Inhibition of the Larmor precession during optical pumping in a weak magnetic field

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We have experimentally and theoretically investigated the behavior of Na atoms under the combined influence of resonant laser light and a magnetic field. Linearly polarized light was tuned to the $(3s)^2S_{1/2}$ $(F=2)\rightarrow(3p)^2P_{1/2}$ $(F'=1)$ component of the D_1 line. The intensity of fluorescent light was observed as a function of the laser intensity for various strengths of the magnetic field B. Trapping in the $M_F=\pm2$ levels of the ground state could be observed to be of decreasing importance for an increasing component of **B** perpendicular to the laser polarization $\hat{\epsilon}$. For laser intensities increasing beyond certain critical values, however, the fluorescence decreased and trapping seemed to become again important. For a quantitative explanation of the observed behavior we describe the system by generalized rate equations for the density matrix. We find that stimulated absorption and emission processes effectively inhibit the Larmor precession, thereby enforcing the trapping effect. Precession can only take place at sufficiently low light intensities.

I. INTRODUCTION

In recent years numerous collision experiments have been performed with laser-excited atoms. For a proper interpretation of the experimental data it is desirable to know and to control the distribution of the excited-state population over the various fine and hyperfine levels and their magnetic sublevels. For an increase of the measured signals it is in addition desirable to obtain a high density of excited atoms. In many cases the lifetime of the excited state is much shorter than the passage time of the atom through the interaction region, so that an atom can in principle be excited several times. Then it is important to ensure that the atoms can again be excited from all sublevels which are populated by spontaneous decay. To this end the application of a magnetic field can be helpful, in particular in those cases where the ground state involved in the laser transitions has more magnetic sublevels than the excited state. Without a magnetic field B, atoms will be trapped in a magnetic sublevel of the ground state, from which they cannot be excited again by the polarized laser light. Application of a magnetic field induces a Larmor precession of the atom about the magnetic field direction which corresponds to a periodic exchange of populations for different magnetic sublevels and which removes the "trap." However, this precession and the resulting removal of the trap can severely be disturbed by the absorption and emission processes induced by the laser light. It is the purpose of this paper to present an appropriate theoretical treatment and to discuss the optimal conditions for obtaining a high density of excited atoms with well-known distributions of magnetic sublevels. We apply this theoretical treatment to explain experimental observations of atomic fluorescence intensities made in our laboratory.

As a specific example we investigate the excitation of Na atoms. Figure 1 shows a diagram of relevant Na energy levels. Laser excitation of the Na $^2P_{3/2}$ state has been studied in detail by Fischer and Hertel.[1] In that case trapping can be avoided by tuning the laser to the $F=2\rightarrow F'=3$ transition. Here we want to discuss excitation of the Na $^2P_{1/2}$ state, especially the $F=2\rightarrow F'=1$ transition. Linearly polarized light can only induce $\Delta M=0$ transitions (in a coordinate frame with the z axis chosen parallel to the light polarization vector $\hat{\epsilon}$). Atoms in the $M=2$ sublevel are therefore trapped. However, this trap can be removed by a magnetic field with a component of **B** perpendicular to the $\hat{\epsilon}$ direction. In our case, excitation takes place in magnetic fields up to $B=2$ G. These fields are weak in the sense that the corresponding Zeeman splitting is small compared to the natural width of the excited Na state, but strong enough to induce appreciable precession during the passage time through the laser beam. Therefore this system is well suited to study trapping and its removal during laser excitation in a magnetic field.

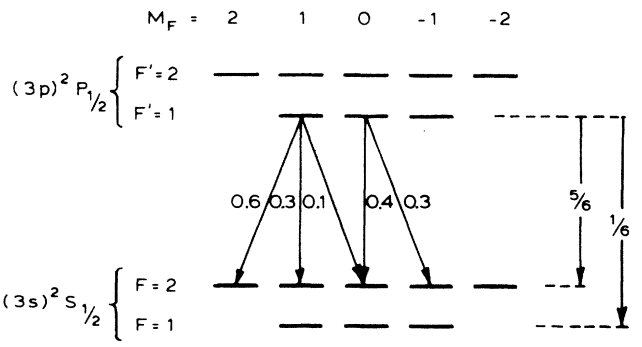

FIG. 1. Some energy levels of Na $(3s)$ and $(3p)$. Squares of Clebsch-Gordan coefficients are inserted for the dipole transitions discussed in this paper.

