Optical–microwave pumping of alkali atoms and population capture

E. B. Aleksandrov and A. K. Vershovskii

(Submitted 1 July 1985)
Opt. Spektrosk. **59**, 1210–1216 (December 1985)

The steady-state distribution of the populations of the hyperfine sublevels of the ground state of alkali atoms is calculated for the case in which the atoms are subjected to a spectrally selective optical pumping on $^2S_{1/2}$–$^2P_{1/2,3/2}$ transitions and a simultaneous pumping by microwave fields which are at resonance with transitions in the hyperfine structure of the ground state, $F = 2$, $M_F = \pm 2, \pm 1 \leftrightarrow F = 1, M_F = \pm 1$. The addition of the microwave pumping is shown to substantially increase the population difference for the O–O transition in the hyperfine structure. During selective optical pumping of the $F = 1$ level, the population inversion which can be achieved for the O–O transition is limited by the effect of population capture. This capture can be eliminated by using incoherent microwave fields. The quality factor of the O–O resonance is calculated as a function of the parameters of the pump. The outlook for the use of composite pumping in frequency-stabilization systems is discussed.

1. INTRODUCTION

The optical pumping and microwave resonance in the system of hyperfine sublevels of the ground state of alkali atoms have retained the interest of researchers and engineers for many years. The reasons are that this system is attractive for model studies of the interaction of a quantum-mechanical system with electromagnetic fields and the applied interest in connection with frequency standardization. Suggestions have recently been made for using two-frequency laser light for optical pumping of the hyperfine structure of alkali atoms. In contrast with ordinary pumping, by the light from a resonant lamp with isotopic filtering, which allows, in the limiting case, an emptying of one of the two hyperfine levels during a uniform filling of the magnetic sublevels of the other, two-frequency laser pumping makes it possible in principle to put all the atoms in a common magnetic sublevel $m_F = 0$.[1] The result is a substantial improvement in the power of the signal of the microwave resonance at the reference transition, F_1, $m_{F_1} = 0$ with F_2, $m_{F_2} = 0$. In pursuing the suggestions, we decided to analyze a version of composite optical–microwave pumping, also used for a preferential filling of one sublevel $m_F = 0$. Figure 1 shows the pumping scheme. The $F_1 = I - 1/2$ and $F_2 = I + 1/2$ hyperfine levels of the $^2S_{1/2}$ ground state are pumped by incoherent pump light at rates W_1 and W_2. The $^2P_{1/2}$ and $^2P_{3/2}$ excited states are completely mixed and depolarized by collisions with atoms of the buffer gas. In addition, microwave transitions are pumped in the hyperfine system; these transitions are separated along the frequency scale by a weak auxiliary magnetic field (as shown by the arrows in Fig. 1).

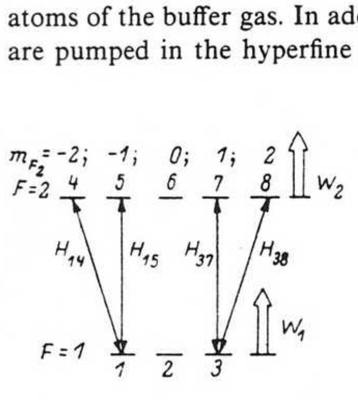

FIG. 1. Scheme for multifrequency pumping of the hyperfine sublevels of the ground state of alkali atoms.

Only in the limiting case of optical pumping of a single level, F_2 (i.e., with $W_1 = 0$), is it possible to predict, without going through calculations, the asymptotic behavior of the process at high pumping rates: A single sublevel, F_1, $m_{F_1} = 0$, will be populated. In other cases, the situation is complicated by the coherence imparted to the states by the microwave fields, with the result that predictions based on the population balance are wrong. An important role (and a negative role, for pumping purposes) is played by the coherent effect of population capture, which has been studied previously for the case of two-frequency pumping of a three-level system.[2–4] In the present paper we report calculations of the distribution of the populations of the hyperfine sublevels and of the optical signal of a reference microwave O–O resonance as a function of the pump parameters. We also discuss the outlook for applications of composite optical pumping.

