**Problem Set # 5**

**DUE:** At the start of Lecture on Monday, April 5.

**Reading:** HLB–RWF 3.2.1 (optional), 9.1.1, 9.4.1 - 9.4.3.

**Problems:**

1. **Photon Plucks**

   The first excited state of Ba is the metastable $6s5d\,^3D$ state. Consider the $J = M_J = 3$ component of $^3D$

   $$|^3D_3\rangle = |s0\alpha\ d2\alpha\rangle.$$

   A photon pluck promotes one $e^-$ via a $\Delta \ell = \pm 1, \Delta m_\ell = 0, \pm 1, \Delta s = \Delta m_s = 0$ transition. If the photon is linearly polarized along the $z$–direction, the selection rule is $\Delta m_\ell = 0$.

   **A.** What are all of the $\ell_1 m_\ell_1 \ s_1 m_s_1 \ \ell_2 m_\ell_2 \ s_2 m_s_2$ basis states accessible via a $z$–polarized photon pluck?

   **B.** Consider first only the $5d \to np$ excitation to $6snp$ Rydberg complexes. What $L$–$S$–$J$–$M_J$ eigenstates are coherently populated at $t = 0$? What are their relative amplitudes?

   **C.** Now consider the $6s \to n'p \ (n' \neq n)$ excitation to doubly excited $5dn'p$ configurations. What $L$–$S$–$J$–$M_J$ eigenstates are coherently populated at $t = 0$? What are their relative amplitudes?

   **D.** The only things not specified are the relative amplitudes within the $6snp$ Rydberg series, within the $5dn'p$ Rydberg series, and for the $6snp$ series relative to the $5dn'p$ series. These are given by the radial integrals

   $$\mu_{n'p5d} = (6p|5d\rangle \ (n'/6)^{-3/2}$$

   $$\mu_{np6s} = (6p|6s\rangle \ (n/6)^{-3/2}$$

   What is the form of the $\Psi(t)$ that results from this single-photon pluck of Ba $|6s\ 5d\ ^3D_3\rangle$?

   I have not provided sufficient information about eigen-energies, photon center-frequency, and photon pulse duration. You should make reasonable choices for these quantities.

   **E.** The ionization threshold for the $6sn\ell$ Rydberg series ($n \to \infty$) is 42032·4 cm$^{-1}$, which is considerably lower than that for the $5dn'\ell$ Rydberg series ($n' \to \infty$) at 46906·3 cm$^{-1}$ (for $J = 3/2$). Suppose you probe the coherent superposition state from part D with a detection pulse at an energy just above the $6sn\ell$ limit but below the $5dn'\ell$ limit. You detect photo-ions as a function of delay between the excitation and detection pulses. What will you see? Which coherences will be detected by this pump/probe scheme and which coherences will be destroyed?

   **F.** The $^3P$ states of the $n'p5d$ and $np6s$ Rydberg series interact with each other via the $1/r_{12}$ interelectronic repulsion operator. The Kepler period of a Rydberg wavepacket is proportional to $[\langle n^{-3}\rangle]^{-1}$. Owing to the difference in energy of the series limits, the *isoenergetic* members of the $n'p5d$ and $np6s$ series have $n' < n$, thus the Kepler period of the $n'p5d$ wavepacket is shorter than that of the $np6s$ wavepacket. The interaction between the two wavepackets is largest when both are inside the ion-core. If you could monitor the amplitude in the $np6s$ wavepacket as a function of time, what would you expect to see?
Atomic Hyperfine Structure

The $H^{\text{eff}}$ for $^{137}\text{Ba}$ ($I = 3/2$) is

$$H = H^{\text{el}} + H^{\text{SO}} + H^{\text{mhf}}.$$  

For Rydberg series ($\delta_\ell$ is the quantum defect, $R = 109,737 \text{ cm}^{-1}$)

$$H^{\text{el}} = -\left[ R/ (n - \delta_\ell)^2 \right] |n\ell n'\ell'\rangle \langle n\ell n'\ell'|.$$  

For two-electron atoms

$$H^{\text{SO}} = \xi(r_1)\ell_1 \cdot s_1 + \xi(r_2)\ell_2 \cdot s_2.$$  

For magnetic hyperfine structure of a two-electron atom

$$H^{\text{mhf}} = [a(r_1)\ell_1 + a(r_2)\ell_2 + b(r_1)s_1 + b(r_2)s_2] \cdot I.$$  

