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Hyperfine-dependent lifetimes induced by singlet-triplet mixing

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The effect of hyperfine-induced singlet-triplet mixing on radiative lifetimes has been studied in neutral strontium. Different lifetimes are, for the first time, observed for different hyperfine components in laser-spectroscopy measurements combining high spectral and temporal resolution. The experimental results are found to be in good agreement with theoretical calculations.

Natural radiative lifetime measurements can, under certain circumstances, serve the purpose of investigating the composition of atomic wave functions. In particular, the lifetime values obtained for sequences of normally long-lived Rydberg states sensitively reflect the admixture of short-lived doubly excited states. Particularly spectacular effects are obtained for the $6sns\ ^1S_0$ and $6snd\ ^1D_2$ sequences of Ba.¹⁻⁴ Natural radiative lifetimes do not normally show any dependence on the F quantum number within a state, since the lifetime is an electronic quantity and has nothing to do with the nuclear properties. However, there can be such a dependence due to hyperfine-induced mixing between levels with different radiative lifetimes. In the present paper such effects are demonstrated for the first time.

During recent years hyperfine-induced singlet-triplet mixing has been studied, particularly for He (Ref. 5) and in sequences of the alkaline earths.⁶⁻⁸ In these studies the influence of the hyperfine structure (hfs) has been used to probe the interaction. There have also been reports on transition probabilities affected by this kind of hyperfine-induced mixing in Hg and more recently in OVII and Fe VIII.⁹ Beigang *et al.*¹⁰ report a strong localized singlet-triplet mixing between the series $5snd\ ^1D_2$ and $5snd\ ^3D_3$ for $n=19$ in ^{87}Sr . These are the states investigated in the present work regarding F -dependent lifetimes.

The difference in energy ΔE between these two states is comparable to the Fermi contact term a_{5s} of the inner s electron, leading to the local mixing. The effect can only exist in the ^{87}Sr isotope ($I=\frac{9}{2}$, natural abundance 7.02%), since it is the only stable one having a nonzero nuclear magnetic moment. On the other hand, measurements on the even isotopes, e.g., ^{88}Sr (natural abundance 82.56%), give the unperturbed lifetimes for the singlet and the triplet states. We describe lifetime measurements on individual hfs components of ^{87}Sr and on the unperturbed states of ^{88}Sr . In order to measure lifetimes of individual hfs components, one must have high-energy resolution as well as high temporal resolution. Therefore, a delayed coincidence method employing a pulse-modulated cw single-mode dye laser was used. This scheme, referred to as PUMOLS (pulse modulated laser spectroscopy), has been applied previously for radiative lifetime measurements on Rydberg sequences in Sr and Ba.^{1,2,11,12}

The PUMOLS technique has been described in detail

earlier,¹³ but a brief description of the method used is presented here. In PUMOLS experiments cw laser light is used to excite free atoms in an atomic beam. The light is externally pulse-modulated at high repetition rates. Suitable pulse lengths and light-off intervals are set depending on the individual lifetimes, i.e., approximately 3τ and 10τ , respectively. The fluorescence photons are detected in a photomultiplier tube (PMT). It is essential that each laser pulse gives rise to, *at the most*, one fluorescence photon at the detector. These photons give the stop signals to a time-to-amplitude converter (TAC) started with trigger pulses directly from the pulse modulator. Necessary precautions, as discussed in Ref. 13, regarding, e.g., pile up, multiple scattering, and distortion by Zeeman quantum beats, must be taken. A multichannel analyzer (MCA) is used for sampling the decay photons and the completed curves are stored in a computer for later evaluation.

In neutral strontium, two-step excitation from the $5s^2\ ^1S_0$ ground state to the $5s\ 19d\ ^1D_2$ Rydberg state, via the $5s\ 5p\ ^1P_1$ intermediate state was employed, as illustrated in Fig. 1. The actual experimental setup used is shown in Fig. 2. For the first step, at 460.7 nm, a multimode dye laser (Coherent Radiation CR 599) running on Coumarin 470 was used. An uv krypton-ion laser (CR 3000 K) was utilized as a pump. To achieve "white" excitation of the intermediate state, the piezo-mounted folding mirror of the dye laser was scanned rapidly with a voltage ramp to smear out the mode structure of the laser. A Stilbene 3 single-mode ring dye laser (CR 699-21) pumped by an argon-ion laser (Spectra Physics SP-171-18) was used for the second step. The light from the ring laser was pulse-modulated externally by an acousto-optic modulator (SORO-MAR-50). The linewidth of the single-mode laser (normally 1 MHz) was only slightly affected by Fourier broadening due to the pulsing, since the pulse length was 1 μs or longer. The strontium atomic beam was produced in a resistively heated oven, inside a vacuum chamber. The excitation was performed at right angles to the atomic beam, which was collimated with apertures to a ratio of about 1:20 for reduction of the Doppler width. The optical detection system, orthogonal to both the atomic and laser beams, consisted of appropriate color and interference filters, together with a Peltier-cooled PMT (EMI 9558 QB).

