Laserspectroscopic Investigations in the Configuration 5d6s6p of Lu I

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In an atomic beam of Lu, six levels of the configuration 5d6s6p in the energy range 18000 to 23000 cm^{-1} were selectively populated with the light of a pulsed dye laser. Time resolved observation of the reemitted resonance light was used to deduce the lifetimes of the excited levels.

Introduction

The classification of the lower levels of Lu I is mainly based on the pioneer work of Meggers and Scribner [1] and Klinkenberg [2]. In a recent investigation Verges and Wyart [3] observed 155 emission lines in the infrared region of $1-3.7\,\mu m$. By means of Fourier transform spectroscopy they measured the magnetic and electric hyperfine constants of the combining levels of 40 lines for the isotope Lu^{175}. With these values it was possible for them to revise and extend the level scheme. In this way the term classification of the configurations $5d^26s$ and 5d6s6p could be completed.

Measurements of hyperfine constants of energy levels are very useful for a detailed study of coupling properties within a given configuration or of possible interaction with other configurations of the same parity. Complementary information for the classification of levels is given by the determination of lifetimes, because electric dipole transition probabilities depend on the coupling properties and on the radial integrals between configurations of different parity.

In the case of the 5d6s6p configuration of Lu I the lifetimes of the lower levels at about 20 000 cm^{-1} (Fig. 1) should mainly be due to transitions to the levels $^2D_{3/2}$ and $^2D_{5/2}$ (1994 cm^{-1}) of the ground state configuration $5d6s^2$. Because of the ΔE^3-dependence of electric dipole transition probabilities the transitions to levels of the configuration $5d^26s$ which lie above 18 851 cm^{-1} should be negligible. As most of the lower levels of the configuration 5d6s6p belong to the quartet system in the LS coupling scheme it is obvious that these levels can only decay to the ground term $5d6s^2$ ^2D through the doublet composition of their eigenfunctions. Therefore lifetimes of these levels are very sensitive to admixtures of doublet wave functions ^2P, ^2D and ^2F.

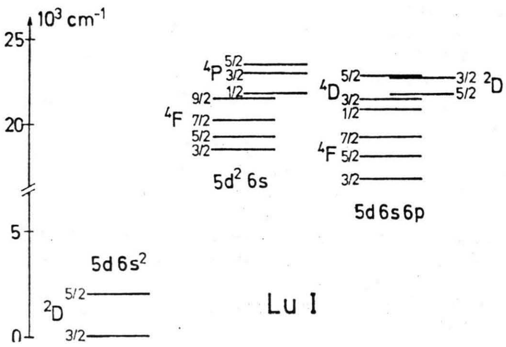

Fig. 1. The interesting lower levels of Lu I.

In our experiments we have investigated six lower levels of the configuration 5d6s6p by selective laser excitation with a pulsed dye laser. By means of time resolved observation of the reemitted resonance light we have measured the lifetimes of these excited levels. These experimental values are compared with values deduced from the transition probabilities of Corliss and Bozman [4] and in the case of $^4D_{1/2}$ with a calculation using eigenfunctions of Wyart [5] and the Coulomb approximation for the relevant radial integral.

Experiments

The atomic beam of Lu was produced by a tantalum oven which was heated by electron impact to a temperature of about 1400 K. For the excitation an amplified tunable dye laser was used which was pumped by a nitrogen laser with a peak power in

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the order of 1 MW. The pulse length was about 3 ns and the repetition rate up to 250 Hz. Inside the vacuum chamber a system of light baffles was put into the path of the laser beam in order to eliminate stray light. The reemitted resonance light was selected by a monochromator with respect to the corresponding transitions.

All investigated levels with exception of $^4D_{1/2}$ have total angular momentum $J = 3/2$ or $5/2$, so they can decay into both levels $^2D_{3/2}$ and $^2D_{5/2}$ of the ground state configuration $5d6s^2$. Vice versa it is possible to excite these levels from both ground state levels because due to thermal excitation the metastable $^2D_{5/2}$ (1994 cm^{-1}) is sufficiently populated at a temperature of about 1400 K. Therefore it was always possible to separate the wavelenght of the excitation and the observation. For example $^4D_{5/2}$(21 195 cm^{-1}) was excited at 520.6 nm from the ground state level $^2D_{5/2}$ and its decay was observed at 471.6 nm by the transition to $^2D_{3/2}$, whereas for the excitation from $^2D_{3/2}$ the transition to $^2D_{5/2}$ was used for the observation. Such a spectral separation of the excitation and the observation is quite useful for time resolved photon counting techniques because photomultipliers usually are sensitive to excessive stray light from the laser pulse which can cause distortions in the time response.

