On the efficiency of laser pumping of hyperfine structure sublevels of Rubidium-87 and Cesium-133 atoms

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Abstract. The evolution of the hyperfine structure magnetic sublevel populations of $^{87}$Rb and $^{133}$Cs atoms in resonant linearly polarized laser fields is theoretically analyzed. Analytical expressions are obtained for stationary populations of the magnetic sublevels of the ground state $^{5}S_{1/2} F_{g}=1, 2$ in $^{87}$Rb and $^{6}S_{1/2} F_{g}=3, 4$ in $^{133}$Cs under optical pumping at the $F_{g} \leftrightarrow F_{e}=F_{g}$ and $F_{g} \leftrightarrow F_{e}=F_{e}$ transitions of the $D_{2}$ lines ($n^{2}S_{1/2} - n^{2}P_{1/2}$) versus the initial populations. The characteristic population stabilization time are obtained by numerical solution for the density matrix equations, depending on the intensity of the laser fields and the detuning of the optical resonance. It is shown that the alternation of the polarization direction of the laser field at the transition $F_{g} \leftrightarrow F_{e}=F_{g}$ almost completely transfers atoms to the lower sublevel of the "clock" M1 transition $F_{g} M=0 \leftrightarrow F_{g} M=0$ in rubidium and cesium frequency standards, which proportionally increases the signal of the recording system.

1. Introduction

At present, the highest accuracy is achieved by the new generation of time and frequency atomic fountain standards [1-5], which based on laser cooling of cesium atoms. In comparison with classical microwave frequency standards based on a beam of thermal cesium atoms [6], the accuracy and stability of independent reproduction of units of time and frequency in atomic fountains is at least an order of magnitude higher due to the minimization of the Doppler shift and amounts to several units of the sixteenth decimal place [3, 4].

A sufficiently high value of the signal-to-noise ratio in the recording system of the frequency standard, which determines the duration of continuous measurements, is proportional to the density of the atomic beam. For cesium fountains, it is necessary to take into account the spin-exchange frequency shift of the "clock" transition, which is also proportional to the density of atoms. Thus, it is required to optimize the relative population of atoms in the initial state of the “clock” transition. A solution of this problem is the use of optical pumping for magnetic sublevels [7]. For the rubidium standard, the restrictions on the spin-exchange frequency shift are less stringent. Different aspects of the optical pumping were studied experimentally and theoretically in $^{87}$Rb [8-14] and Cs [15-24].

In this work, efficiency analysis of the optical pumping of the hyperfine sublevel with the total atomic angular momentum and its projection $^{5}S_{1/2} F_{g}=1 M=0$ in $^{87}$Rb atoms and $^{6}S_{1/2} F_{g}=3 M=0$ in $^{133}$Cs is carried out. The pumping scheme is based on resonant transitions $F_{g}=1 \leftrightarrow F_{e}=1$ and $F_{g}=2 \leftrightarrow F_{e}=2$ in rubidium, and $F_{g}=3 \leftrightarrow F_{e}=3$ and $F_{g}=4 \leftrightarrow F_{e}=4$ in cesium within the $D_{2}$ lines and...
linearly polarized laser fields. For these transitions, explicit asymptotic population values of the "dark" \( M = 0 \) sublevels (unaffected by laser fields) are obtained, which are independent of laser parameters. The time that is needed for the dark-level populations to approach practically their asymptotic values is determined numerically by solving the kinetic equations for the density matrix, depending on the laser frequency detuning from resonance and the radiation intensity.

2. Bichromatic laser field pumping of clock levels

Figure 1 shows schematics of the lowest level hyperfine structure of \(^{87}\text{Rb}\).

![Figure 1](image-url)

For efficiency of an optical pumping in a multilevel system, the ratio of the spontaneous transition probabilities to "dark" states is important. As can be seen from table 1 the D\(_2\) line is preferable for Rb, since the branching coefficients for spontaneous transitions from excited \( F_e M = \pm 1 \) states to the lower \( F_e M = 0 \) sublevel is higher than for the D\(_1\) line. The same is true for a Cs atom.

