Energy-level shift of $5sns\ ^3S_1$ Rydberg states of ^{87}Sr

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Doppler-free two-photon spectroscopy was applied to excite hyperfine-induced transitions from the $5s^2\ ^1S_0$ ground state to the $F = I$ component of the hyperfine multiplet of the $5sns\ ^3S_1$ Rydberg series of ^{87}Sr ($n = 50, \ldots, 85$). The experimental data confirm the interpretation of hyperfine-induced singlet-triplet mixing which is related to the singlet-triplet energy separation.

Hyperfine-induced singlet-triplet mixing in two-electron systems gives rise to significant modifications of hyperfine structure and isotope shifts. This was demonstrated in the case of ^{3}He for $ns\ ^3S$ ($n = 4$–6) and $nd\ ^3D$ ($n = 3$–6) levels.[1-4] As the amount of singlet-triplet mixing increases rapidly with increasing quantum number n,[3] singlet-triplet mixing is of critical importance, in particular, for the character of high Rydberg states.[1]

The influence of this hyperfine interaction in high Rydberg states ($10 \leqslant n \leqslant 70$) of alkaline earth atoms was observed in a recent paper.[5] For the odd-mass isotopes ^{43}Ca, ^{87}Sr, and ^{135,137}Ba an additional level shift was found depending systematically on the decreasing singlet-triplet energy difference for high principal quantum numbers.

In this Brief Report we present new results for transitions to the $F = I$ component of the hyperfine multiplet of the $5sns\ ^3S_1$ Rydberg states of ^{87}Sr excited by Doppler-free two-photon transitions from the $5s^2\ ^1S_0$ ground state. These transition probabilities were induced by hyperfine interaction between the two terms of the $5sns$ configuration. The data shown here confirm the earlier presented calculations (Ref. 5 and references therein) for the hyperfine-induced singlet-triplet mixing of high Rydberg states in two-electron systems. The excitation of these states allows for the observation of hyperfine structure variations, if one electron is systematically removed from the atomic core.

Doppler-free two-photon spectroscopy was used to investigate $5sns\ ^1S_0$ and $^3S_1$ Rydberg levels from the $5s^2\ ^1S_0$ ground state. A cw dye ring laser with up to 200-mW frequency-stabilized single-mode output power in the blue spectral range and a thermionic detection were applied. Information about the experimental setup is given in Ref. 5. In order to allow for the observation of very high Rydberg states, the excitation zone of the thermionic diode was electrically shielded from the detection zone[6] thus reducing the influence of Stark shifts and broadening. A reduction of Stark broadening is required to obtain narrow ionization signals for high principal quantum numbers. In addition, a Faraday rotator was used as

an optical isolator between dye laser and thermionic diode to get the optimum overlap of the two counter-propagating beams without disturbing the single-mode characteristics of the dye laser. In this way a high detection sensitivity was achieved and transitions to Rydberg states with a principal quantum number up to $n = 85$ were detected.

In two-electron systems the $msns$ configuration can form $^1S_0$ and $^3S_1$ terms. From the $5s^2\ ^1S_0$ ground state it is only possible to excite transitions to the $5sns\ ^1S_0$ Rydberg states via two-photon excitation for terms in pure Russel-Saunders coupling. This is shown in the right-hand side of Fig. 1, where the $5s61s\ ^1S_0$ Rydberg state is displayed. The stable Sr isotopes ^{86,87,88}Sr are clearly resolved and the additional energy shift of the odd isotope with a nuclear spin $I = \frac{9}{2}$ can be seen. This shift is caused by the hyperfine interaction between the $F = I$ component of the $^3S_1$ hyperfine multiplet and the $^1S_0$ level with a total angular momentum $F = I$. Both states repel each other if the energy separation becomes comparable to the Fermi contact term as already described in Ref. 5.