2. POPULATIONS OF THE HYPERFINE SUBLEVELS DURING COMPOSITE PUMPING

To calculate the steady-state populations of the hyperfine sublevels during optical–microwave pumping, we take the approach of Ref. 5, where the problem of the O–O resonance in rubidium vapor was solved. In our case, instead of a single monochromatic field, applied to the 2–6 transition (Fig. 1), there are four fields, so that the problem is much more complicated. On the other hand, the problem simplifies substantially in the particular case in which the levels pertinent to the microwave pumping of the 1–4 and 3–8 transitions and also the 1–5 and 3–7 transitions are symmetric in pairs, i.e., when the moduli of the interaction matrix elements are equal ($|H_{14}| = |H_{38}|$ and $|H_{15}| = |H_{37}|$) and the frequency separations of the corresponding pairs of resonances are identical. In this situation, by virtue of the broadband nature of the optical pumping, it can be asserted that the populations of levels 1 and 3, 4 and 8, and 5 and 7 are equal in pairs. The microwave fields give rise to coherences in the system; by virtue of the symmetry of the problem, we need consider only ρ_{14}, ρ_{15}, ρ_{45}, and their conjugates.

We describe the incoherent optical pumping as a process of the relaxation of the populations of levels i at a rate W_1 for $i = 1$–3 or W_2 for $i = 4$–8, added to a thermal relaxation, whose rate q we assume to be identical for all of the sublevels. In this case, the coherences ρ_{14} and ρ_{15} relax at a rate $\gamma = (W_1 + W_2)/2 + q$ while ρ_{45} relaxes at a rate $\gamma_2 = W_2 + Q$.

Under our assumptions the system of equations for the density matrix elements of the ground state is

$$i\hbar\dot{\rho}_{11} = \rho_{14}H_{41} - \rho_{41}H_{14} + \rho_{15}H_{51} - \rho_{51}H_{15} - i\hbar\gamma_1\rho_{11} + i\hbar Q/8, \quad (1)$$

$$i\hbar\dot{\rho}_{14} = \rho_{41}H_{14} - \rho_{14}H_{41} - i\hbar\gamma_2\rho_{44} + i\hbar Q/8, \quad (2)$$

$$i\hbar\dot{\rho}_{55} = \rho_{51}H_{15} - \rho_{15}H_{51} - i\hbar\gamma_2\rho_{55} + i\hbar Q/8, \quad (3)$$

$$i\hbar\dot{\rho}_{14} = (\rho_{11} - \rho_{44})H_{14} - \hbar\omega_{14}\rho_{14} - i\hbar\gamma\rho_{14} - \rho_{54}H_{15}, \quad (4)$$

$$i\hbar\dot{\rho}_{15} = (\rho_{11} - \rho_{55})H_{15} - \hbar\omega_{15}\rho_{15} - i\hbar\gamma\rho_{15} - \rho_{45}H_{14}, \quad (5)$$

$$i\hbar\dot{\rho}_{45} = \rho_{41}H_{15} - \rho_{15}H_{41} - \hbar\omega_{45} - i\hbar\gamma_2\rho_{45}, \quad (6)$$

$$i\hbar\dot{\rho}_{22} = -i\hbar\gamma_1\rho_{22} + i\hbar Q/8, \quad (7)$$

$$i\hbar\dot{\rho}_{66} = -i\hbar\gamma_2\rho_{66} + i\hbar Q/8, \quad (8)$$

$$i\hbar\dot{\rho}_{26} = -i\hbar\gamma\rho_{26}, \quad (9)$$

where $\gamma_1 = W_1 + q$, $Q = \gamma_1(2\rho_{11} + \rho_{22}) + \gamma_2(2\rho_{44} + 2\rho_{55} + \rho_{66})$, and Ω_{ik} is the frequency of the transition between sublevels i and k.

System (1)–(9) is supplemented with a normalization condition corresponding to the case in which we ignore the populations of the excited states:

$$\sum_k \rho_{kk} = 1. \quad (10)$$

As in Ref. 5, we use a quasiresonant approximation, taking into account the effect of each microwave field on only its own transition. This approach justifies the approximation of a rotating wave, according to which the interaction with the microwave fields is described by

$$H_{ik} = \hbar V_{ik} e^{+i\Omega_{ik}t}. \quad (11)$$

The coherences ρ_{14} and ρ_{15} are assumed to be sinusoidal functions of the time with the frequencies of the pump fields at the corresponding transitions, while ρ_{45} obviously oscillates at the frequency $\Omega_{45} = \Omega_{41} - \Omega_{15}$.

