Radiative lifetimes of $5d^{10}nl$ states of Hg^+

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The radiative lifetimes of excited Hg^+ $5d^{10}nl$ states are reported. They are measured by the multichannel delayed-coincidence method and for some of them Hartree-Fock calculations are made. These data are compared with other authors' theoretical and experimental results. Radiative lifetimes of $n\,^2S$, $n\,^2D$, and $n\,^2G$ states are discussed as functions of the principal quantum number.

INTRODUCTION

The subject of the present work is the radiative lifetimes of the $5d^{10}nl$ excited states of Hg^+. Mercury is among the most frequently used elements in spectral sources. Further investigation of the radiative constants of Hg^+ could explain the processes taking place in these sources, in particular, cw and pulsed mercury-ion lasers. These lasers operate on transitions between low-lying ^2S, ^2P, ^2D, and ^2F states of Hg^+.[1] It is assumed that the charge exchange process between helium ions and mercury atoms is responsible for the well-known red (6149-Å) and infrared (7944-Å) laser lines. On the other hand, the mercury atom is a system with a $5d^{10}$ close electron core and one excited electron; hence its atomic constants (excited-state energy, radiative lifetimes, etc.) might be expected to exhibit, to some extent, hydrogenlike behavior. However, this behavior might be influenced by relativistic effects due to the great atomic number and, especially, by electron-configuration interaction. It is clear that the lack of information about the radiative lifetimes of excited states and transition probabilities prevents the understanding of the processes in the He-Hg–mixture lasers and other mercury sources as well as of the mercury-ion spectrum. So far, three experimental studies have been devoted to ($5d^{10}nl$)-state radiative lifetimes employing beam-foil-[2] and beam-gas-level crossing[3,4] techniques, but only a few states were studied. Oscillator strengths of some spectral lines belonging to the $5d^{10}np\,^2P$, $ns\,^2S$, and $nd\,^2D$ states are calculated in Refs. 5 and 6.

It should be mentioned that there exists the "Beutler" system of terms which are formed when one electron from the $5d^{10}$ shell is excited in addition to the one already excited, $5d^96snl$. These states emit spectral lines, which sometimes have comparable or even higher intensities than those of the doublet system of terms. Moreover, there are spectral lines connecting both systems of terms. Prior to our previous studies,[7,8] the radiative-lifetime data of excited "Beutler" states were not available in the literature at all. Recently, radiative lifetimes of the lowest ground "Beutler" states $5d^96\,^2D_{5/2,7/2}$ were determined in an ion trap.[9] In the present work a systematic investigation of the radiative lifetimes of the $5d^{10}$ ns, np, nd, nf, and ng excited states is reported. The available literature's data are critically reviewed in order to outline the common current picture of the discussed problem. As a result, radiative lifetimes in some spectral series are also studied as functions of the principal quantun number.

EXPERIMENT

A multichannel delayed-coincidence method and nonselective electron excitation were applied in the present work. Note that the high-lying ionic states could be measured only in this way, although this type of excitation has some disadvantages, mainly cascade population corrections. The investigated states were obtained when electron pulses passed through mercury vapor in a diffusion cell. The temperature of the cell walls was maintained higher than that of the mercury container. The pressure of the saturated vapors was determined by the temperature of the container,[10] maintained with an accuracy of 0.5 °C. The electron gun had a 1.8-mm-diam oxidic cathode, heated indirectly. The electron beam was shaped by a system of three grids, located at distances of 1 mm from each other, followed by an anode at 10 mm from the last one. The first grid determined the electron-beam current. The second grid was a modulation one. A negative potential with respect to the cathode potential and rectangular pulses from a generator were simultaneously applied to the second grid forming the electron pulses. The generator pulses had a 33-V