For high principal quantum numbers the interaction becomes strong enough, so that a considerable amount of singlet character is mixed into the $^3S_1$ ($F = I$) wave function. In this case the transition probability to the $^1S_0$ states should decrease and transitions to $^3S_1$ ($F = I$) components become possible. Such hyperfine-induced transition probabilities have been observed for $5s19d\ ^1D_2$ and $^3D_3$ Rydberg levels in ^{87}Sr where a local fine-structure degeneracy leads to a strong hyperfine interaction.[7] Both effects which were not considered in Ref. 5 are also shown in Fig. 1. The intensity of the ionization signal is proportional to the isotopic abundancy. Therefore, the ratio between the ionization signals of ^{86}Sr (9.86%) and ^{87}Sr (7.02%) should be 1:0.71. It is obvious that this is not the case for the $5s61s\ ^1S_0$ state where the ^{87}Sr component is too small. The missing intensity can be found in the ionization signal of the $^3S_1$ ($F = I$) component which is shown on the low-energy side of Fig. 1. The intensity is in agreement with calculations of the amount of mixing between the two states. In
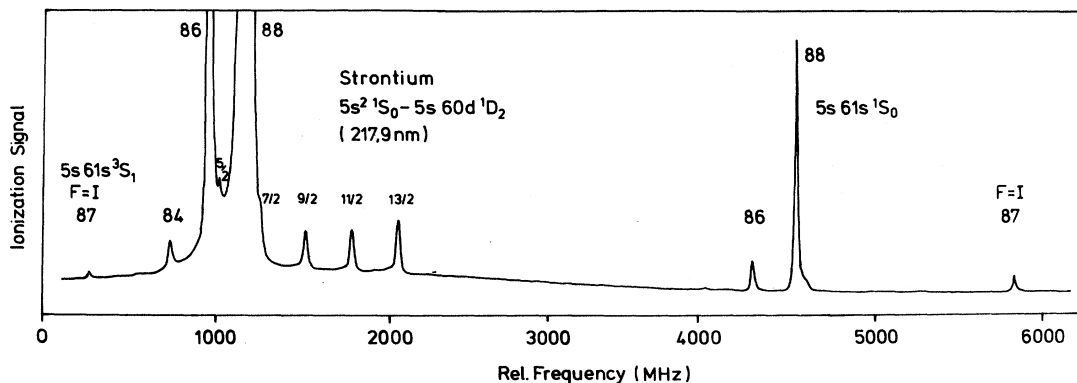
FIG. 1. Doppler-free two-photon excitation spectra from $5s^2\,{}^1S_0$ ground state into $5s\,60d\,{}^1D_2$ and $5s\,61s\,{}^1S_0$ and ${}^3S_1$ $(F = I)$ Rydberg levels. Signals due to even isotopes are labeled by mass number, while hyperfine components of ^{87}Sr are marked by their F values.

principal, both the intensity ratio and the frequency shift for both series can be used to determine hyperfine-induced singlet-triplet mixing. For the calculation of the mixing ratio the perturbed wave functions are taken as a linear combination of pure Russel-Saunders coupled singlet and triplet wave functions:

$$\Psi_1 = |\Psi({}^1S_0)\rangle_{F=I}$$
$$= \alpha|\Psi({}^1S_0^0)\rangle_{F=I} - \beta|\Psi({}^3S_1^0)\rangle_{F=I} \ , \qquad (1)$$

$$\Psi_3 = |\Psi({}^3S_1)\rangle_{F=I}$$
$$= \beta|\Psi({}^1S_0^0)\rangle_{F=I} + \alpha|\Psi({}^3S_1^0)\rangle_{F=I} \ ,$$

where $\alpha^2 + \beta^2 = 1$.

The Hamiltonian has to be extended by the Fermi contact term

$$W = \text{const}\,\vec{I}\cdot\vec{S} \text{ to } H = H_0 + W \ .$$

Solving the secular equation

$$\det \begin{vmatrix} E_1 + \langle\Psi_1|W|\Psi_1\rangle - \tilde{E} & \langle\Psi_1|W|\Psi_3\rangle \\ \langle\Psi_3|W|\Psi_1\rangle & E_3 + \langle\Psi_3|W|\Psi_3\rangle - \tilde{E} \end{vmatrix} = 0 \qquad (2)$$

results in the perturbed level energies $\tilde{E}$ and mixing coefficients α and β. The matrix elements are taken from Ref. 8. The mixing ratio between singlet and triplet states is directly proportional to the relative transition probability to ${}^1S_0$ and ${}^3S_1$ $(F = I)$ levels.

The calculated admixtures α^2 and β^2 are shown in the upper part of Fig. 2. At $n = 61$ the admixture of singlet into triplet wave functions is already $\sim 20\%$; at $n \sim 115$ the same intensities are expected for transitions to ${}^1S_0$ and ${}^3S_1$ $(F = I)$ levels. When reaching the ionization limit the mixing ratio levels off at a ratio of 4:5 which is the ratio of the relative intensities of the $F = I - \frac{1}{2}$ and $F = I + \frac{1}{2}$ hyperfine components

of the ${}^2S_{1/2}$ ground state of Sr$^+$.