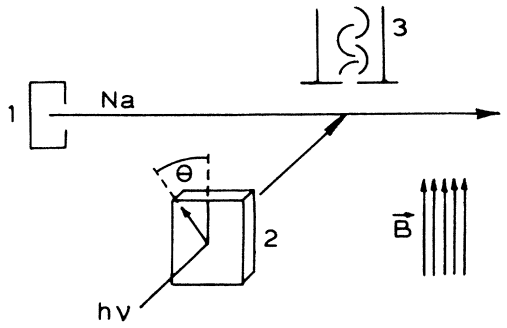

FIG. 2. Schematic view of the apparatus. (1) Na oven. (2) Polarization rotator for the laser light. (3) Photomultiplier for detection of fluorescent light.

II. EXPERIMENT

We have excited Na atoms in a thermal atomic beam (divergence 0.05°) from an effusive oven by means of a cw dye-laser beam, directed perpendicularly to the atomic beam. Figure 2 shows a schematic diagram of the apparatus. The laser frequency was tuned to the $^2S_{1/2}$ $(F=2)\rightarrow{}^2P_{1/2}$ $(F'=1)$ transition by maximizing the atomic fluorescence as a function of laser frequency in the appropriate range. The stability of the laser frequency was better than 1 MHz. The interaction region between laser and atomic beam had a length of $l=5$ mm. At an oven temperature of $T=634$ K this corresponds to an average passage time of $t=l\langle v^{-1}\rangle=l(2m/\pi kT)^{1/2}=8.4$ μs. This interaction time is much larger than the mean lifetime of the excited atoms (16 ns) and therefore the atoms could in principle be excited many times as they travel through the laser spot.

We have excited the atoms by linearly polarized light. As an indication for the number of excited atoms we have measured the intensity of the resonance fluorescent light F by means of a photomultiplier. In one set of measurements we have determined F as a function of the angle θ between the linear polarization vector $\hat{\epsilon}$ and the magnetic field **B** in the interaction region. **B** had a strength of $B=0.5$ G and was mainly the earth's magnetic field, with the component perpendicular to the rotation plane of $\hat{\epsilon}$ being compensated by a Helmholtz coil. A resulting curve is shown in Fig. 3. It demonstrates that the magnetic field perpendicular to $\hat{\epsilon}$ has an important influence and helps to obtain a large number of excited atoms. In a second set of measurements we have determined the fluorescence F as a function of the laser intensity I. In this case $\hat{\epsilon}$ was chosen perpendicular to **B** and the strength of **B** was taken as parameter, thus obtaining different curves $F_B(I)$. The result is shown in Fig. 4. The most remarkable feature is the fact that in some cases the fluorescence intensity decreases with increasing laser intensity.

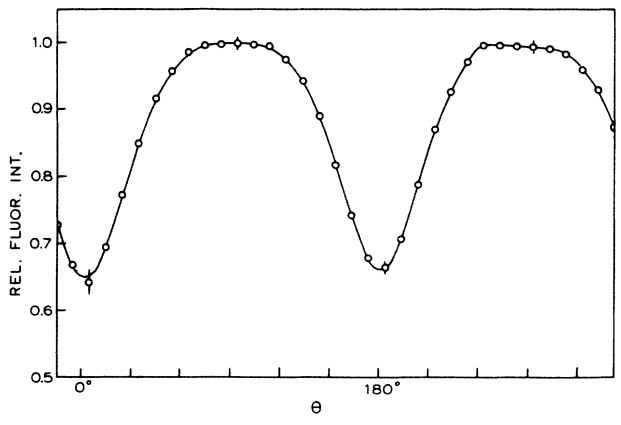

FIG. 3. Relative fluorescence intensities at varying orientation of the linear polarization vector $\hat{\epsilon}$ of the laser light. At $\theta=0$, $\hat{\epsilon}$ is parallel to the magnetic field **B**.

