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Low-Energy Level Structure of Neutral Cerium (Ce I)

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The low portion of an extensive level structure derived from analysis of the optical spectrum is reported. Positions, J values, and g_J factors are given for 98 levels, including all 91 levels expected below 10 000 cm^{-1}. A previous report on this analysis showed the ground level to be $4f5d6s^2\ {}^1G_4^\circ$ and gave the lowest levels of $4f5d^2 6s$. Comparison of the observed odd-parity levels with calculations by Goldschmidt and Salomon shows that all 86 of the odd levels tabulated here belong to these configurations. All but seven of these odd levels are assigned LS names, although the calculations show that many of them have low LS purities, and a few have strong mixtures of the two configurations. The much simpler system of even levels below 10 000 cm^{-1} includes only the six levels of $4f^2 6s^2\ {}^3H$ and ${}^3F$, beginning with ${}^3H_4$ at 4762.718 cm^{-1} above the ground-state level. The table of even levels also includes $4f^2 6s^2\ {}^1G_4$ and the lowest two levels of each of the lowest two terms of $4f^2 5d6s$, ${}^5I_{4,5}$, and ${}^5K_{5,6}$.

INTRODUCTION

One reason for special interest in the outer structure of the cerium atom is the rapid increase in the binding energy of the $4f$ electron through the sequence Ba–Pr ($Z = 56$–59). Well before any analysis of the arc spectrum of Ce existed, its complexity supported the deduction that at $Z = 58$ the $4f$ binding energy was about equal to that for $5d$ and $6s$. The ground configuration of La I ($Z = 57$) is $5d6s^2$, and $5d^2 6s$ is also very low. Thus after the discovery[1] in 1953 that $4f^3 6s^2$ was the ground-state configuration of Pr I ($Z = 59$), each of the configurations $4f^2 6s^2$, $4f5d6s^2$, and $4f5d^2 6s$ remained a reasonable possibility as lowest in Ce I. Racah's finding[2] that the $4f5d^2$, $4f5d6s$ configurations[3] extended lowest in Ce II made the Ce I $4f^2 6s^2$ possibility remote. An atomic-beam resonance measurement of three Ce I g_J factors differing by several percent from any expected LS-coupling values gave further evidence that the normal configuration had at least one $5d$ electron.[4]

In 1963 an analysis of the Ce I spectrum[5] gave energies, g_J values, LS designations, and configuration assignments for nine levels of $4f5d6s^2$ and for five levels of $4f5d^2 6s$. These were the lowest levels of the lowest two configurations, and the ground level was shown to be $4f5d6s^2\ {}^1G_4^\circ$. By 1967 the analysis had been greatly extended.[6] All levels were known to well above 10 000 cm^{-1}, including those of the three lowest $4f^2 6s^2$ terms. A theoretical interpretation of the low odd levels had also been accomplished.[7,8] Although only a small portion of the extended analysis can be given here, this report includes what are for several purposes the most important and interesting results.

OBSERVATIONS

The cerium-line list contains some 25 000 wavelengths.[9] These data and a description of the observations will eventually be published elsewhere. The most accurate wavelengths have been obtained in the region 3800–7100 Å from spectrograms taken in the eighth through the fifteenth orders of a plane-grating spectrograph.[10] An electrodeless lamp was the source. The probable wave-number error for averages from these plates is less than 0.01 cm^{-1}. The infrared spectrum has been observed out to 2.42 μm by Verges,[11] and was important in confirming the low even Ce I levels.

Extensions of the previously described Zeeman data[5] were obtained from infrared observations[12] to 9106 Å at a field of 24 000 G, and from observations in the visible region by Vander Sluis[13] with a

TABLE I. Odd-parity energy levels of Ce I to 10 000 cm^{-1}.