The lower part of Fig. 2 compares the additional energy shift of ${}^1S_0$ and ${}^3S_1$ $(F = I)$ Rydberg levels as calculated from Eq. (2) with experimental data obtained for ${}^1S_0$ and ${}^3S_1$ $(F = I)$ levels. For the calculation the singlet-triplet separation was determined using level energies for the ${}^1S_0$ series measured with an accuracy of $+0.001$ cm^{-1} (Ref. 9) and calculated energies for the ${}^3S_1$ series with a quantum defect $\delta_3 = 3.386$ as observed in a pulse laser experiment.[10]

The experimental energy shift of the $5sns\,{}^1S_0$ series can be deduced directly from the spectra (see also Fig. 4 in Ref. 5). The shift of the ${}^3S_1$ $(F = I)$ levels,

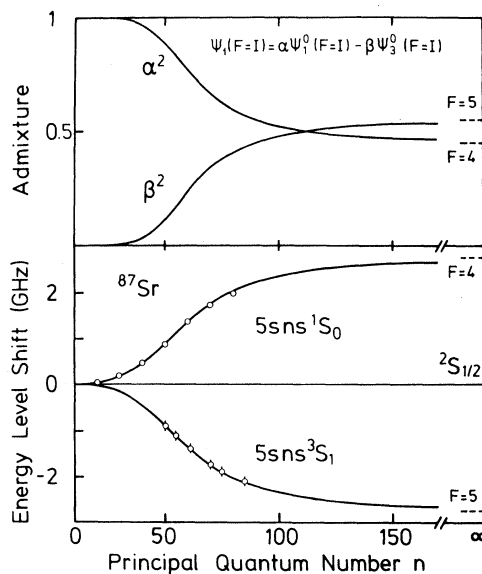FIG. 2. (a) Calculated admixtures α^2 and β^2 as a function of n. (b) Comparison between calculated energy shifts of ${}^1S_0$ and ${}^3S_1$ $(F = I)$ Rydberg levels and experimental data (open circles).

however, cannot be determined directly as no transition to the $^3S_1$ levels of even Sr isotopes are allowed with two photons from the $5s^2\,{}^1S_0$ ground state. Therefore, the frequency difference between transitions to the $^1S_0$ and $^3S_1$ $(F = I)$ levels with the same principal quantum number n was measured. After subtracting the energy difference between singlet and triplet states the resulting frequency value represents the sum of the energy shift of $^1S_0$ and $^3S_1$ $(F = I)$. Due to the uncertainty of the singlet-triplet difference these data show slightly larger error bars as can be seen in Fig. 2. But there is still very good agreement with the theoretical data.

The energy shift of the $F = I$ component of the $5sns\,{}^3S_1$ hyperfine multiplet results also in a strong violation of the Landé interval rule. The $F = I - 1$ and $I + 1$ components experience no perturbation and converge with increasing n to the $F = I - \frac{1}{2}$ and $I + \frac{1}{2}$ hyperfine components of the $^2S_{1/2}$ ion ground state without changing their energy separation. The $F = I$ component, however, ends in the lower $^2S_{1/2}$ hyperfine component which causes for high principal quantum numbers a deviation from the interval rule as shown in Fig. 3. Similar deviations have been observed for $^3D_1$ and $^1D_2$ states of Hg[11-13] and recently for high Rydberg states of Yb[14] and Ba.[15]

The experimental data presented in this Brief Report confirm the earlier observations and interpretations of hyperfine-induced energy shifts in high Rydberg states of alkaline earth elements.[5] The decreasing transition probability to members of the $^1S_0$ series
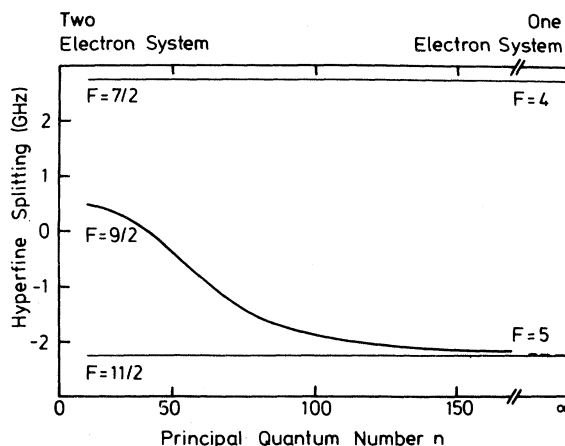

FIG. 3. Calculated variation of the $^3S_1$ hyperfine multiplet as a function of principal quantum number n, leading to the hyperfine multiplet of a one-electron system.

for high principal quantum numbers is in agreement with calculations. The observation of transitions to the $^3S_1$ $(F = I)$ Rydberg levels with increasing intensity for increasing principal quantum numbers n also supports our theoretical assumptions.

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