amplitude, 10–100-ns duration, and a repetition rate of up to 3 MHz. These parameters depended on the radiative-lifetime values under study. Further, the electrons were accelerated by the third grid potential, which was equal to that of the anode. Hence, the third grid and the anode formed an equipotential space, where electrons moved without acceleration and where excitation of the investigated states took place. The electron-beam density was 12 A/m^2. The electron excitation energy was in the range of 30–150 eV, i.e., near the excitation function maxima. It is well known that when electron excitation energy surpasses the energy-state threshold, the probability of excitation of higher-lying states and hence appearance of cascade population of the investigated states increases. This may occur even when the excitation energy is only 0.5 eV above the threshold.[11] Note that the threshold measurements, in general, are possible only for the states emitting the strongest spectral lines in the investigated spectrum. On measuring near the threshold and above it, cascade population occurs, and the treatment of the decay curves is rather difficult. Hence, if the employed spectral lines are weak and therefore the threshold measurements are impossible, it would be better to choose a higher excitation energy which allows registration of the decay curves with well-expressed components. This increases the precision of the data that are obtained during the numerical processing.

In the present experiment, spectral lines were analyzed by means of a 40-cm light power grating (MDR-2)-type monochromator, which has a dispersion of 20 Å/mm, and were registered by a FEU-106 photomultiplier in the spectral range of 2000–8000 Å. The photomultiplier was used in the photocounting regime.

We used the traditional version of the delayed-coincidence method, with transformation of the time intervals to amplitude distribution by a time-to-amplitude converter and further data storage in a multichannel analyzer. The high repetition rate of the excitation pulses allows the rise of the signal-to-noise ratio. The useful signal did not exceed 1 kHz in the most favorable cases. The measurements were carried out in time interval ranges from 128 to 10 240 ns. We used a simple and precise method to calibrate our system.[12,13] The calibration error was less than 1%. The time resolution (of 2.3 ns) for the whole system was determined by measuring the $6s'\,^2D_{3/2}$ Kr^+ excited-state radiative lifetime.[14] This value of 0.3 ns was obtained in a line-shape experiment.[15] On the other hand, the lifetimes comparable with time resolution of the system can be measured if the section equal to approximately 3.5 times the resolution time is discarded, during the numerical treatment, from the initial part of the decay curves.[16] We took advantage of this procedure when the decay curves of short-living states were analyzed.

In our experiments, decay curves had in their maxima approximately 15×10^3 counts. The maxima-to-background ratio was from 5:1 to 20:1 and the maxima-to-statistical distribution ratio was in the range of 100:1. The background values were taken into account both from the end of decay curves and from the domain before the excitation pulses. The cascade contribution was given by the ratio of the amplitudes of the short and long exponents at the point where the short exponent had its maximum value. These amplitudes were obtained during the numerical treatment of the decay curves.

The decay curves had single- and double-exponential shape. The numerical processing of the curves was made by the REGN program system[17] separating the components with close decrements as well as extracting those with small amplitudes. For numerical procedure, 120–150 points were used. The decay curves' parameter errors, derived from numerical calculations, were in the range of 2–5 %, estimated by statistical distribution. Radiative lifetimes were determined after extrapolation to zero mercury pressure. This extrapolation was based on 10–15 points, taking into account individual errors of each experimental point. The saturation vapor pressure was varied in the 1–10 mTorr interval. The errors shown in Table I correspond to the doubled standard deviation.

RESULTS AND DISCUSSION

The investigated states and transitions are shown in Fig. 1. The radiative lifetimes measured in the present work are presented in Table I. The theoretical results obtained by us using the Hartree-Fock method are also

