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On the anomalous low spontaneous emission rates for p-series of sodium due to the effect of natural Förster resonance

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We are concerned here with a possibility of dipole matrix element strong blockade for excited atoms in the vicinity of Förster resonance. The latter occurs when the difference $\Delta\delta = \delta_{l'} - \delta_l$ between the quantum defects of l- and ($l' = l \pm 1$)-adjacent series becomes equal to 0.5. The comparison between lifetimes of Rydberg p-states in alkaline atoms indicates anomaly large (fivefold) values in the case of sodium. We explain this fact with the configuration of p- and s-series in Na which practically meets the Förster resonance realization requirement. Important specific futures of the Förster resonance discussed here and related to radiative kinetics of Rydberg states are of interest for interpretation of fluorescence spectra observed in astrophysics.

Keywords: Spectra, Na emission, blockade, external fields

1 Introduction

The effects of Förster resonance or the double Stark resonance (Förster, 1948; Walker & Saffman, 2005) (hereinafter, DSR) in quantum systems are beginning to find applications in quantum information devices (Reetz-Lamour et al., 2006) and in control of cold atoms with a laser field (Walker & Saffman, 2005). The essence of the DSR phenomenon depicted in the atom energy diagram (see Fig. 1) is that the energy of the excited $(n, l+1)$ state of atomic $(l+1)$-series lies exactly in the middle between the energies of the two neighboring states $(n+1, l)$ and (n, l) from the adjacent l-series.

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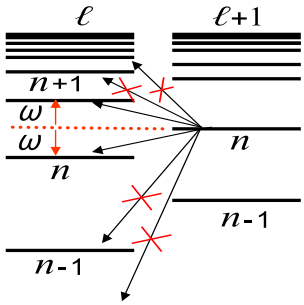


Figure 1 Schematic energy levels diagram for alkali atoms which corresponds to the Förster resonance realization between quantum states belonging to $(l+1)$- and l-adjacent series.

Here n and l are the principal and orbital quantum numbers, respectively. At DSR the potential energy of a diatomic configuration (quasi-molecular) $\{np, np\}$ coincides with the energy of a diatomic system $\{(n+1)\ s,\ n\ s\}$, resulting in their populations sharing due to the long-range dipole–dipole or van der Waals interactions. In terms of quantum information science, this corresponds to the formation of an "entangled" state.

An interesting feature of atomic systems under DSR is a possibility of a significant weakening dipole matrix elements amplitudes for the "long" transitions while maintaining the probability of the "short" transitions (Arefieff et al., 2015) between the quantum states (see Fig. 1). The simplest example exhibits a three-dimensional oscillator of frequency ω which energy levels ε (Sommerfeld, 1934)

$$\varepsilon = \omega(2n_r + l + 3/2), \tag{1}$$

satisfy DSR for all l-series. Here n_r is the radial quantum number for the fixed l-series. Atomic units are used unless stated otherwise. The harmonic oscillator emits radiation at a single frequency corresponding to its natural one ω, that results in a blockade of all optical dipole transitions except for transitions between the neighboring states (see Fig. 1).

As a tool for the implementation of DSR in Rydberg states of alkali atoms one usually uses a Stark shift of p-levels in an external electric field of a few 10^{-9} at.u. (Ryabtsev et al., 2010). Atomic l-series with $l > 2$ are not considered, since for them the energy structure of the excited states becomes similar to that of the hydrogen atom, i.e. the states with the same n do not differ in energies.

2 Radiative kinetics in the vicinity of the double Stark resonance

In the vicinity of DSR we can expect a significant decrease in the constants of radiative processes. Since the probability of a spontaneous emission is directly connected with the dipole matrix elements, locking the latter under DSR conditions should lead to a delay in the spontaneous decay of excited states. This section will analyze the serial pattern (i.e., the dependence on the principal quantum number in the fixed l-series)

of radiative lifetimes for s- and p-series of alkali atoms, the configuration of which lies near the DSR. At the same time, we consider in brief the modeling of the radiative kinetics for model atoms with Sommerfeld potential (Sommerfeld, 1934).