Under special conditions (to be specified by you), $H^{\text{SO}}$ and $H^{\text{mhf}}$ simplify to

$$H^{\text{SO}} = \zeta(N, L, S) L \cdot S$$
$$H^{\text{mhf}} = C(N, L, S, J) I \cdot J$$

where $N$ refers to the electronic configuration. These simplified forms of $H^{\text{SO}}$ and $H^{\text{mhf}}$ are useful for depicting the pattern of splittings within an “isolated” state. Electronic transitions are controlled by

$$\langle n_\ell_1 \ell_1 m_\ell_1 |\hat{x} \hat{y} \hat{z}| n_\ell_1' \ell_1' m_\ell_1' \rangle,$$

which is a matrix element of a one-electron operator that operates exclusively on the spatial (not spin) part of a single spin-orbital.

A. Use the simplified forms of $H^{\text{SO}}$ and $H^{\text{mhf}}$ to construct the spin-hyperfine structure of $^{137}\text{Ba}$ in the $6s5d \, 3D$ and $6snp \, 3P$ states.

**HINT:** $$(L + S)^2 = J^2$$
$$(J + I)^2 = F^2$$

B. Starting from the $^{137}\text{Ba} \, 6s5d \, 3D_3$ level (with hyperfine $F$–components $F = 3/2, 5/2, 7/2, \text{and} \ 9/2$ thermally populated), draw a level diagram on which you illustrate all of the allowed fine-hyperfine transitions from $3D_3$ to $3P_{J,F}$. The rigorous selection rule for electric dipole transitions (a vector operator) is $\Delta F = 0, \pm 1$. The nearly rigorous selection rules $\Delta J = 0, \pm 1$, $\Delta L = 0, \pm 1$, and $\Delta S = 0$ may also be taken seriously here.

C. In order to derive the fine-hyperfine quantum beat signal obtained by pulsed excitation of $3P_{J',F'} \leftrightarrow 3D_{J=3,F}$ transitions, you need to compute all of the relative transition amplitudes for the short-pulse excitation “pump” transition and for the delay–scanned detection “probe” transition. For simplicity you can use the $6snp \, 3P_{J,F} \rightarrow 6s6d \, 3D_{3,F}$ probe transition. In order to calculate the relative transition amplitudes in the $|s_1s_2\ell_1\ell_2LSJIF_M F\rangle$ basis set, you must perform a series of coupled→uncoupled transitions: $|s_1s_2\ell_1\ell_2LSJIF_M F\rangle$.
\[ |s_1 s_2 \ell_1 \ell_2 L S J M J I M I \rangle \]
\[ |s_1 s_2 \ell_1 \ell_2 L M L S M S I M I \rangle \]
\[ |\ell_1 m_\ell_1 \ell_2 m_\ell_2 s_1 s_2 S M S I M I \rangle \]
\[ |\ell_1 m_\ell_1 s_1 m_\ell_1 \ell_2 m_\ell_2 s_2 m_\ell_2 s_2 M S I M I \rangle . \]

All of the beat notes in your quantum beating signal are explicitly known half-integer multiples of a common factor with relative amplitudes controlled by \( \langle 6d|z|np \rangle \langle np|z|5d \rangle \) times factors computed by you. Would the relative intensities and phases of the beat notes be affected if the pump and probe lasers were polarized perpendicularly (i.e., \( x,z \)) rather than parallel (i.e., \( z,z \))? Optional: compute and compare the beat patterns for probe (\( z \)), pump (\( z \)) to probe (\( x \)), pump (\( z \)).

D. As the principal quantum number of the \( 6snp \) \( ^3P \) state increases, the spin-orbit coupling constant decreases as \( n^{-3} \) but the contribution of the \( 6sm_s \) spin-orbital to the \( b(r_1)s_1 \cdot I \) hyperfine term remains constant. At some point the hyperfine splittings become larger than the spin-orbit splittings. What happens to the level structure and quantum beat amplitudes? A qualitative answer is acceptable. **HINT:** The hyperfine structure of \( Ba^+ \) \( 6s^2S \) is highly relevant here.