Since the $19d\ ^3D_3$ state for the even isotopes cannot be excited from the $5s\ 5p\ ^1P_1$ level, the metastable state

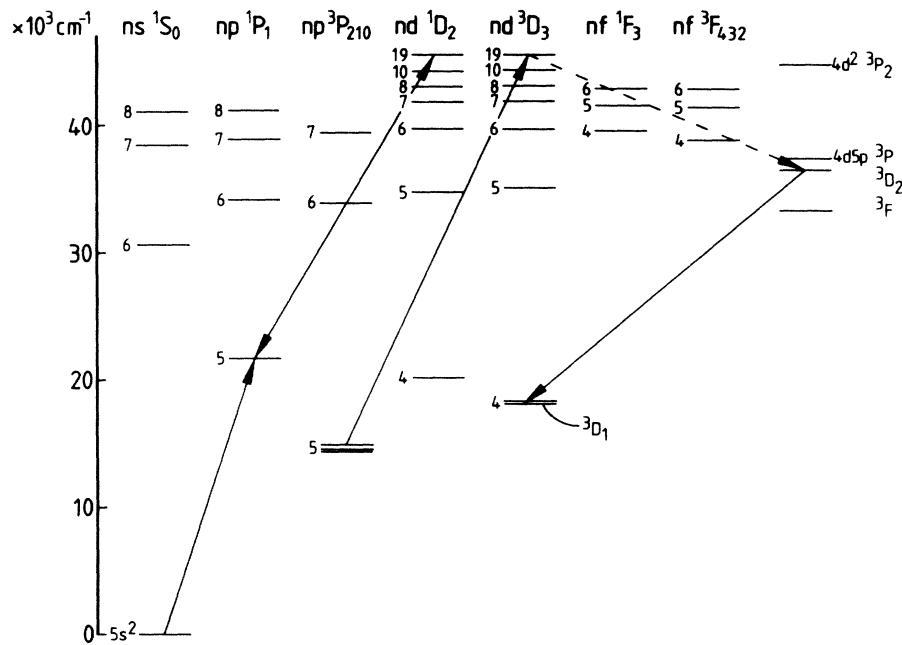


FIG. 1. Schematic energy-level diagram for neutral strontium. The transitions used for excitation and detection are indicated.

$5s5p\ ^3P_2$ had to be used. This called for a pulsed laser system as the wavelength was in the uv region (326.4 nm). The metastable population was achieved by applying a discharge in the atomic beam about 1 cm above the oven, and letting the released electrons collide with the atoms. A Nd-YAG (yttrium aluminum garnet) pumped dye laser (Quanta-Ray DCR-2 and PDL-1) and a Raman shifter (Quanta-Ray RS-1) were then used to excite the atoms to the triplet state. The decay was recorded with a transient recorder (Biomation 8100) and an external mass memory in which the transients were added. However, the route of decay was not as expected. It was found that the 3D_3 state mainly decayed through transitions involving both

valence electrons. There was no distinguishable fluorescence at the laser wavelength, but a strong signal was found at the wavelength of the $4d5p\ ^3D_2-5s4d\ ^3D_1$ line. This decay route is indicated in Fig. 1. The lifetime deduced from this cascade can be trusted since the intermediate doubly excited state has a much shorter lifetime than the $19d\ ^3D_3$ state.

The hyperfine structures of the $18-20d\ ^1D_2$ states were recorded by scanning the single-mode laser, and they were found to be in agreement with those obtained in Ref. 10. Only in the $19d$ scan did extra peaks appear. The peaks belong to the $19d\ ^3D_3$ state and have F quantum numbers from $\frac{5}{2}$ to $\frac{13}{2}$. The triplet components with $F = \frac{3}{2}$ and $\frac{15}{2}$ do not mix into the singlet and consequently will not be seen in a scan of the $19d\ ^1D_2$ state. The lifetimes were then measured for different F components. No F dependence was found for $18d$ and $20d$, while for the $19d$ state pronounced differences were observed. Possible systematic errors were minimized by performing measurements on the hfs components with equal detected signal strengths, achieved through laser-beam intensity regulation, for constant atomic density. Further, the even-isotope singlet signal and the ^{87}Sr hfs component signals ($^1D_2, ^3D_3$) were measured alternately.