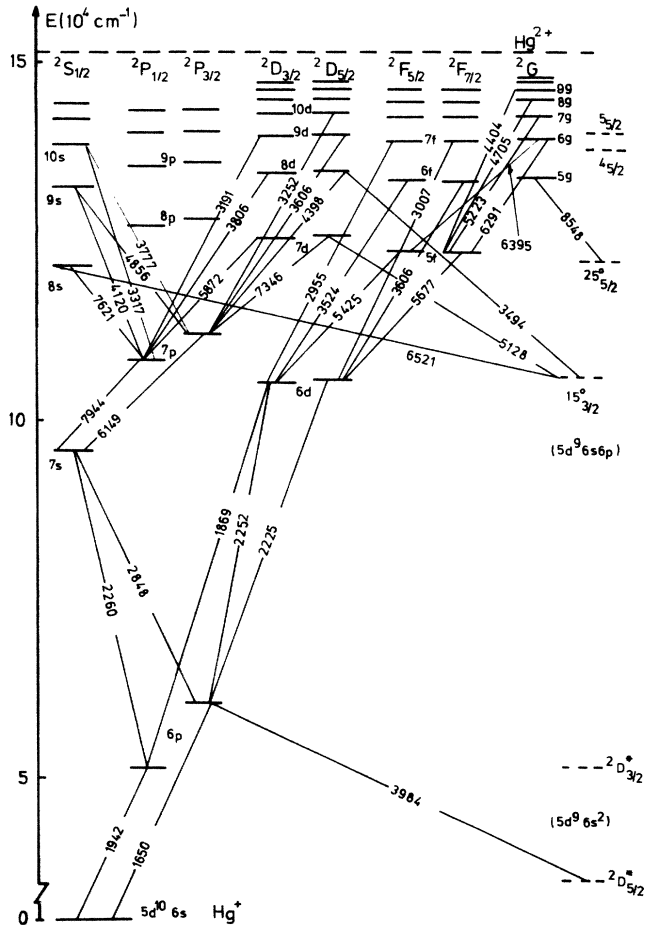

FIG. 1. Grotrian diagram of $5d^{10}nl$ states of Hg^+.

TABLE I. Radiative lifetimes of $5d^{10}nl$ excited states of Hg^+ (data in nanoseconds).

State	λ (Å)	Transition	Experiment				Theory		
			This work	Ref. 2	Ref. 3	Ref. 4	This work Length form	This work Velocity form	Ref. 5
$7s\ ^2S_{1/2}$	2847.7	$7s\ ^2S_{1/2}-6p\ ^2P_{3/2}$		1.9(2)			2.92	3.06	1.93
	2260.3	$7s\ ^2S_{1/2}-6p\ ^2P_{1/2}$							
$8s\ ^2S_{1/2}$							5.09	5.34	3.9
$9s\ ^2S_{1/2}$	4855.7	$9s\ ^2S_{1/2}-7p\ ^2P_{3/2}$	7.5(4)	10(15)			8.77	9.22	8.8
	4120.6	$9s\ ^2S_{1/2}-7p\ ^2P_{1/2}$							
$10s\ ^2S_{1/2}$	3317.3	$10s\ ^2S_{1/2}-7p\ ^2P_{1/2}$	3.8(4)				14.45	15.17	20.2
$6p\ ^2P_{1/2}$	1942.3	$6p\ ^2P_{1/2}-6s\ ^2S_{1/2}$		1.9(3)		2.3(3)			1.6
$6p\ ^2P_{3/2}$	1649.9	$6p\ ^2P_{3/2}-6s\ ^2S_{1/2}$				1.92(12)			0.9
$7p\ ^2P_{1/2}$	7944.5	$7p\ ^2P_{1/2}-7s\ ^2S_{1/2}$	18.8(12)						19.7
$7p\ ^2P_{3/2}$	6149.5	$7p\ ^2P_{3/2}-7s\ ^2S_{1/2}$	3.1(2)						2.82
$6d\ ^2D_{3/2}$	1869.4	$6d\ ^2D_{3/2}-6p\ ^2P_{1/2}$				1.7(3)	1.85	1.95	1.07
$6d\ ^2D_{5/2}$	2224.7	$6d\ ^2D_{5/2}-6p\ ^2P_{3/2}$		2.2(3)	1.9(3)	1.9(3)			1.3
$7d\ ^2D_{3/2}$	5871.7	$7d\ ^2D_{3/2}-7p\ ^2P_{1/2}$	5.0(6)				4.76	5.09	3.53
$7d\ ^2D_{5/2}$	7346.4	$7d\ ^2D_{5/2}-7p\ ^2P_{3/2}$	6.7(5)						4.76
	5128.4	$7d\ ^2D_{5/2}-15^0$							
$8d\ ^2D_{3/2}$	3806.4	$8d\ ^2D_{3/2}-7p\ ^2P_{1/2}$	10.6(6)	8.5(6)			9.54	10.26	7.8
$8d\ ^2D_{5/2}$	4398.6	$8d\ ^2D_{5/2}-7p\ ^2P_{3/2}$	10.7(5)						10.9
$9d\ ^2D_{3/2}$	3191.0	$9d\ ^2D_{3/2}-7p\ ^2P_{1/2}$	11.3(6)				16.74	18.08	
$9d\ ^2D$	3608			14.4(15)					
$10d\ ^2D_{5/2}$	3252.2	$10d\ ^2D_{5/2}-7p\ ^2P_{3/2}$	18.9(17)				26.96	27.52	
$5f\ ^2F_{5/2}$	5425.2	$5f\ ^2F_{5/2}-6d\ ^2D_{3/2}$	3.2(2)	7.2(6)	2.7(1)		6.34	6.27	
$5f\ ^2F_{7/2}$	5677.2	$5f\ ^2F_{7/2}-6d\ ^2D_{5/2}$	8.6(9)		7.6(5)				
$5g\ ^2G$							14.46	14.47	
$6g\ ^2G$	6291.3	$6g\ ^2G-5f\ ^2F_{7/2}$	26.8(20)				24.51	24.53	
	6394.9	$6g\ ^2G-5f\ ^2F_{5/2}$							
$7g\ ^2G$	5222.8	$7g\ ^2G-5f\ ^2F_{7/2}$	40.7(24)				38.30	35.91	
$8g\ ^2G$	4704.6	$8g\ ^2G-5f\ ^2F_{7/2}$	66.6(43)						
$9g\ ^2G$	4404.4	$9g\ ^2G-5f\ ^2F_{7/2}$	90(7)						

