
		P_2+F_2	0,0	42.2
147.2	17	P_2+F_2	1,2	48.3
159.5	13	P_2+F_2	1,0	61.8
165.2	11	P_2+F_2	2,2	67.9
179.4	5	P_2+F_2	2,0	81.5
194.1	4	P_0+F_4	0,3	97.7
214.9	8	P_0+F_4	0,x	16.4
230.9	12	P_0+F_4	0,1	32.2
250.3	18	P_0+F_4	0,2	50.5
321.7	17	P_1+F_3	1,0	23.4
337.9	20	P_1+F_3	1,2	40.2
344.6	4	P_1+F_3	1,3	45.7
368.0	2	P_1+F_3	0,2	70.2
374.8	5	P_1+F_3	0,3	75.7
389.3	8	P_1+F_3	1,1	89.3
405.0	3	P_1+F_3	1,3'	04.6
419.1	2	P_1+F_3	0,1	19.3
432.4	1	P_1+F_3	0,3'	34.6
573.1	2	I_6+F_3	0,2	72.6
620.1	2	I_6+F_3	0,1	21.2
667.3	8b	I_6+F_3	1,0	66.7
681.0	5	I_6+F_3	1,2	83.5
732.8	7	I_6+F_3	1,1	32.6
758.1	2	P_1+F_4	1,3'[b]	57.3
811.0	7	P_1+F_4	1,x	07.9
822.0	8	P_1+F_4	1,1	23.6
839.0	5	P_1+F_4	0,x	37.9
851.5	4	P_1+F_4	0,1	53.6

[a]Sharp impurity lines, chiefly Nd^{3+}.
[b]Absorption from first excited Stark component.

into this region are observed, in particular also the combinations of the components of $^3P_1$, $^3P_2$, and $^1I_6$ with the low-lying levels ^3H and ^3F, while in Varsanyi's fluorescence experiments there is only evidence for combinations with $^3P_0$. This seeming discrepancy is explained by the fact that excitation of $^3P_{1,2}$ and $^1I_6$ in concentrated $PrCl_3$ yields no or only very weak fluorescence, which is different from the situation in $PrCl_3$ highly diluted with $LaCl_3$, where, for instance, excitation of $^3P_2$ produces strong fluorescence from $^3P_0$. The ion pair absorption lines show no noticeable polarization.

The existence of the ion pair absorption lines is in agreement with nonlinear coupling between neighboring Pr^{3+} ions and the more formal theoretical considerations of Dexter.[2]

[1]F. Varsanyi and G. H. Dieke, Phys. Rev. Letters 7, 442 (1961).
[2]D. L. Dexter, Phys. Rev. 126, 1962 (1962).