Due to low signal strength the atomic beam could not be sufficiently collimated to reduce the Doppler broadening enough to resolve the $F = \frac{11}{2}$ and $\frac{13}{2}$ components of the singlet state. Therefore, one value is given for these components and it should be interpreted as a weighted mean of the two individual values.

In Fig. 3 two experimental curves can be seen which show the difference in lifetime for the unresolved $F = \frac{11}{2}$ and $\frac{13}{2}$ peak, and the $F = \frac{9}{2}$ peak for ^{87}Sr . All components belong to the $19d\ ^1D_2$ state. The measured lifetime values for selected hyperfine components and the two ^{88}Sr values are compiled in Table I.

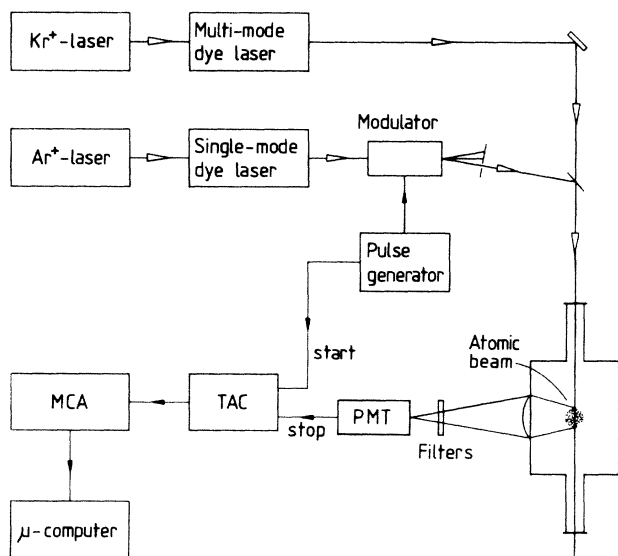


FIG. 2. Experimental setup for the PUMOLS measurements.

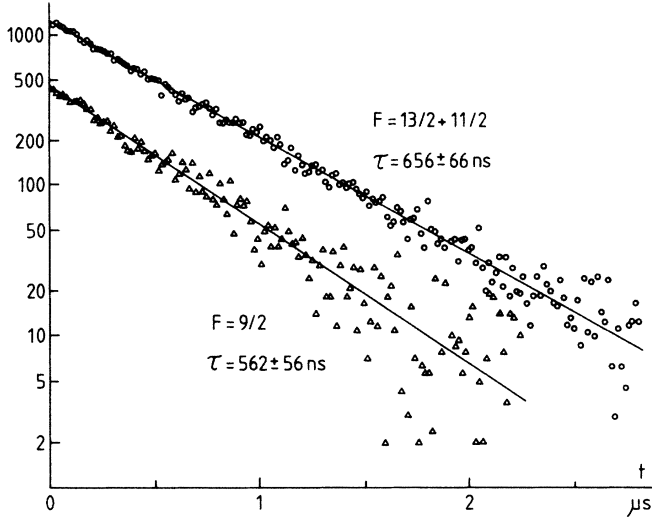


FIG. 3. Experimental decay curves for two hyperfine peaks, belonging to the $19d\ ^1D_2$ state, shown on a \log_{10} -linear plot with the best fit (drawn line). Background counts have been subtracted.

The wave functions Ψ for the odd-isotope hfs component with a specific F number can be written approximately as linear combinations of the unperturbed wave functions $\bar{\Psi}$ for the even isotopes

$$\Psi(^1D_2'', F) = a_F \bar{\Psi}(^1D_2) + b_F \bar{\Psi}(^3D_3),$$

$$\Psi(^3D_3'', F) = -b_F \bar{\Psi}(^1D_2) + a_F \bar{\Psi}(^3D_3).$$

When calculating transition probabilities from these mixed states to a given lower state $\bar{\Psi}_0$, one obtains cross terms for constructive and destructive interference. When

TABLE I. Observed lifetimes.

| Isotope | State | hfs level | τ (ns) |
|------------------|-------------|---|--------------|
| ^{88}Sr | 1D_2 | | 766 ± 77 |
| | 3D_2 | | 395 ± 79 |
| ^{87}Sr | " 1D_2 " | $(F = \frac{13}{2}) + (F = \frac{11}{2})$ | 656 ± 66 |
| | | $F = \frac{9}{2}$ | 562 ± 56 |
| | " 3D_2 " | $F = \frac{5}{2}$ | 652 ± 65 |
| | | $F = \frac{7}{2}$ | 583 ± 58 |

lifetimes are calculated, however, summation over the F components of the lower level is carried out and the interference terms cancel. For the radiative decay rates Γ one obtains

$$\Gamma(^1D_2'', F) = a_F^2 \bar{\Gamma}(^1D_2) + b_F^2 \bar{\Gamma}(^3D_3),$$

$$\Gamma(^3D_3'', F) = b_F^2 \bar{\Gamma}(^1D_2) + a_F^2 \bar{\Gamma}(^3D_3).$$

Here $\bar{\Gamma}$ denotes the decay rate of the corresponding pure states in the even isotopes. If the radiative lifetimes of the 1D_2 and 3D_3 states are different in the even isotopes, as was found in ^{88}Sr , one will obtain, for the mixed states, decay rates that depend on the hyperfine-induced mixing coefficients a_F and b_F .