shown. The wave functions of the excited electron were determined in the space of a frozen core of Hg^{2+} ion without spin-orbital interaction. Transition probabilities were determined in both the velocity and length forms of the transition operator. In Table I, two values of theoretical radiative lifetimes are shown accordingly. All possible transitions from investigated states were taken into account. The present work's data are compared with other authors' results that were obtained by beam-foil techniques[2] and the level crossing method after beam-gas excitation.[3,4] Our data are also compared to theoretical oscillator-strength values[5] that were converted for convenience into radiative-lifetime values. These oscillator strengths are calculated by means of a semiempirical wave function obtained by taking into account the exchange effects in the relativistic and nonrelativistic cases only; only for the low-lying $7s\ ^2S_{1/2}$, $8s\ ^2S_{1/2}$, $6p\ ^2P$, and $6d\ ^2D$ states have all transitions been considered.[5] The other state's theoretical lifetimes should be assumed to be an upper limit of estimation. In Table I, radiative lifetimes that were calculated in the relativistic case are shown.

Some comments on the peculiarities of the investigated spectral series and a comparison with other authors' results should be made.

$n\ ^2S_{1/2}$. In the present work, the radiative lifetime of $10\ ^2S_{1/2}$ was measured using spectral line 3317 Å ($10\ ^2S_{1/2}-7p\ ^2P_{1/2}$).[18] The classification of this spectral line is to some extent controversial. In earlier studies,[19,20] a spectral line with the wavelength 3317.62 Å was assigned to the $5d^96s6p\ ^2D^3_{5/2}-5d^96s6p\ ^4P_{5/2}$ transition, i.e., it was assumed to belong to the "Beutler" system of terms. The radiative lifetime of the $^2D^3_{5/2}$ state that was obtained by us, using another spectral line 3638.39 Å (Refs. 17 and 18) is 6.0 ns.[8] Hence the 3317.3-Å spectral line comes from the $10s\ ^2S_{1/2}$ state. The other spectral line that was emitted by the $10\ ^2S_{1/2}$ states 3777 Å ($10\ ^2S_{1/2}-7\ ^2P_{3/2}$) had a small intensity and was used only for qualitative measurements, which confirmed the value obtained by the 3317-Å spectral line. On the other hand, the $10s\ ^2S_{1/2}$ state's radiative-lifetime value diverts from the commonly adopted radiative-lifetime dependence on principal quantum number, and exhibits a significant discrepancy from the theoretical result. Theoretical data of the $10\ ^2S_{1/2}$ state were obtained in the present work, taking into account all possible transitions, while in Ref. 5, only the $10s\ ^2S_{1/2}-7p\ ^2P_{1/2,3/2}$ spectral-line oscillator strengths were calculated. Note that this discrepancy might be due to an incorrect spectral line