**Table 1.** Branching coefficients for spontaneous transitions between Zeeman sublevels \( F_e = 1,2M' \rightarrow F_g = 1M, F_f = 2M \) under consideration in D\(_1\) and D\(_2\) lines of \(^{87}\text{Rb}\).

| Line Upper level | \( F_g = 1M \) | \( F_f = 2M \) |
|------------------|----------------|----------------|
|                  | 0              | \( \pm 1 \)    | \( \pm 1 \)    | \( \pm 2 \)    |
| D\(_1\) \( F_e = 1M' = 0 \) | 0            | 1/12           | 1/3            | 1/4            |
| \( M' = \pm 1 \) | 1/12           | 1/12           | 1/12           | 1/4            | 1/2            |
| D\(_2\) \( F_e = 1M' = 0 \) | 0            | 5/12           | 1/15           | 1/20           |
| \( M' = \pm 1 \) | 5/12           | 5/12           | 1/20           | 1/10           |
| D\(_1\), D\(_2\) \( F_e = 2M' = 0 \) | 1/3           | 1/12           | 0              | 1/4            |
| \( M' = \pm 1 \) | 1/4            | 1/4            | 1/12           | 1/6            |
| \( M' = \pm 2 \) | 1/2            | 1/6            | 1/3            |
The transition scheme of simultaneous bichromanic pumping is shown in figure 2. The excited states $F_e, M = 0$ (dashed) are not populated during the process since optical transitions from lower sublevels are forbidden in linear polarized laser field. Note that Zeeman splitting of sublevels by a weak C-field can be neglected in the consideration. From the symmetry the results for positive and negative $M$ are equal.

![Figure 2: Optical pumping scheme of $^{87}$Rb with $F_g = 1 \leftrightarrow F_e = 1$ and $F_f = 2 \leftrightarrow F_e = 2$ transitions in a linear polarized bichromatic laser field (solid arrows). Dashed arrows indicate spontaneous transitions with $M = 0$ “dark” states.](image)

The asymptotic sublevel populations (diagonal density matrix elements $n_{FM} = \rho_{FM, FM}$) for pumping in two simultaneous transitions that do not have common levels \[25\] read for Rb

$$
\begin{align*}
n_{gM=0} &= n_{gM=0} + \sum_{M>0} P_{g0,gM} n_{gM} + \sum_{M>0} P_{g0,0M} n_{0M}, \\
n_{gM=0} &= n_{gM=0} + \sum_{M>0} P_{f0,gM} n_{gM} + \sum_{M>0} P_{f0,0M} n_{0M}.
\end{align*}
$$

(1)

Here the transition matrices $f(g)M \rightarrow eM = 1 \rightarrow g(f)M = 0$ are

$$
P_{g(f)0,f(g)M} = 2b_{f(f)0,e}|(1 - b_{f(g)e})^{-1}|_{M}
$$

(2)

where $I$ is the identity matrix, $b_{g(f)M,eM^*}$ is the branching coefficient of the radiative transition probability between hyperfine structure magnetic sublevels. The total momentum projection quantization axis is collinear both to laser polarization and an atomic beam direction.

From (1) and (2) for $F_g = 1 \leftrightarrow F_e = 1$ and $F_f = 2 \leftrightarrow F_e = 2$ transitions of the line $D_2$ in $^{87}$Rb follow

$$
\begin{align*}
n_{10} &= n_{10}^0 + \frac{46}{25} n_{11}^0 + \frac{34}{25} n_{21}^0 + \frac{43}{25} n_{22}^0, \\
n_{20} &= n_{20}^0 + \frac{4}{25} n_{11}^0 + \frac{16}{25} n_{21}^0 + \frac{7}{25} n_{22}^0.
\end{align*}
$$

(3)

In the case of equilibrium initial populations

$$
n_{10} = 0.74, \quad n_{20} = 0.26.
$$

(4)

Note that for a linear polarization the Zeeman coherencies stay in zero initial values since their equations do not couple to the populations. Thus the resulting state is an incoherent mixture.

In the $^{133}$Cs atom the “dark” sublevels are $F_g = 3M = 0$ and $F_f = 4M = 0$ under the simultaneous action of two laser fields at the transitions $F_g = 3 \leftrightarrow F_e = 3$ and $F_f = 4 \leftrightarrow F_e = 4$ of the D$_2$ line. Their
asymptotic populations have relatively cumbersome form given in [25]. For equilibrium initial populations

\[ n_{30} = \frac{482907}{698782} \approx 0.691, \quad n_{40} = \frac{215875}{698782} \approx 0.309. \]  

(5)

3. Multistage pumping scheme

It follows from (4) and (5) that the pumping produces a mixed state, which is an incoherent sum of the sublevels of the clock transition. To obtain a pure state \( F_gM = 0 \) when using the Ramsey method, it is required to clear the upper sublevel \( F_fM = 0 \). This can be achieved without the loss of atoms in a beam by applying repeated pumping at the transition \( F_f = 2 \leftrightarrow F_e = 2 \) in an alternative field polarized direction at angle \( \beta \) (for certainty by rotating around \( y \)-axis) respect to the quantization axis.