2.1 Radiative lifetimes of Rydberg states

The partial probabilities $A_{nl}^{n'l'}$ of spontaneous transitions (Einstein coefficients) between excited states are important characteristics of Rydberg atoms. For a fixed (n, l)-level the integral probability of all allowed optical transitions to the lower-lying states

$$A_{nl} = 1/\tau_{nl} = \sum_{n'l'} A_{nl}^{n'l'} \tag{2}$$

determines its natural width A_{nl} along with radiative lifetime τ_{nl}. Analysis of serial patterns for the Rydberg states ($n \gg 1$) is conveniently performed using analytical relationships

$$1/\tau_{nl} = \frac{4}{\pi\sqrt{3}c^3} \frac{C_l}{n_{ef}^3 (l + 0.5)^2} \tag{3}$$

found in Bezuglov et al. (1991) within the frame of one-channel quantum defect treaty. Here $n_{ef} = n - \delta_l$ is the effective quantum number. The coefficient C_l is a constant for a given l-series and obviously determines the efficiency of radiative processes. Table 1 shows the data on C_l obtained in Bezuglov et al. (1991) for s-, p- and d-series. The comparison between C_{l-1} values for Rydberg p-states in alkaline atoms indicates their anomalously small values in the case of sodium.

Table 1 C_l coefficient values for s-, p-series of alkali atoms (Bezuglov et al., 1991).

Atom	s-series	p-series	d-series
H I	0.012	1.00	1.00
Li I	0.025	0.069	1.12
Na I	0.016	0.014	0.55
K I	0.017	0.051	0.18
Rb I	0.017	0.075	0.51
Cs I	0.018	0.061	0.82

From the presented results we can infer the existence of some interconfiguration interaction mechanism differently revealing itself in different alkali atoms. It is possible to specify a manifestation of DSR as one of these mechanisms resulting in the partial blockade of optical transitions in Na. Especially that the difference $\Delta\delta_{SP}$ between the quantum defects of s- and p-sodium series is 0.493 (Zimmerman et al., 1979), i.e. it is less than the "magical" number of 0.500 needed for the implementation of DSR by the value of 0.007 only. Changes in the values of both the effective quantum number n_{ef} and the serial difference $\Delta\delta$ between quantum defects upon increasing the radial quantum number n_r is given in Table 2 for s-, p-, d-states of Na. It is well seen that the configuration of the Rydberg states in s- and p-series almost completely meets the requirement of DSR realization.

For comparison, Table 3 shows the data (Hezel et al., 1992) for quantum defects of Rydberg states for s-, p-, d-series along with their difference for all alkali atoms.

Table 2 Values of the effective quantum number n_{ef}, and the difference $\Delta\delta$ between the quantum defects in the case of Na (Gontis & Kaulakys, 1987).

n_r	$l=0$ n_{ef}	$l=1$ n_{ef}	$l=2$ n_{ef}	$\Delta\delta_{SP}$	$\Delta\delta_{DP}$
1	2.643	3.133	3.988	0.490	0.855
5	6.651	7.133	7.986	0.492	0.843
10	11.651	12.135	12.988	0.494	0.843
15	16.651	17.147	17.988	0.496	0.841
19	20.651	21.147	21.988	0.498	0.841

Our interpretation of the anomalously large values of radiative lifetimes is associated with an almost exact DSR implementation in the case of sodium.

Table 3 Qantum defect δ_l for s-, p-series of alkali and hydrogen atoms (Hezel et al., 1992).

	Li	Na	K	Rb	Cs	H
s	0.40	1.35	2.19	3.13	4.06	0
p	0.04	0.85	1.71	2.66	3.59	0
$\Delta\delta_{SP}$	0.36	0.50	0.48	0.47	0.47	0

An interest in the fluorescence spectrum of sodium, incipient in astrophysics at the first analysis of the comets luminosity (Levin, 1964), increased after the discovery of the sodium cloud-nebulas in the vicinity of Jupiter's Galilean moons – Io and Europa, as well as Jupiter itself (Matson et al., 1978; Leblanc et al., 2005).

2.2 Sommerfeld atom

It is useful to note an interesting fact of relatively small (4–6%) s- and p-series detuning of DSR in terms of the quantum defects difference $\Delta\delta_{SP}$ for practically all alkali atoms (see Table 3). This, however, leads to a large (fivefold) reduction of radiative lifetimes τ_{nl} values compared to the "resonance" case of Na atom. A similar sharp dependence of radiative rate constants on parameters of atomic systems in the vicinity of DSR was revealed in our recent paper (Arefieff et al., 2015) where we have taken as an example one-electron model atom with the Sommerfeld potential (Sommerfeld, 1934)