To obtain the mixing coefficients the Fermi contact term $W = a_{5s} \mathbf{I} \cdot \mathbf{s}$ was added to the total Hamiltonian $H = H_0 + W$. The energy matrix, which breaks up into 2×2 submatrices, one for each F value (except for $F = \frac{3}{2}$ and $\frac{15}{2}$, which do not mix, since there are no such states for 1D_2), was then diagonalized. In a purely LS -coupled 1D_2 state there will be no hyperfine splitting.¹⁴ For Sr,

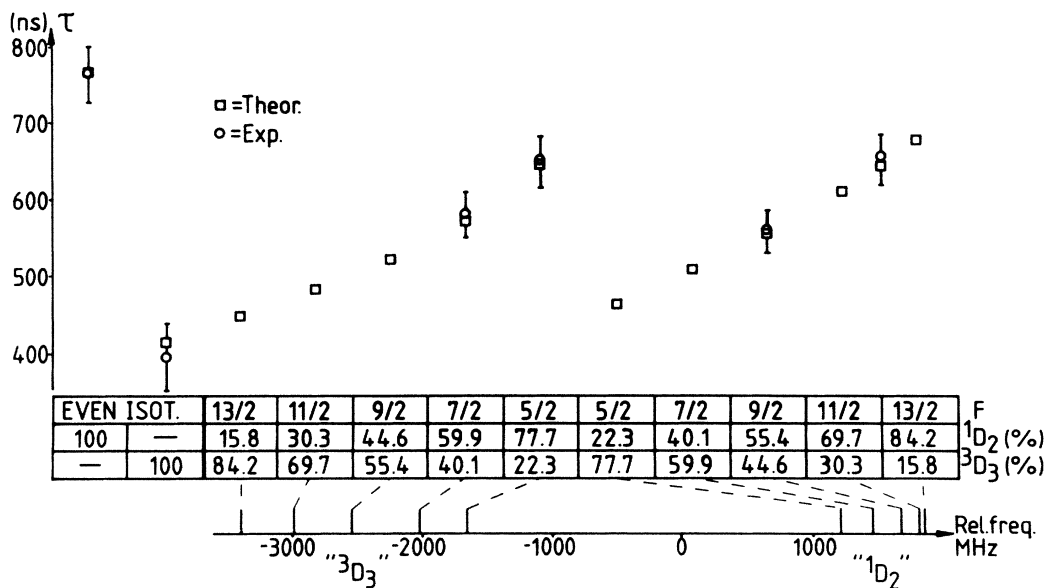


FIG. 4. Comparison between the theoretical and experimental lifetime results. The eigenvector composition is shown at the bottom of the figure, together with a schematic representation of the relative positions for the hyperfine components. The energy structure is taken from Ref. 9.

however, intermediate coupling exists. Before setting up the energy matrix the wave function for the 1D_2 state therefore has to be expanded in wave functions for purely *LS*-coupled states; $^3D_2^o$ and $^1D_2^o$.¹⁴

$$\Psi(^1D_2) = \alpha' \Psi(^3D_2^o) + \beta' \Psi(^1D_2^o).$$

Values for α' and β' are found in the literature.^{10,15}

The mixing coefficients and energy eigenvalues depend critically on the separation between the centers of gravity of the unperturbed 3D_3 and 1D_2 levels. We used the value $\Delta E = 1.78$ GHz found by Beigang *et al.*¹⁰ when studying the energy structures.

Figure 4 shows the theoretical and experimental lifetime results. As can be seen, good agreement between the values is found. The squares of the calculated hyperfine-

induced mixing coefficients a_F and b_F are also indicated in the figure. From these numbers it can be deduced that the designation of the $F = \frac{5}{2}$ and $\frac{7}{2}$ states, labeled " 1D_2 " and " 3D_3 " in the figure, should really be interchanged if the states are named according to dominant eigenvector components.

In conclusion, we have for the first time experimentally shown and theoretically interpreted how *F*-dependent lifetime values can occur as a result of strong hyperfine-induced level mixing. The observations give further insight into hyperfine-induced perturbations, that have so far only been studied using hfs level shifts.

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