Configuration	Term	J	Level (cm^{-1})	g_J [a]	g_J [b]
$4f5d6s^2$	$^1G°$	4	0.000	0.9462	0.9454
$4f5d6s^2$	$^3F°$	2	228.849	0.7661	0.7651
		3	1663.120	1.077	1.0774
	c and d	4	3100.151	1.081	1.0770
$4f5d6s^2$	$^3H°$	4	1279.424	0.8889	0.8898
		5	2208.657	1.033	1.0321
		6	3976.104	1.157	1.1603
$4f5d6s^2$	$^3G°$	3	1388.941	0.7360	0.7349
	c and d	4	3312.240	1.083	1.0858
	c	5	4199.367	1.150	1.1502
$4f(5d^26s\ ^4F)$	$^5H°$	3	2369.068	0.5975	0.5998
	c	4	2437.629	0.9854	0.9859
	c and e	5	4417.618	1.179	1.1779
		6	4746.627	1.165	1.1659
		7	5802.108	1.237	
$4f5d6s^2$	$^1D°$	2	2378.827	0.937	0.9365
$4f(5d^26s\ ^4F)$	$^5I°$	4	3196.607	0.6668	0.6661
		5	3764.008	0.906	0.9069
		6	4455.756	1.118	1.1171
		7	5315.803	1.215	1.2162
		8	6809.128	1.250	
$4f(5d^26s\ ^4F)$	$^3G°$ c	5	3210.583	1.160	1.1628
	c	3	4160.283	0.730	0.7293
	c	4	4173.494	1.029	1.0295
$4f5d6s^2$	$^3D°$	1	3710.513	0.616	0.6155
		2	4766.323	1.153	1.1495
		3	5006.719	1.235	1.2367
$4f5d6s^2$ + $4f(5d^26s\ ^2D)$	$^3P°$ c	0	3974.503		
	c	1	4020.954	1.489	1.4940
	c	2	6303.984	1.419	
$4f(5d^26s\ ^4F)$	$^3S°$ c	1	5097.777	1.886	1.8826
$4f(5d^26s\ ^4F)$	$^5D°$ c	2	5210.906	1.225	1.2269
	c	3	5519.751	1.244	1.2453
	c	0	5571.156		
	c	4	5572.074	1.315	1.3166
	c and f	1	5637.233	1.389	
$4f(5d^26s\ ^4F)$	$^5G°$ c	2	5409.236	0.773	
		3	6234.792	1.047	1.0497
		4	6856.559	1.150	
		5	7467.160	1.179	
		6	8055.526	1.207	
$4f(5d^26s\ ^4F)$	$^5F°$	1	5674.829	0.140	
		2	5904.006	0.905	
	c	3	6337.061	1.232	
		4	7174.156	1.373	
		5	7933.558	1.345	
$4f(5d^26s\ ^2F)$	$^3H°$ c	4	6475.540	0.897	
$4f5d6s^2$	$^1F°$ c	3	6621.892	1.147	
$4f5d^26s$		5	6663.226	0.953	
$4f(5d^26s\ ^4F)$	$^3F°$ c	2	6836.628	0.683	0.6808
	c	3	7169.751	1.13	
	c	4	7890.429	1.242	
$4f(5d^26s\ ^2F)$	$^1G°$ c	4	7348.299	0.964	

TABLE I. (*continued*)

Configuration	Term	J	Level (cm^{-1})	g_J [a]	g_J [b]
$4f(5d^26s\ ^4F)$	$^3I^{\circ}$ [c]	6	7 696.210	1.076	
	[c]	5	7 715.236	0.934	
		7	8 587.973	1.155	
$4f5d^26s$		5	7 841.955	1.063	
$4f(5d^26s\ ^2D)$	$^1P^{\circ}$ [c]	1	7 853.119	0.983	
$4f(5d^26s\ ^4P)$	$^5G^{\circ}$	2	8 088.912	0.403	
		3	8 307.309	0.957	
		4	8 762.126	1.054	
	[c]	5	9 462.705	1.19	
		6	11 030.470	1.193	
$4f(5d^26s\ ^4F)$	$^5P^{\circ}$	2	8 101.187	1.735	
		3	8 270.249	1.504	
		1	8 430.846	2.04	
$4f(5d^26s\ ^2F)$	$^1S^{\circ}$	0	8 351.167		
$4f(5d^26s\ ^4F)$	$^3P^{\circ}$ [c]	2	8 366.098	1.525	
	[c]	1	8 695.201	1.285	
	[c]	0	9 119.094		
$4f5d^26s$		5	8 400.730	0.917	
$4f5d^26s$		4	8 509.209	0.954	
$4f5d^26s$		6	8 603.531	1.225	
$4f(5d^26s\ ^2F)$	$^1F^{\circ}$ [c]	3	8 902.306	1.128	
$4f5d^26s$		5	8 991.451	1.067	
$4f(5d^26s\ ^4P)$	$^3D^{\circ}$ [c]	3	9 135.099	1.274	
	[c]	2	9 425.529	1.207	
	[c]	1	9 903.122	0.813	
$4f(5d^26s\ ^2F)$	$^3D^{\circ}$ [c]	2	9 200.707	1.376	
$4f(5d^26s\ ^2F)$	$^3I^{\circ}$	6	9 333.222	1.047	
$4f5d^26s$		1	9 369.628	1.065	
$4f(5d^26s\ ^2F)$	$^3F^{\circ}$	2	9 709.012	0.799	
$4f5d^26s$	$^3G^{\circ}$ [c,g]	3	9 787.220	0.868	
$4f(5d^26s\ ^2G)$	$^1I^{\circ}$ [c]	6	9 830.608	1.081	
$4f(5d^26s\ ^4F)$	$^5S^{\circ}$	2	9 947.822	1.63	
$4f5d^26s$	$^3G^{\circ}$ [c,g]	3	9 996.647	(0.82) [h]	