classification as well as to strong electron-configuration mixing of the $5d^{10}ns$ electron configuration mainly with the $5d^8 6s^2 7s$ and $5d^9 6p^2$ ones.

$np\ ^2P_{1/2,3/2}$. The spectral lines 7944.5 and 6149.5 Å, which arise from $7\ ^2P$ doublet levels, are both laser lines and laser levels, respectively, of the He-Hg–mixture laser.[1] Considerable difference between the measured lifetimes of $7\ ^2P$ multiplet states is confirmed by the theoretical calculations,[5] where only the transitions to $7s\ ^2S_{1/2}$ states are taken into account. This deviation is due to strong spin-orbital interaction. Spectral lines from higher-lying $5d^{10}np$ states turned out to be too weak to be studied experimentally.

$nd\ ^2D_{3/2,5/2}$. There exists a great number of transitions connecting $7d\ ^2D$ and $8d\ ^2D$ states with Beutler states, but there is no information about such transitions for higher-lying $nd\ ^2D$ states. The radiative lifetime of $7d\ ^2D_{5/2}$ was measured by means of the 7346-Å ($7d\ ^2D_{5/2}-6p\ ^2P_{3/2}$) line, which is a laser one, and by the 5128-Å ($7d\ ^2D_{5/2}-15^0_{3/2}$) line to the Beutler system of states. Both spectral lines gave identical results. The other state of that doublet $7d\ ^2D_{3/2}$ was studied using spectral line 5871.76 Å. According to the *Tables of Spectral Lines*,[21] this line is blended by the 5871.97-Å atomic line, whose intensity is probably three times smaller. The state $9d\ ^2D_{5/2}$ emits the spectral line 3605.80 Å ($9d\ ^2D_{5/2}-7p\ ^2P_{3/2}$), coinciding with the spectral line 3605.86 Å ($6f\ ^2F_{7/2}-6d\ ^2D_{5/2}$). The 3605.8-Å spectral-line decay curve had a complicated three-exponential shape. The value of 9 ns can be obtained from the first exponent. That is the reason why we did not measure the $9d\ ^2D_{5/2}$ state. Andersen and Sørensen[2] have reported the radiative lifetime of the $9d\ ^2D$ state, measured by 3608 Å. There is no spectral line with a such wavelength in Ref. 21 or in the original works,[18,19,20,22] either. On the other hand, if there is an error in Ref. 2, it is not clear (i) how 3605.80 and 3605.86 Å have been separated, and (ii) how the radiative lifetime of $9d\ ^2D-14.4$ ns has been obtained. The experimental lifetimes of the $5d^{10}nd\ ^2D$ states can be compared with theoretical data. The theoretical lifetimes for $7d$ and $8d$ states are shorter than the experimental ones, while the measured values for $9d$ and $10d$ states are considerably shorter than the theoretical data.