We can now write expressions for the steady-state populations of the hyperfine sublevels in the simplest case, and that of the most practical interest, $|V_{ik}| = V$ for resonant fields ($\Omega_{ik} = \omega_{ik}$):

$$\rho_{11} = \frac{Q_0}{8}\,\frac{8V^2 + \gamma\gamma_2}{\gamma(8V^2 + \gamma_1\gamma_2)}, \quad (12)$$

$$\rho_{44} = \frac{1}{2}\left(\frac{Q_0}{8}R - \rho_{11}\right), \quad (13)$$

$$\rho_{55} = \rho_{44}, \quad (14)$$

$$\rho_{22} = \frac{Q_0}{8}\,\frac{1}{\gamma_1}, \quad (15)$$

$$\rho_{66} = \frac{Q_0}{8}\,\frac{1}{\gamma_2}, \quad (16)$$

where

$$Q_0 = \frac{4\gamma_1\gamma_2}{R\gamma_1\gamma_2 + \gamma}, \quad (17)$$

$$R = \frac{(8V^2 + \gamma\gamma_2)^2}{\gamma(8V^2 + \gamma_1\gamma_2)(4V^2 + \gamma\gamma_2)} + \frac{4V^2 + 2\gamma\gamma_2}{\gamma_2(4V^2 + \gamma\gamma_2)}. \quad (18)$$

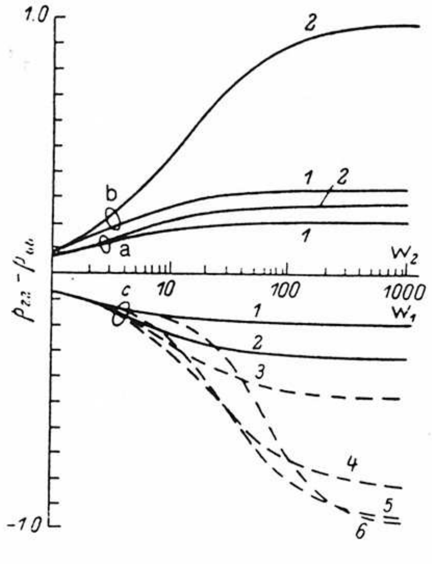

FIG. 2. Population difference for the O–O transition. a—$W_1/W_2 = 0.2$: b—$W_1/W_2 = 0$; c—$W_1/W_2 = 0$. 1—$V = 0$; 2,a,b—$V = W_2$, $\delta = 0$; 2,c—$V = W_1$, $\delta = 0$; 3—$V = W_1$, $\delta = 1$; 4—$V = W_1$, $\delta = 10$; 5—$V = W_1$, $\delta = 100$; 6—$V = W_1$, $\delta = 1000$.

The solid lines in Fig. 2 show the population difference $\rho_{22} - \rho_{66}$ vs the intensity W_1 of the optical pump (under the condition $W_1 > W_2$) or W_2 ($W_2 > W_1$), for various amplitudes of the monochromatic fields V. All quantities with the dimensionality of a linewidth (W_1, W_2, and V) are expressed in units of q.

The case of pumping to the lower level ($W_2 > W_1$) is illustrated in two situations: $W_1/W_2 = 0.2$ [Fig. 2(a)] and $W_1/W_2 = 0$ [Fig. 2(b)]. As can be seen from Fig. 2, the addition of the microwave pump with $W_1 = 0$ leads to a significant increase in the population difference: As expected, in the case of intense composite pumping nearly all the atoms go into a common level, $M_{F_1} = 0$. Figure 3 makes it possible to see in more detail the role of the ratio of intensities of the optical and microwave pumps in this case. We see that at a given intensity W_2 of the optical pump the increment in the population difference due to the microwave pump reaches saturation when the microwave broadening of magnetic-field-dependent transitions becomes comparable to the optical broadening.