[a] Values from optical Zeeman-effect observations (present work) except for level at 9996.6 cm^{-1}.

[b] Atomic-beam magnetic resonance values from Ref. 14, rounded off. The J value for the level at 4746 cm^{-1} and the optical g_J value for the 5006-cm^{-1} level as given in Table I of Ref. 14 are misprints.

[c] The indicated term assignment comprises less than 50% of the calculated composition of the level (Ref. 8).

[d] More than 50% of the composition of each of these two levels belongs to the combination $4f5d6s^2$ ($^3F^{\circ}+{}^3G^{\circ}$). The almost arbitrary name assignments are here reversed from those given tentatively in Ref. 5.

[e] More than 50% of the composition of this level is $^3G^{\circ}$ in character; this $^3G^{\circ}$ character is derived about equally, however, from $4f5d6s^2$ and $4f(5d^26s\ ^4F)$.

[f] The calculated $4f5d6s^2\ ^3P^{\circ}$ component for this level is slightly greater than the component for the assigned name.

[g] The levels with $J=3$ at 9787 and 9997 cm^{-1} each have greater than 50% $4f5d^26s\ ^3G^{\circ}$ character; the lower purity for each level results from a strong mixture of terms in its $5d^26s\ L'S'$ parentage. Parental designations for these levels are thus omitted here. It is also not yet certain which of the alternate ways of assigning these levels to the two calculated eigenvectors is correct.

[h] Calculated value from Ref. 8. The alternate calculated value (see Ref. g above) is 0.84.

field of 54 000 G. Zeeman patterns have now been measured for several thousand Ce I lines, but the reduction and compilation of these measurements have not been completed.

ANALYSIS

The analysis of Ce I has revealed it as a super-

TABLE II. Low even-parity energy levels of Ce I.

Configuration	Term	J	Level (cm^{-1})	g_J
$4f^26s^2$	^3H	4	4762.718	0.8051[a]
		5	6238.934	1.035
		6	7780.202	1.169
$4f^26s^2$	^3F	2	8235.605	0.680
		3	9206.305	1.083
		4	9379.148	1.139
$4f^26s^2$	^1G	4	11361.895	1.101
$4f^2(^3H)5d6s(^3D)$	^5I	4	12114.115	0.655
		5	12467.827	0.948[a]
$4f^2(^3H)5d6s(^3D)$ [b]	^5K	5	12366.834	0.710
		6	12960.950	0.90
$4f(5d6s6p\ ^4F^{\circ})$ [b] or $4f^2(^3H)5d6s(^3D)$ [b]	^5G	2	12992.115	0.388

[a]Rounded-off value from Ref. 14. The value from optical Zeeman-effect observations is 0.806 ± 0.002.

[b]The indicated order of coupling and parental LS designations are guesses.

position of two types of spectra. One of these, a dense spectrum of more than 20000 observed lines, results from transitions from upper even-parity levels to low odd levels. There are 85 odd levels lying below 10000 cm^{-1}, and all belong to the $4f5d6s^2$, $4f5d^26s$ group. These are given in Table I, along with one higher level needed to complete a term. Transitions from over 800 known upper even levels to the levels in Table I classify about 15000 lines, including the great majority of the strong lines.