In the correspondingly rotated reference frame with a new \( z' \)-axis, the asymptotic populations for the monochromatic pumping of \(^{87}\text{Rb} \) atoms have the form

\[
\begin{align*}
n_{10}(\beta) &= n_{10}^0(\beta) + \frac{4}{7} n_{21}^0(\beta) + \frac{1}{7} n_{22}^0(\beta), \\
n_{11}(\beta) &= n_{11}^0(\beta) + \frac{3}{7} n_{21}^0(\beta) + \frac{6}{7} n_{22}^0(\beta), \\
n_{20}(\beta) &= n_{20}^0(\beta) + \frac{4}{7} n_{21}^0(\beta) + \frac{1}{7} n_{22}^0(\beta),
\end{align*}
\]

(6)

where the initial values of the sublevel populations with the quantization axis \( z' \) in the rotated coordinate system are determined by values in (4) and by the Wigner’s \( D \)-functions (see, for example, [26])

\[
\rho_{FM,FM}^0(\beta) = \sum_{m} d_{FM}^F d_{FM}^M (\beta) \rho_{FM,FM}^0 d_{FM}^F \rho_{FM,FM}^M (\beta).
\]

(7)

It follows from (6) that the pumping transmits a significant fraction of atoms from states \( F_f = 2M \) to \( F_e = 1M \).

Note that for a \( \sigma \) polarization \( (\beta = 90^\circ) \) (4) and (7) give the following nonzero density matrix elements in the rotated frame

\[
\begin{align*}
n_{20}^0(90^\circ) &= \frac{1}{4} n_{20}, \\
n_{22}^0(90^\circ) &= \frac{3}{8} n_{20}, \\
\rho_{20,22}(90^\circ) &= \frac{\sqrt{3}}{4\sqrt{2}} n_{20}, \\
\rho_{-2,22}(90^\circ) &= \frac{3}{8} n_{20}.
\end{align*}
\]

(8)

The independent equation systems for Zeeman and optical coherences read

\[
\begin{align*}
\frac{d\rho_{2-2,2}}{d\tau} &= -\frac{i}{2} u - b_{22,22} \rho_{2-2,2}, \\
\frac{d\rho_{2-2,2}}{d\tau} &= -\frac{i}{2} u - b_{22,22} \rho_{2-2,2}, \\
d\frac{d\rho_{2-2,2}}{d\tau} &= -i\Omega_{22}^2 (\rho_{2-2,2} + \rho_{2-2,2}) - i\Delta_u - \frac{1}{2} u, \\
d\frac{dv}{d\tau} &= -i\Delta_v, \\
u(\tau) &= \Omega_{22}[\exp(-i\Delta_v \tau) \rho_{2-2,2} + \exp(i\Delta_v \tau) \rho_{2-2,2}], \\
v(\tau) &= \Omega_{22}[\exp(-i\Delta_v \tau) \rho_{2-2,2} - \exp(i\Delta_v \tau) \rho_{2-2,2}],
\end{align*}
\]

(9)

and
where \( \tau = t/\tau_e \) is the time scaled in the natural lifetime \( \tau_e = 26.2 \text{ ns} \) of the excited level \( F_e = 2 \) indicated by a prime, \( \Delta_2 = \tau_e(\omega_{p2} - \omega) \) and \( \Omega_2 \) are the detuning of the laser frequency \( \omega \) from the transition frequency \( \omega_{p2} \) and the Rabi frequency between two Zeeman sublevels \( F_f = 2M = 2 \) and \( F_e = 2M_e = 2 \) in units of the natural width \( \Gamma_e^{-1} \), correspondingly. Asymptotic solutions of (9) and (10) give zero values of the coherences that can be proved numerically. Thus the initial values for a subsequent application of the bichromatic pumping (3) follow from (6) upon reverse rotation of the coordinate system:

\[
    n_{FM}^0 = \sum_M \left[ d_{FM}^M (-\beta) \right]^2 n_{FM}(-\beta).
\]

The three stage pumping of \(^{87}\text{Rb} \) with sequential \( \pi^- , \sigma^- \) and \( \pi^- \)-polarization at the \( F_f = 2 \leftrightarrow F_e = 2 \) transition with account of (3), (6), (8)-(11) gives

\[
    n_{10} = n_{10}^0 + \frac{9657}{11200} \approx n_{10}^0 + 0.86n_{20}^0
\]

\[
    n_{20} = \frac{1543}{11200} \approx 0.14n_{20}^0
\]

where the initial population densities are defined in (4), which finally gives

\[
    n_{10} \approx 0.964, \quad n_{20} \approx 0.036.
\]

The pumping efficiency of the \( F_e = 1M = 0 \) sublevel is about the factor 7.47 respect to its initial equilibrium value 1/8. To avoid an additional error in the detecting signal a small amount of atoms in the \( F_f = 2M = 0 \) state can be removed from the beam applying a cyclic resonant transition from this state in a laser travelling wave. As a result, the remaining atoms of the beam will be in a pure state \( F_g M = 0 \).