$nf\ ^2F_{5/2,7/2}$. Spectral line 5677.17 Å and state $5f\ ^2F_{7/2}$ are, respectively, the laser line and upper laser level of the He-Hg–mixture laser.[1] As for the $7p\ ^2P$ states, a significant difference between the radiative lifetimes of deeply-lying $5f\ ^2F$ states is observed. The cascade population contribution to those states is also different. For $5f\ ^2F_{5/2}$ it is 10%, whereas for $5f\ ^2F_{7/2}$ it is up to 60%. There are some problems in blending the spectral line 3605.86 Å ($6f\ ^2F_{7/2}-6d\ ^2D_{5/2}$) with the line 3605.80 Å ($9d\ ^2D_{5/2}-7p\ ^2P_{3/2}$) which we have already mentioned; 3524.17 Å ($6f\ ^2F_{5/2}-6d\ ^2D_{5/2}$) is blended by the atomic line 3524.27 Å.

$ng\ ^2G$. There are, in the literature, no radiative-lifetime data for these states to compare with our results. It should be pointed out that there is a good agreement between experimental and theoretical data for ^2G states.

PRINCIPAL-QUANTUM-NUMBER DEPENDENCES OR RADIATIVE LIFETIMES

The system with an occupied close electron core and one excited electron, as the $5d^{10}nl$ electron configurations of Hg^+, might reveal a hydrogenlike behavior of the excited-state energies and radiative lifetimes when the principal quantum number n varies. Radiative lifetimes of these spectral series have a simple dependence on the effective quantum number n^*, since $\tau=C(n^*)^\alpha$, where $n^*=n-\Delta$, Δ is the quantum defect, and C,α are characteristic constants for every spectral series (α should be equal to 3 in the hydrogen case). However, it was verified experimentally that for many atomic and ionic series, α differs from 3.[23] On the other hand, the electron-configuration interaction and the local perturbation give rise to a deviation from the $\tau=\tau(n^*)$ rule. This rule might be expected to hold true if the one-electron approximation for an electron configuration is valid. This can be checked by investigating the dependence of the effective orbital parameter $q(E)$ as a function of the excited-state energy.[24] The effective orbital parameter is

$$q(E)=Z^*(2/|E|)^{1/2}-2l-1\ ,$$

where E is the experimental excited-state energy in atomic units, l is the orbital quantum number, and Z^* the effective charge. When the one-electron approximation is valid for the investigated spectral series,[24] $q(E)$ increases linearly with increasing energy, if the atomic core contains electrons with the same orbital quantum number as the valent one and, vice versa, if the electron core does not confine the electrons with the same orbital quantum number as the valent one, the $q(E)$ decreases with increasing state energy.

The radiative lifetimes as a function of the effective quantum number for $nd\ ^2D$, $ng\ ^2G$, and to some extent for $ns\ ^2S$ spectral series, are presented in Fig. 2. In this figure, the experimental results from Table I, as well as

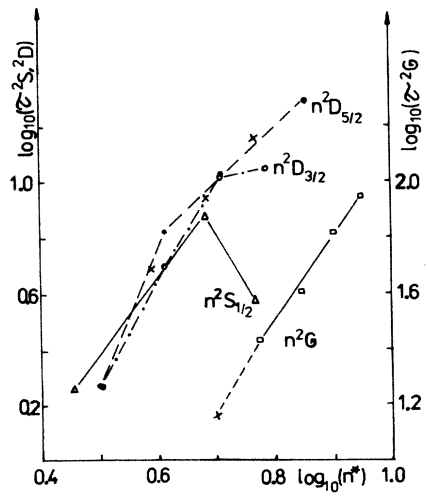

FIG. 2. Radiative lifetimes τ of $ns\ ^2S$, $nd\ ^2D$, and $ng\ ^2G$ states as a function of effective quantum number n^*; $\times$, theoretical radiative lifetimes of $ns\ ^2S$ and $5g\ ^2G$ states.