A much less rich spectrum, probably no more than a tenth of the observed lines, is due to transitions terminating on low even levels. These include only six levels below 10000 cm^{-1}, belonging to the two lowest terms of $4f^26s^2$ (Table II). The range of Table II has been extended to include the $4f^26s^2\ ^1G$ term and the lowest levels of the next lowest even configuration $4f^25d6s$.

Most of the odd levels below 5000 cm^{-1} have from 200 to more than 300 classified transitions. Even though the level values in Table I were obtained at an earlier stage in the analysis, the averaging gave relative separations for several of these levels consistent to ± 0.001 cm^{-1}. The uncertainty for the least accurate odd levels and for the even levels in Table II is about ± 0.01 cm^{-1}.

The Zeeman effect is essential for the analysis of a spectrum of this complexity. Observed g_J values are given for all levels with $J \neq 0$ reported here except one. Childs and Goodman[14] have recently measured very accurately the g_J values of 33 Ce I levels by atomic-beam magnetic resonance. They based the correlation of their values to the Ce I levels on agreement with corresponding optical g_J values and on the intensities in the beam, no other characteristics of the levels being directly obtained from their measurements. The values of Childs and Goodman are included in Table I, rounded off to four decimal places. Comparison shows the corresponding optical values of g_J to be accurate to better than ± 0.002 on the average. Seven optical values that were thought to be especially well determined (given to four places in Table I) have an average accuracy of ± 0.0010. The higher levels usually have less accurate g_J values, and a few for which the probable error is ± 0.01 or more are given to only two places.

Goldschmidt and Salomon first calculated the odd levels of Ce I ($4f5d6s^2 + 4f5d^26s + 4f5d^3$) when only 22 odd levels were known. (The notation indicates that their calculations include configuration interactions.) These calculations, and later refinements fitting more experimental levels, were important guides in extending the analysis. The interpretation of levels of very mixed composition is not possible without reliable calculations, and some odd levels having high or low J values were found by specific searches around the predicted positions. By 1967 satisfactory agreement between the calculations[7] and observations had been achieved for most of the levels in Table I. Goldschmidt and Salomon have continued this work to give increasingly more accurate predictions.[8]

Comparison of these predictions with the full list of more than 200 observed odd levels (0–32000 cm^{-1}) shows that all expected levels have been found to well above 10000 cm^{-1}. Extension of the comparison to 15000 cm^{-1} gives ambiguities in the assignments of some levels for which g factors are not known. It is clear that the lowest level with a major component from $4f5d^3$ lies well above 10 000 cm^{-1}. No odd configuration other than the three included in the calculations need be considered in the range below 10 000 cm^{-1}.

Seventy-three of the 86 levels in Table I have been arranged to form 27 complete LS terms. Six of the remaining 13 levels are also assigned such names. The arrangement was guided by the calculated eigenvectors,[8] although only 39 of the 86 levels have a single $4f5d6s^2$ LS or $4f(5d^26s\ L'S')$ LS component greater than 50%. Some distinctions and comments are made in the table, and it should be clear that many of the designations are mainly a bookkeeping convenience. Interaction between the $4f5d6s^2$ and $4f5d^26s$ configurations contributes significantly to the impurities of a few levels, but most of the strongly mixed compositions are caused by deviations from LS coupling. The interested reader is referred to the work of Goldschmidt and Salomon[8] for quantitative compositions of the levels, including the seven levels of $4f5d^26s$ left undesignated in Table I.

The eight terms listed for $4f5d6s^2$ leave $^1H^\circ$ and $^1P^\circ$ "missing." The former was lost as a designation through contributing small components to a number of levels, while most of the $^1P^\circ$ composition is distributed among several levels above 12 000 cm^{-1}.

The 19 complete terms given for $4f5d^26s$ include all seven quintets based on $5d^26s$ ^4F, and four of the seven possible triplets having this parentage.

As is usual with the lowest few levels of $4f^N$ configurations, the $4f^26s^2$ $^3H^\circ$ levels in Table II belong to a rather good LS term and have g_J factors close to the LS values. The relative positions of all seven identified levels of $4f^26s^2$ are very similar to those of the corresponding levels of $4f^2$ in Ce III; it is thus unlikely that this low portion of Ce I $4f^26s^2$ is significantly perturbed by other low even configurations ($4f^25d6s$, $4f5d6s6p$, $4f^25d^2$) having no counterpart in Ce III.