A similar pumping scheme in a cesium atom gives the following values of the asymptotic populations of the clock sublevels \( F_g = 3 M = 0 \) and \( F_f = 4 M = 0 \) after the third stage

\[
    n_{30} \approx n_{30}^0 + 0.785n_{10}^0, \quad n_{40} \approx 0.215n_{40}^0,
\]

where the initial values are defined in (5). Finally, the resulting populations are

\[
    n_{30} \approx 0.934, \quad n_{40} \approx 0.066,
\]

with the pumping efficiency of the \( F_g = 3 M = 0 \) sublevel about the factor 14.9 at initial equilibrium populations 1/16.

4. **Numerical results**

Analytical results presenting above are independent of laser parameters, which however define time duration of the pumping process or a minimal size of a laser beam crossed by an atomic beam. Figures 3 and 4 illustrate temporal behavior of the Zeeman sublevel populations and coherences.
Figure 3. Evolution of the populations of Zeeman hyperfine sublevels of the ground state in $^{87}\text{Rb}$ (a), its excited state sublevel populations (b), and optical coherences (c) during the bichromatic $\pi$-polarized laser pumping on $F_g = 1 \leftrightarrow F_e = 1$ and $F_f = 2 \leftrightarrow F_e = 2$ transitions. The laser intensities are 5 mW/cm$^2$ and the frequency detuning $\Delta_1 = \Delta_2 = 0$. Thick full line presents $F_g = 1 M = 0$, dashed line is $F_g = 1 M = \pm 1$, thin full line is $F_f = 2 M = 0$, dotted line is $F_f = 2 M = \pm 1$ and chain line is $F_f = 2 M = \pm 2$.

In figure 4 the results of monochromatic pumping on $F_f = 2 \leftrightarrow F_e = 2$ transition under a $\sigma$-polarized laser field following the bichromatic pumping (figure 3) are shown. The initial values of sublevel populations and Zeeman coherences in rotated reference frame are from (8). The curves for the $F_g = 1 M$ sublevels present additional contribution to corresponding populations. In figure 4d the chain curve that started from zero value corresponds to the excited state Zeeman coherence.

Figure 4. The same as in figure 3 (a)-(c) and the Zeeman coherences (d) during the monochromatic pumping on the $F_f = 2 \leftrightarrow F_e = 2$ transition under a $\sigma$-polarized laser field (equations (9) and (10)). The curves are defined as in figure 3, but the quantization direction is along the polarization vector.

A similar two-laser scheme was used in experiments with a cesium fountain [22]. A strong polarization gradient at $F_f = 4 \leftrightarrow F_e = 4$ transition for a parabolic trajectory of atoms was created in the region of a two-dimensional optical lattice created by retroreflection of incoming laser polarization at 45° with respect to the vertical direction. Additional transverse cooling of atoms in a beam according
to the Sisyphus mechanism compensates their heating during Zeeman pumping by the \( F_g = 3 \leftrightarrow F_e = 3 \) transition. A significant enhancement of detecting signal was observed when using the pumping procedure.

5. Conclusions

We present a possibility of enhancing the optical laser pumping of the clock sublevels \( n^2S_{1/2} F,M = 0 \) in \(^{87}\text{Rb} \) \((n = 5)\) and \(^{133}\text{Cs} \) \((n = 6)\) atoms in a sequence of bichromatic and monochromatic transition schemes. In particular, the results of analytic studies of the multistage pumping with \( F_s \leftrightarrow F_e = F_g \) and \( F_s = F_e = F_g \) transitions in \( n^2S_{1/2} \leftrightarrow n^2P_{3/2} \) under the linear polarized laser fields show practically total population transfer from initially populated states to a single \( F_s M = 0 \) final state by varying the polarization direction on \( F_s \leftrightarrow F_e = F_g \) transition. The statistically equilibrium population value is increased during three-stage pumping approximately 7.5 times for rubidium and 15 times for cesium. Numerical studies give estimation of necessary duration of the resonant pumping process about 160 lifetime periods \( \tau \) in Rb at 5 mW/cm\(^2\) laser intensities with its extension to \( 10^3 \tau \) at 0.5 mW/cm\(^2\). These values fit conditions to realize the laser pumping system.

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