In the case of pumping to the upper level [$W_1 > W_2$, Fig. 2(c)] the situation is more complicated. To keep the figures clear, we show the calculated results only for $W_2 = 0$. In this

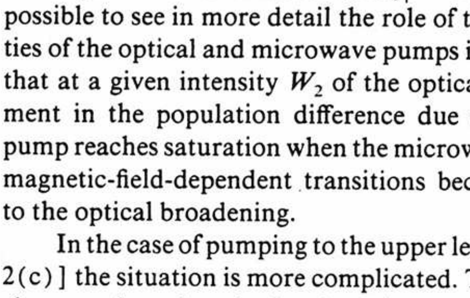

FIG. 3. Population difference for the O–O transition during pumping with monochromatic microwave fields ($\delta = 0$, $W_1/W_2 = 0$). 1—$V = 0$; 2—$V = 0.01\,W_2$; 3—$V = 0.1\,W_2$; 4—$V = 0.3\,W_2$; 5—$V = W_2$.

case, the addition of the microwave pump is far less effective: The difference $\rho_{22} - \rho_{66}$ has a limit of $-1/3$, not the -1 which we would expect on the basis of balance considerations. A complete calculation of the populations shows that in this case the lower state F_1 is completely emptied by the pump light, but two-thirds of all the atoms are stuck in the sublevels $i = 4, 5, 7, 8$, despite the microwave fields, which are applied in an effort to pull the atoms into the light, into the state F_1.

We are dealing here with a phenomenon known in optics as population capture.[2–4] This effect has previously been studied in an inverted-V arrangement of three levels, coupled by two coherent fields. When the population of the common level decays (because of spontaneous emission or ionization) the sum of the populations of the two other levels tends toward a constant value on an unbounded increase in the strength of the alternating fields, while the population of the common level approaches zero. In our case, the same situation arises in a (noninverted) V arrangement of microwave transitions: The common lower level is emptied by the optical pumping.

The physical meaning of population capture is that the coherent fields interacting with a triad of levels (e.g., 1, 4, and 5) create a rotating magnetic dipole moment, which is described by the coherence ρ_{45}. A rotation phase is established in which the fields V_{14} and V_{15} essentially disengage from the magnetic top. As a result, the flux of atoms to level 1 caused by the microwave fields is reduced to a minimum determined by the relaxation of the phase of the top and independent of the microwave power.

In order to eliminate population capture, it is necessary to disrupt the coherences ρ_{45} and ρ_{78} rapidly, e.g., by using a noisy modulation of the phase of the microwave fields, so that the width δ of their spectrum exceeds the width of the transitions 4–5 and 7–8.[6] The result of the application of the noisy fields to the system can be found most simply from Eqs. (1)–(10) for monochromatic fields by introducing an additional effective relaxation of coherences at a rate equal to the width δ of the spectrum of the fields. The relaxation constants γ and γ_2 in Eqs. (4)–(6) are replaced by $\Gamma = \gamma + \delta$ and $\Gamma_2 = \gamma_2 + 2\delta$, respectively.

The solutions of this system for ρ_{22} and ρ_{66} are again given by Eqs. (15)–(17), where now we have

$$R = \frac{[2V^2(3a+1)+\Gamma\Gamma_2]^2}{[2V^2(\gamma_1(a+1)+2\Gamma_2)+\gamma_1\Gamma\Gamma_2][2V^2(a+1)+\Gamma\Gamma_2]} + \frac{2[2V^2+\Gamma\Gamma_2]}{\gamma_2[2V^2(a+1)+\Gamma\Gamma_2]}, \tag{18a}$$

$$a = \Gamma_2/\gamma_2. \tag{19}$$

The calculated results are shown by the dashed lines in Fig. 2(c) for several values of the width δ of the microwave field spectrum. We see that the population inversion now approaches a complete inversion with increasing pump intensity.

3. QUALITY FACTOR OF THE O–O RESONANCE DURING COMPOSITE PUMPING

What is the outlook for the use of composite pumping in a rubidium frequency standard? To answer this question, we consider a fifth coherent field, V_{26}, whose frequency we associate with the frequency of the 2–6 transition. The field V_{26} changes the population difference $\rho_{22} - \rho_{66}$ and thereby changes the optical absorption of the system, which plays the role of a resonance signal. In addition to performing its function of forming an error signal, however, the field V_{26} participates in a pumping process, reducing not only the population difference $\rho_{22} - \rho_{66}$ but also the sum of these populations. Let us assume, for example, that we are dealing with a purely optical pumping to the F_2 level, which ultimately [Fig. 2(c)] allows us to put 0.2 of all the atoms in sublevel 6. When the field V_{26} is turned on, the atoms from this sublevel are subjected to optical excitation through sublevel 2, and from the excited state they become uniformly distributed among the sublevels of the ground state. As a result, the population of sublevel 6 falls off, while those of the side sublevels 4, 5, 7, and 8 increase. In this situation, the use of auxiliary microwave fields to empty the side sublevels has an additional advantage.