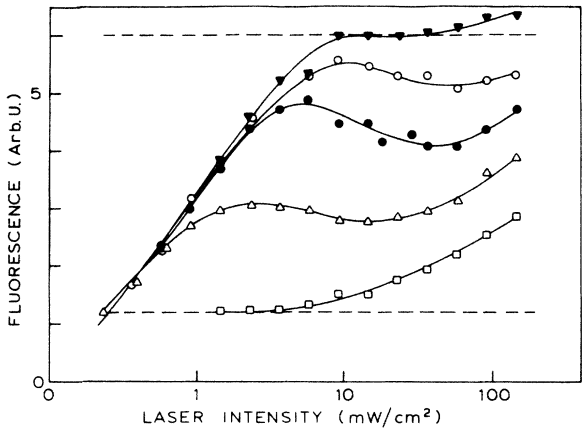

FIG. 4. Measured fluorescence intensities as a function of laser intensity for different fields **B** perpendicular to the linear polarization vector $\hat{\epsilon}$. □: $B=0$; △: $B=0.27$ G; ●: $B=0.54$ G; ○: $B=1.08$ G; ▼: $B=2.16$ G.

III. QUALITATIVE CONSIDERATIONS

For an understanding of the data shown in Figs. 3 and 4 one has to realize that fluorescence photons emitted by Na $^2P_{1/2}$ have an isotropic angular distribution. This has been verified experimentally by measuring the fluorescence intensity in two different directions simultaneously. Therefore the measured fluorescence intensity and its variation is a direct measure for the average number of excited atoms.

At first glance a straightforward explanation for the θ dependence seems to be at hand: at $\theta=0°$, where $\hat{\epsilon}$ is parallel to **B**, there is no removal of the $M=\pm2$ traps. This yields a large number of trapped atoms and results in a low fluorescence intensity. At $\theta=90°$ and 270°, on the other hand, the component of **B** perpendicular to $\hat{\epsilon}$ has its maximum, the $M=\pm2$ traps are most efficiently removed, and this yields a maximum of excited atoms and fluorescence intensity.

The results, shown in Fig. 4, however, indicate that at high radiation densities other effects become important.

To obtain an appropriate description of these processes we proceed as follows.

(i) We discuss the limits of efficiency for a magnetic field in the removal of a trap.

(ii) We show that an independent treatment of Larmor precession and optical pumping is insufficient.

(iii) We give an appropriate quantum-mechanical description by means of generalized rate equations.

(iv) We compare this description with the experimental data.

A. Limits of efficiency for the magnetic field

We want to calculate the average number N of photons that are spontaneously emitted from a Na atom on its way through the excitation region, being initially in the $F=2$ hyperfine level of the $^2S_{1/2}$ ground state (see Fig. 1). Apart from trapping in the $M=2$ sublevels we have to take into account that decay from $F'=1$ can occur to both hyperfine levels $F=1,2$ of the ground state. From $F=1$ the atom cannot be excited again and is lost. The branching ratio for transitions $F'=1\rightarrow F=2$ and $F'=1\rightarrow F=1$ can be obtained from the dipole transition amplitude D given by[2]

$$D_{FF'}=\langle JIF||\mu||J'I'F'\rangle$$
$$=(-1)^{J+I+F+1}\langle J\,|\,\mu\,|\,J'\rangle$$
$$\times\sqrt{(2F+1)(2F'+1)}\begin{Bmatrix} J & F & I \\ F' & J' & 1 \end{Bmatrix}\delta_{II'}\,, \qquad (1)$$

with μ the dipole operator, and J, I, and F the electronic, nuclear, and total angular momentum, respectively. This yields a branching ratio of 5:1, which means that a fraction of $\frac{1}{6}$ of the atoms is lost in each excitation-decay cycle. Due to this the maximum number of photons for the case that the $M=\pm 2$ traps are completely removed is

$$\bar{N}_{\max}=\sum_{n=0}^{\infty}(\tfrac{5}{6})^n=6\,. \qquad (2)$$

If, on the other hand, the $M=\pm 2$ traps are not at all removed ($B=0$), one finds (for the case that stimulated absorption is much faster than spontaneous decay)

$$\bar{N}_{\min}=\tfrac{3}{5}\sum_{n=0}^{\infty}(\tfrac{5}{6}\times\tfrac{3}{5})^n=1.2\,. \qquad (3)$$

Here the branching ratios for decay into the various $F=2$ magnetic sublevels have been taken into account, which can be obtained from the Wigner-Eckart theorem. Under the influence of a magnetic field the observed fluorescence intensity can therefore at most vary by a factor of 5.