However, the Ce I $4f^26s^2$ level and term separations are smaller than the corresponding separations in Ce III $4f^2$ by an average of about 7%. This decrease when the $6s$ electrons are present[15] is an order of magnitude greater than was predicted by a Hartree-Fock calculation, which gave differences of less than 1% between values for the relevant $4f$ interactions in the two cases.[16] The Hartree-Fock result is in better accordance with similar comparisons between $4f^N6s^2$ (neutral) and $4f^N$ (doubly ionized) in lanthanides heavier than cerium. The relatively large observed effect of the $6s$ electrons in Ce shows that the collapse of the $4f$ shell is not quite complete in the rare-earth sense. A further collapse between Ce and Pr is evidenced by the normal configurations Pr I $4f^36s^2$ and Pr II $4f^36s$.

Only a few even levels above 12 000 cm^{-1} have so far been assigned to configurations. Wilson's theoretical predictions[16] for $4f^25d6s$ and $4f5d6s6p$ were a very helpful guide for the expected low structure of these configurations. The interpretation of some of the lowest levels is doubtful, however, and calculations including several configurations and their interactions will be needed for progress. Some levels found below 14 000 cm^{-1} belong to $4f5d6s6p$, and as indicated in Table II this configuration may extend below 13 000 cm^{-1}. Consideration of the three configurations named in Table II and three related configurations, $4f^25d^2$, $4f6s^26p$, and $4f5d^26p$, is already sufficient to show that interpretation of the even levels of Ce I is a formidable problem. Goldschmidt and her group have begun calculation of appropriate matrices with configuration interaction.

In summary, the levels in Tables I and II establish the relative positions of the four lowest-extending configurations in Ce I and comprise a complete list below 10 000 cm^{-1}. They are taken from an analysis that includes well over a thousand levels. The work necessary for further interpretation and some extension of this analysis is underway.

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Damping of Plasma Oscillations in Atoms*

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Using the Thomas-Fermi model of the atom, we calculate the damping of collective oscillations due to the excitation of bound electrons to free states. We find that in some cases the decay time is much longer than the period of the oscillation. Our results indicate that weakly damped plasma oscillations can exist in atoms.

I. INTRODUCTION

The idea that electrons in heavy atoms may oscillate collectively is an old one dating back at least as far as the work of Bloch[1] in 1933. Bloch used the Thomas-Fermi model of the atom and treated the electrons as a degenerate electron gas which could be described by fluid equations. When perturbed, this gas oscillated about its equilibrium configuration under the influence of electrical and pressure forces in the same way as plasma oscillations occur in a gaseous plasma. This work of Bloch was extended by Jensen[2] and has recently become of interest again through the work of Wheeler and others.[3-6] There is some experimental evidence[7] for atomic energy levels above the ionization energy which may be due to atomic plasma oscillations. The subject has been reviewed by Kirzhnits and Lozovik.[8]

However, it is not really known whether such plasma oscillations can exist. As Kirzhnits and Lozovik remark, "The most acute problem for the atomic plasmon is its damping."[8] If a plasma oscillation is damped out in a time which is short in comparison with its period, then the concept of a collective oscillation is meaningless. Our work attempts to resolve the question of the existence of atomic plasma oscillations by calculating the damping.

II. CALCULATION OF DAMPING COEFFICIENT

Since the energy $\hbar\omega$ of a quantized plasma oscillation in an atom is generally above the ionization potential of the atom, these oscillations can decay by exciting bound electrons to free states. This damping mechanism is very similar to Landau damping in infinite homogeneous plasmas. The principal difference is that in the latter case the transitions responsible for damping are between free states of the electrons; in atoms the transitions are between bound and free states.

To arrive at a suitable expression for the damping coefficient γ, we start out with the integrodifferential equation for the electrostatic potential in a finite quantum-mechanical plasma, as derived by Cheng and Harris[9]:

$$-\nabla^2\Phi + \sum_{m,n} 4\pi e^2 \frac{f(m)-f(n)}{\hbar\omega-(E_m-E_n)+i\eta} \times \langle m|\Phi|n\rangle \psi_n^* \psi_m = 0 . \qquad (1)$$

Here Φ is the potential, ψ_n is the wave function of an electron with the set of quantum numbers denoted by n, E_n is the energy of this state, $f(n)=0$, 1 is the occupation number of the state, ω is the frequency of the oscillation, and η is a positive infinitesimal. Multiplying this equation by Φ^* and