The problem of the O–O resonance during composite pumping can be solved by introducing an interaction with a field H_{26} in Eqs. (7)–(9):

$$\hbar\dot{\rho}_{22} = \rho_{26}H_{62} - \rho_{62}H_{26} - i\hbar\gamma_1\rho_{22} + i\hbar Q/8,$$
$$i\hbar\dot{\rho}_{66} + \rho_{62}H_{26} - \rho_{26}H_{62} - i\hbar\gamma_2\rho_{66} + i\hbar Q/8,$$
$$i\hbar\dot{\rho}_{26} = (\rho_{22} - \rho_{66})H_{26} - \hbar\omega_{26}\rho_{26} - i\hbar\gamma\rho_{26}.$$

In the approximation of a thin layer, the number of pump photons which are absorbed by the atoms per unit time is proportional to the quantity

$$S = Q_0 - q + \frac{A}{\Delta^2 + \Gamma_{\text{eff}}^2}\cdot \tag{20}$$

where $\Delta = \omega_{26} - \Omega_{26}$ is the frequency deviation from the O–O resonance,

$$A = 4|V_{26}|^2 \frac{\gamma(\gamma_1 - \gamma_2)^2}{(R\gamma_1\gamma_2 + \gamma)^2} \tag{21}$$

$$\Gamma_{\text{eff}}^2 = \gamma^2 + 4|V_{26}|^2 \frac{\gamma(R\gamma + 1)}{R\gamma_1\gamma_2 + \gamma}\cdot \tag{22}$$

and Q_0 and R are given by Eqs. (17)–(18a). The resonance signal proper is the increment in the number of absorbed photons which is related to the application of the field V_{26}:

$$S'(\Delta) = \frac{A}{\Delta^2 + \Gamma_{\text{eff}}^2}\cdot \tag{23}$$

The resonance signal is Lorentzian with a half-width Γ_{eff}. The quality factor of the resonance—a quantity proportional to the accuracy with which the deviation of the frequency Ω_{26} is measured—is determined by the expression $\Phi = S'(0)/N\Gamma_{\text{eff}}$, where N is the mean square amplitude of the noise against whose background the resonance signal is observed. If the measurement system is designed properly, the noise is determined entirely by the shot noise of the photocurrent of the detector; i.e., it is proportional to $\sqrt{W_1 + W_2}$. The quality factor becomes

$$\Phi = \frac{A}{\sqrt{\beta(W_1 + W_2)}\,\Gamma_{\text{eff}}^3}, \tag{24}$$

where β is a constant. For given pump rates W_1 and W_2, the quality factor reaches a maximum value of