$$U_\alpha = -\frac{1}{r} + \frac{\alpha}{2r^2}. \tag{4}$$

Here α is the parameter of the problem. Such an option for the centrally symmetric atomic potential has been proposed in Sommerfeld (1934) for calculations of relativistic corrections in the hydrogen atom, but has worked well in calculations of probabilities of radiative transitions in alkali atoms under the framework of the so-called approximation of the effective orbital quantum number l_{ef} (Grouzdev, 1982). A distinctive feature of any treaty dealing with the potential (2), is the possibility of an analytical description of quantized energy values along with explicit expressions of wave functions via well-studied and widely used in quantum theory hypergeometric

functions (Landau & Lifshitz, 1977). It is possible, in particular, to describe simply the effect of DSR between adjacent $\{l,\ l-1\}$ series with a suitable choice of the parameter $\alpha = \alpha_l = 3(l^2 - 1/16)$. The results obtained in this way in Arefieff et al. (2015) demonstrate a significant decrease in the partial probabilities $A_{nl}^{n'l'}$ of radiative transitions in the vicinity of the DSR, which is consistent with the effect of blocking the dipole matrix elements, based on an analogy with 3D oscillator.

We should note a drawback of work by Arefieff et al. (2015) due to the use of a simplified one-parameter amendment to the Coulomb potential simultaneously for all quantum states. Strictly speaking, in the real many-electron atom an optical (valence) electron, belonging to different atomic series, suffers different interactions with the core electrons, that should be modeled by the potential (4) with a variety of the parameter α values.

3 Modeling radiative lifetimes under the approximation of effective orbital quantum number

There is a convenient method (see, e.g., Grouzdev (1982)) to find a specific parameter α_{nl} value for the considered atomic (n, l)-state using experimental data on its energy ε_{nl}. Namely, the radial motion of the valence electron is determined by the effective potential

$$U_{ef} = -\frac{1}{r} + \frac{l(l+0.5)+\alpha}{2r^2}, \tag{5}$$

where the total coefficient of the second (centrifugal) member is expressed through the effective orbital quantum number: $l(l+0.5)+\alpha = l_{ef}(l_{ef}+0.5)$. Both the energy spectrum

$$\varepsilon_{nl} = -1/2n_{ef}^2, \quad n_{ef} = n_r + l_{ef} + 1, \tag{6}$$

and the corresponding radial wave functions for the atomic potential (5), are well known (Landau & Lifshitz, 1977). It becomes possible, thus, to find the probabilities $A_{nl}^{n'l'}$ of optical transitions, depending on the variations of the quantum defect δ_p for p-series at constant values δ_s within the s-series. In calculating the spontaneous decay rates (2), we used the data on the quantum defects (Gontis & Kaulakys, 1987) (see

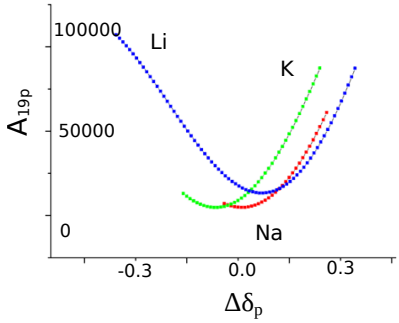


Figure 2 Natural width A_{nl} [s^{-1}] of 19p-state ($l = 1$, $n = 19$) for Li, Na, K in varying the quantum defect δ_p of p-state.

also Table 2). Figure 2 represents the natural width A_{nl} of 19p-state in Li, Na, K as a function on the quantum defect variation $\Delta\delta_p$ of the initial excited 19p-state. Figure curves indicate clearly that A_{nl} minimum occurs at a practically unshifted ($\Delta\delta_p = 0$) value of the quantum defect in the only case of sodium atom. This fact is the direct consequence of the discussed above s- and p-series features in Na that has the Rydberg states configuration the most exact matching the DSR requirement $\Delta\delta_{SP} = 0.5$.

4 Conclusion

It is shown that the three level structure of s-, p-series in an excited sodium atom turns out to be similar to that of 3D quantum oscillator, which is characterized by suppression of the "long" dipole transitions. This feature explains, first of all, the known "quasi-stability" of Rydberg sodium p-states described in the literature (Bezuglov et al., 1991). The other characteristic feature of sodium Rydberg states is concerned with a phenomenon of the double Stark (or Förster) resonance. The latter is also demonstrated to be well correlated with the radiative processes blockade. At the same time, the above examples show the possibility of the influence of environmental parameters on the excited atom characteristics and, in particular, on the control of Rydberg atoms/molecules fluorescence spectra in the Earth's upper atmosphere (Golubkov, 2011) and in astrophysical objects.

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