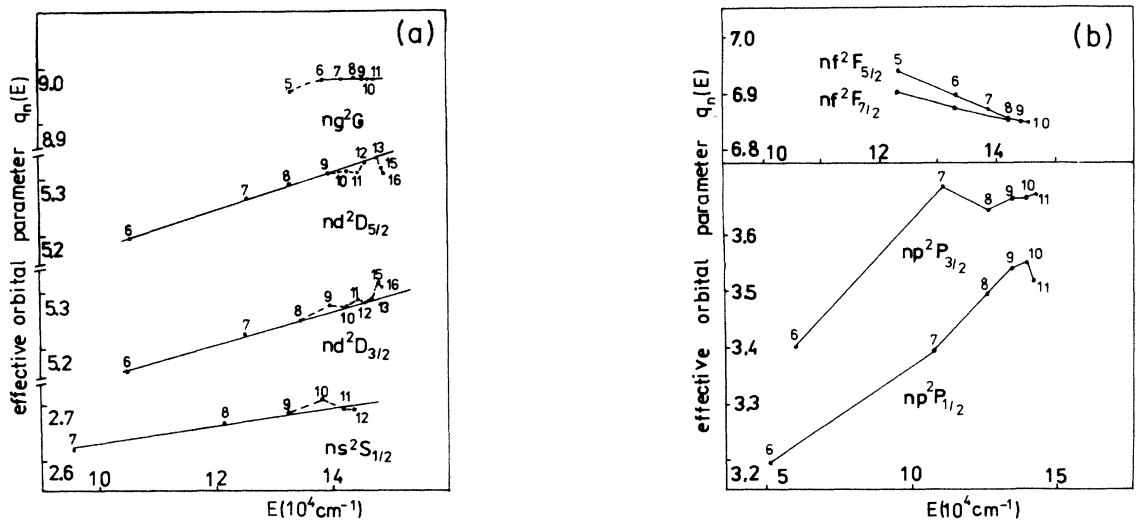

FIG. 3. (a) Effective orbital parameter q as a function of excited-state energy, $ns\ ^2S$, $nd\ ^2D$, and $ng\ ^2G$ states. (b) Effective orbital parameter q as a function of excited-state energy, $np\ ^2P$, and $nf\ ^2F$ states.

the theoretical values of $n\ ^2S_{1/2}$ ($n=8-10$) and $5g\ ^2G$ states, are presented. It turns out that only the $ng\ ^2G$ dependence is linear with constants $C=0.18$ ns and $\alpha=3.02$, i.e., hydrogenlike dependence occurs. In Fig. 3 the effective orbital parameters $q(E)$ as functions of the excited-state energy are presented. For the $ng\ ^2G$ series, this function is linear, which correlates with the $ng\ ^2G$ radiative-lifetime dependence from Fig. 2. On the other hand, the $nd\ ^2D$ and $ns\ ^2D$ series deviate from the straight line for those states, where radiative-lifetime dependences are perturbed (see Fig. 2). One can see on Fig. 3 that the $np\ ^2P$ series also is perturbed, the perturbation being maximal for the $7p\ ^2P$. For the $nf\ ^2F$ series, when the state energy increases, the value of $q(E)$ decreases, although the atomic core confines electrons which have the same orbital quantum number. Therefore, in that case, the one-electron approximation is not valid.

Similar radiative-lifetime dependences $\tau=\tau(n^*)$ were investigated in the doublet spectrum[23] of Cd^+. There are some differences from the Hg^+ spectrum. In the Cd^+ case, the $nd\ ^2D$ radiative lifetimes have a linear dependence as a function of state energies with the coefficient $\alpha=3.07$, while the $nf\ ^2F$ series has a local perturbation at $8f\ ^2F$ states and the $4d^9 5s 5p\ ^2F_{7/2}$ level was found as a perturbator. In this case the two-channel quantum-defect theory was applied.

CONCLUSION

The radiative lifetimes of excited $5d^{10}nl$ states of Hg^+ that are measured in the present work and a few available experimental data obtained by other authors are in comparatively good agreement. The experimental results are also in good agreement with the theoretical ones. Some differences are observed for those states where experimental values diverge from the hydrogenlike function of radiative lifetimes in principal quantum number. We suppose that the electron-configuration mixing of $5d^{10}nl$ with "Beutler" electron configurations should be taken into account in the theoretical calculation, since, as one can see from Fig. 3, the effective orbital parameter diverges for the ns and nd series at approximately the same energies.

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