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Natural Radiative Lifetimes in the 5snd 1D2 Series of Sr I

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Natural radiative lifetimes have been measured for the 5snd 1D2 series in Sr I for the previously not measured n=10, 11 and 12 states. The measurements connect earlier measurements in lower and higher states in this heavily perturbed series. The lifetimes were determined using pulsed laser excitation and time-resolved detection.

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Introduction

The 5snd 1D2 series in Sr I is strongly perturbed both by configuration interaction with doubly excited states and by singlet-triplet mixing. The perturbations can be observed as irregularities in the basic energy level scheme [1, 2, 3] and in other observable quantities such as radiative lifetimes [4], Landé factors [5] and hyperfine structures [6]. Wavefunctions for the states in a perturbed series are usually obtained from experimental energy levels using multi-channel quantum-defect theory (MQDT) [7]. Radiative lifetimes for the 5snd 1D2 states in Sr I were measured by Gornik for n≤9 [8] and Graffström et al. for 13≤n ≤22 [4]. The lifetime values for the higher states strongly deviated from the trend given by the lower states. In the present paper we report on measurements of the n=10, 11 and 12 states. Pulsed laser excitation of an atomic beam was employed and different excitation and detection schemes were used for the three states. A localized perturbation not showing up in measurements of other quantities like, g-factors and hyperfine structures, was observed in the measured lifetimes.

Experimental Set-Up

In the original set-up two-step laser excitation and delayed coincidence technique were employed. For the first step, populating the 5s 5p 1P1 level, we used a krypton-ion laser (Coherent Radiation CR 3,000 K) with a power of 2 W pumping a multimode CR-599 dye laser with Coumarin 1. For the second step an argon-ion laser (Spectra Physics 171-18) pumping a single-frequency Stilbene I ring laser (CR-699-21) was used. The wavelengths as measured by a digital wavelength-meter were found to be in good agreement with those given in [1]. An acousto-optic modulator (SORO-MAR-50) controlled by a pulse generator modulated the laser beam for the second step. A period corresponding to ~10 τ and a pulse duration of ~3 τ gave optimum working conditions. The two beams were made to overlap by means of a beamsplitter, collimated with a lens and then crossed the atomic beam at a right angle. The atomic beam was produced in a resistively heated oven containing Sr metal. Apertures were used to reduce straylight from the oven. The atomic beam was terminated on a liquid nitrogen cool trap. The decay of the investigated level was detected with a photomultiplier equipped with suitable filters to avoid background from cascading and from deexcitation of the first level. A pulse generator created the start pulses for a time-to-amplitude converter, while the first photon arriving at the PMT generated a stop signal. The pulses from the converter were fed into a multichannel analyser interfaced to a microcomputer and the decay curve was stored as a data file on a floppy disc. For the n=11 and 12 states weak fluorescence signals were obtained on the excitation wavelength. The main decay channel turned out to
be to the $5s\,5p\,^3P_1$ and $^3P_2$ states. However, for the $n=10$ level no fluorescence at all was observed when scanning the laser over the $5s\,5p\,^3P_1-5s\,10d\,^1D_2$ wavelength. This transition is also missing in the energy level measurement of [2, 3]. In a new approach to measure this state we used the $5s\,5p\,^3P_1-5s\,10d\,^1D_2$ intercombination line for excitation. The $^3P_1$ level was populated by driving a discharge along the atomic beam into the oven. For the further excitation from the metastable level $5s\,5p\,^3P_1$ we used a frequency doubled Nd:YAG pumped dye laser (Quanta Ray PDL-1). This excitation scheme gave a good signal. The radiative decay was recorded using a transient recorder. About a thousand transients were added. The decay curves were sent to the computer and evaluated in a least-squares procedure to get the radiative lifetimes. For each state, measurements were performed on several occasions and particular attention was paid to ensure that there was no influence due to pile up, multiple scattering, collisions and Zeeman quantum beats.

Results

In Fig. 1 a typical fluorescence curve for one of the studied new levels ($n=12$) is shown. The results are summarized in Fig. 2 where we have plotted the radiative lifetime on a logarithmic scale versus the logarithm of the effective principal quantum numbers ($n^*$). The previous results by Gornik [8] and Grafström et al. [4] are plotted in the same diagram. The effect of the perturbations in the series can now be studied in detail.

In an unperturbed Rydberg series the lifetimes are expected to increase monotonically. However, for the $^1D_2$ states there is an overall shortening of the lifetime values for the higher states. This is caused by an interaction with the $4d\,6s\,^3D_2$ state which is mixed into a large number of levels with a maximum admixture around $n=16$. In this region there is also a strong singlet-triplet mixing. Further the plot shows a dip at the $n=12$ state which is energetically close to the $4d^2\,^3P_2$ level. A $^3P_2$ state can not perturb a $^1D_2$ state directly but if LS coupling is not strict the $4d^2\,^3P_2$ state will have some $^1D_2$ character. In the MQDT analysis by Esherich [1] the only indication of a perturbation from $^3P_2$ was somewhat larger errors in the fit around $n=12$ than for other states. Inclusion
of the 4d2 perturbers in the analysis did not improve on the total agreement with all measured states, i.e. the width of the autoionizing 4d2 1D2 level. Our lifetime results, however, show a perturbation in the n = 12 region. The lifetime of the 4d2 3P2 state has been measured to 7 ns [4] which is much shorter than the lifetime of the states in the series. Therefore even a small admixture has a drastic effect. The radiative decay rate \( \Gamma \) of a perturbed Rydberg state can be estimated by
\[
\Gamma(n^*) = a \cdot \frac{\gamma}{(n^*)^6} + b \cdot \Gamma_{\text{perturber}}.
\]

The constants \( \gamma \) and \( \beta \) can be determined by investigating the trend of the unperturbed levels. Neglecting the effect of the 4d6s 3D2 perturber the admixture of the 4d2 3P2 state into the \( n=10, 11 \) and 12 states can be estimated. If we assume the lifetimes of the lower states to be unaffected by perturbations the admixture of the \( 3P_2 \) state is 0, 1.5 and 3% respectively. The lifetime of the 5s 10d 1D2 state follows the trend of the lower states. However, in the measurements it was found that the probability for the 5s 5p 1P1 – 5s 10d 1D2 transition is very low, indicating a more complex situation.

To summarise we have measured the lifetimes for three states in the 5 snd 1D2 series of strontium, connecting the results of two earlier measurements. Our results give an input for an extended analysis of perturbed Rydberg series.

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**References**