For the time resolved observation of the resonance light the technique of the delayed coincidence was used. The principal part of the electronics consisted in a time to amplitude converter (TAC) which was started by the laser pulse and stopped by the first observed photon. This technique can only be applied if the mean counting rate is considerably less than one photon per laser pulse. Otherwise the exponential decay curves are distorted with the result that the measured values of lifetimes tend to be too short. Therefore in our experiments the computer controlled counting rate was in the order of 0.1 (or less) photons per laser pulse. The residual pile-up effect was eliminated numerically.

The output of the TAC was stored via a data processing system by a computer which was programmed as a multichannel analyzer. Further details are given in a previous paper [6]. From the exponential decay curves the lifetimes were deduced. The values are listed in Table 1.

Table 1. Results of the lifetime measurements in some levels of the configuration 5d6s6p.

Investigated level	Energy [cm^{-1}]	Transition wavelengths [nm] $^2D_{3/2}$	$^2D_{5/2}$	Lifetime [μs] this work	others
$^4F_{5/2}$	18505	540.2	605.5	0.43(2)	2.93 [a]
$^4D_{1/2}$	20762	481.5	—	1.02(6)	6.34 [a]
$^4D_{3/2}$	21195	471.6	520.6	2.45(15)	7.68 [a]
$^2D_{5/2}$	21462	465.8	513.5	0.080(4)	0.35 [a]
$^2D_{3/2}$	22124	451.8	496.6	0.043(3)	0.21 [a] 0.049(1) [b]
$^4D_{5/2}$	22222	449.8	494.2	0.82(5)	4.51 [a]

[a] Lifetimes deduced from gf-values of Corliss and Bozman [4].
[b] Lifetime by Goebel [7].

Discussion

For most of the investigated levels no direct comparison with other experimental values was possible. Only in the case of $^2D_{3/2}$ a level crossing experiment [7] yielded the value $\tau = 0.049(1)\,\mu s$. For the other levels one can tentatively use the gf-values of Corliss and Bozman [4]. Under the assumption that no strong transitions to other configurations than the configuration $5d6s^2$ of the ground state exist, one can deduce from these gf-values the lifetimes listed in Table 1. A comparison with our values shows that they are systematically longer by a factor of about five.

For a theoretical calculation of the transition probabilities one needs the wavefunctions of the excited states and the ground state. Because of the overlapping of the even configuration $5d6s^2$ of the ground state with $5d^26s$, $5d^3$ and $6s6p^2$, and because of the overlapping of the odd configuration 5d6s6p of the excited states with $5d^26p$ and $6s^27p$ one must regard simultaneously all these configurations. However the analysis of Vergès and Wyart [3] shows that 98% of the ground state and 92 – 98% of the excited states have the composition $5d6s^2$ and 5d6s6p, respectively. Therefore the other configurations will be neglected.

Next one has to consider a suitable coupling scheme of the excited states with respect to the parentage approximation. Using LS coupling the approximate parentage scheme is 5d6s(^1D) and 5d6s(^3D), respectively. The addition of the 6p electron yields ^2P, ^2D and ^2F states for the parent ^1D, and ^2P, ^2D, ^2F or ^4P, ^4D, ^4F states for the parent ^3D. The actual wave functions are compositions of these states. As an example, the wave function of $^4D_{1/2}$(20 762 cm^{-1}) is given by [5]

$$|5d6s6p\,^4D_{1/2}\rangle = -0.048\,|(^1D)\,^2P_{1/2}\rangle - 0.126\,|(^3D)\,^2P_{1/2}\rangle - 0.113\,|(^3D)\,^4P_{1/2}\rangle - 0.968\,|(^3D)\,^4D_{1/2}\rangle .$$

For the calculation of the transition probabilities to the ground state 5d6s^2 ^{2}D it is necessary to consider only the doublet composition of the wave functions. For the example above the first two parts with $^2P_{1/2}$ determine the transition. For the radial integral the Coulomb approximation gives $\langle 6p\,|\,er\,|\,6s\rangle = 3.2\,ea_0$ resulting in a transition probability $A(^4D_{1/2},\,^2D_{3/2}) = 8.91 \cdot 10^5\,s^{-1}$. Assuming that only this transitions is mainly responsible for the lifetime of $^4D_{1/2}$, the value $\tau = 1.12\,\mu s$ is deduced, which is in reasonable agreement with our experimental value. Further theoretical calculations with respect to the eigenfunctions and the radial integrals together with the experimental determination of the branching ratios of the transitions are neccessary to deduce reliable gf-values.

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