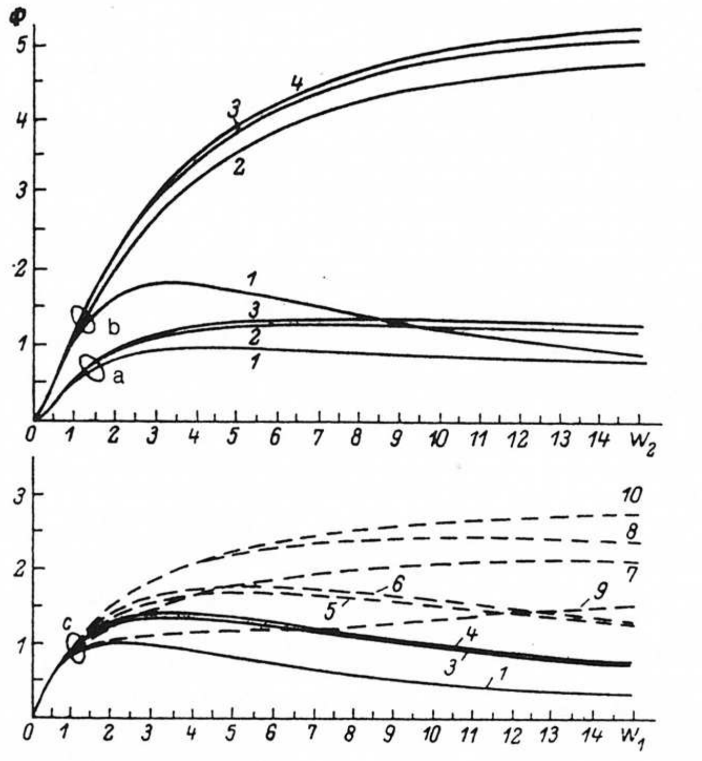

FIG. 4. Quality factor of the O–O resonance versus the pump intensity. a—$W_1/W_2=0.2$; b—$W_1/W_2=0$; c—$W_2/W_1=0$. 1—$V=0$; 2—$V=0.5W_2$, $\delta=0$; 3,a,b—$V=W_2$, $\delta=0$; 3,c—$V=W_1$, $\delta=0$; 4,a,b—$V=10W_2$, $\delta=0$; 4c—$V=10W_1$, $\delta=0$; 5—$V=W_1$, $\delta=1$; 6—$V=10W_1$, $\delta=1$; 7—$V=W_1$, $\delta=10$; 8—$V=10W_1$, $\delta=10$; 9—$V=W_1$, $\delta=100$; 10—$V=10W_1$, $\delta=100$.

$$\Phi_{opt}=\frac{2}{3\sqrt{3\beta}}\frac{(W_1-W_2)^2}{\gamma(R\gamma+1)(R\gamma_1\gamma_2+\gamma)\sqrt{W_1+W_2}}, \tag{25}$$

at

$$\Gamma_{eff}=\sqrt{3}\gamma,\quad |V_{26}|^2=\frac{(R\gamma_1\gamma_2+\gamma)\gamma}{2(R\gamma+1)}. \tag{26}$$

Figure 4 shows curves of Φ_{opt} versus the pump intensity for several values of V and δ. As before, the solid lines correspond to monochromatic microwave fields (or their absence), while the dashed lines correspond to noisy fields.

As can be seen from Fig. 4, the microwave pump can substantially raise the quality factor of the O–O resonance, by a factor as high as three, during pumping to the F_1 level. There are two important circumstances to be noted, however. First, a significant improvement is achieved only in the case of a high spectral selectivity of the optical pumping; i.e., the condition $W_1 \gg W_2$ or $W_2 \gg W_1$ must be satisfied. Second, an increase in the quality factor requires a more intense optical pumping. For example, the attainment of the maximum quality factor during composite pumping to the level F_1 corresponds to a fivefold increase in the light intensity. At the same time, there will be a lowering of the long-term frequency stability of the reference transition through a mechanism of optical shifts of the resonance. Furthermore, the introduction of additional microwave fields is potentially a source of field-induced shifts of the resonant frequency. Because of these circumstances, caution must be exercised in recommending the use of composite pumping for a rubidium frequency standard with a gas-filled cell.

However, with regard to beam systems which have been suggested for frequency stabilization with optical pumping, the optical–microwave conversion is clearly promising. It is to this case which the results in Section 2 refer. In beam systems, the pumping region is spatially separated from the measurement region, so that we do not need to deal with the problem of optical and microwave shifts of the reference resonance. The optical–microwave modification of schemes for laser pumping of atomic beams is particularly pertinent. In this case it is a simple matter to achieve an ideal spectral selectivity of the excitation, and the required pump power can be realized (as can be seen from Figs. 1 and 2, in order to reach the $\rho_{22}-\rho_{66}$ saturation region it is necessary to achieve a pump rate on the order of 100 times the relaxation rate, which is determined in this case by the transit time of the atoms through the pumping region). In terms of the ultimate result, composite laser-microwave pumping is equivalent to two-frequency laser pumping, but the composite pumping is preferable: The requirements on the frequency spread and frequency stability of the laser light are greatly relaxed since two-frequency laser pumping presupposes a selectivity of the excitation of not only the hyperfine sublevels of the ground state but also of the excited state.

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