B. Why Larmor precession and optical pumping cannot be treated separately

In the first place one can try to take the influence of the magnetic field into account by treating the Larmor precession in the usual way independent of the radiative transitions. This was done, e.g., by Kroon *et al.*[3] All wave functions are expanded into eigenstates along the magnetic field direction. These eigenstates have time-dependent phase factors $\exp(im\mu_B gBt/\hbar)$ with μ_B the Bohr magneton, g the Landé factor and m the magnetic quantum number in this coordinate frame. In the coordinate frame with $\hat{z}$ parallel to $\hat{\epsilon}$—in which the selection rules for optical excitation are valid—this time dependence corresponds to a periodic interchange of populations for the various magnetic sublevels. In this picture one would expect the $M=\pm 2$ populations to be "rotated" to other sublevels within a quarter Larmor period (in our case with $B=0.5$ G, $\tau_e/4=2\,\mu$s).

However, this picture of Larmor precession is only valid if the relative phases of the population amplitudes involved are not disturbed during the precession. In case of strong laser excitation and decay such an unperturbed phase development cannot be assumed. For example, contributions to the $M=2$ population caused by a pumping process from $M=1$ to $M=2$ (via excitation to and decay from the $M'=1$ level) will disturb the coherences involving the $M=2$ state. An appropriate description cannot be given in terms of populations, and a density matrix treatment has to be used.

IV. THEORY

A. Evolution equations

We use generalized rate equations to describe the time dependence of the density matrices $\rho_g(t)$ and $\rho_e(t)$ for the ground state and the excited state of the atom respectively. In this treatment Rabi oscillations are omitted. This is justified for total interaction times between atoms and laser light that are much longer than the natural lifetime. The Zeeman level splitting is assumed to be small compared with the sum of the bandwidth and the natural width.

We start with the following equations[4] in which the first terms describe the free evolution of the atom in the magnetic field, the second ones loss and gain, respectively, due to spontaneous decay processes, and the third ones stimulated absorption and emission:

$$\frac{d}{dt}\rho_e(t)=-\frac{i}{\hbar}[H_e,\rho_e(t)]-A'\rho_e(t)+\frac{\pi}{\epsilon_0\hbar^2 c}I_{\text{eff}}[\hat{\epsilon}\cdot\mu_{eg}\rho_g(t)\mu_{ge}\cdot\hat{\epsilon}^*-\tfrac{1}{2}\hat{\epsilon}\cdot\mu_{eg}\mu_{ge}\cdot\hat{\epsilon}^*\rho_e(t)-\tfrac{1}{2}\rho_e(t)\hat{\epsilon}\cdot\mu_{eg}\mu_{ge}\cdot\hat{\epsilon}^*]\,, \qquad (4)$$

$$\frac{d}{dt}\rho_g(t)=-\frac{i}{\hbar}[H_g,\rho_g(t)]+\frac{\omega_0^3}{3\pi\epsilon_0\hbar c^3}\mu_{ge}\cdot\rho_e(t)\cdot\mu_{eg}$$
$$+\frac{\pi}{\epsilon_0\hbar^2 c}I_{\text{eff}}[\hat{\epsilon}^*\cdot\mu_{ge}\rho_e(t)\mu_{eg}\cdot\hat{\epsilon}-\tfrac{1}{2}\hat{\epsilon}^*\cdot\mu_{ge}\mu_{eg}\cdot\hat{\epsilon}\rho_g(t)-\tfrac{1}{2}\rho_g(t)\hat{\epsilon}^*\cdot\mu_{ge}\mu_{ge}\cdot\hat{\epsilon}]\,. \qquad (5)$$

Here $\rho_e(t)$ and $\rho_g(t)$ have dimensions of $(2F+1)\times(2F+1)$ and therefore represent 3×3 and 5×5 matrices, respectively. The effective light intensity I_{eff} is given by

$$I_{\text{eff}}=\int d\omega\, I(\omega)\frac{A/2\pi}{(\omega-\omega_0)^2+(A/2)^2}\ , \tag{6}$$

with $I(\omega)$ the frequency-dependent laser intensity. The loss term A' can be written as

$$A'\rho_e=A(1+s)\rho_e\ , \tag{7}$$

where A is the Einstein coefficient for spontaneous transition $e\rightarrow g$ given by

$$A=\frac{\omega_0^3}{3\pi\epsilon\hbar c^3}\,|\langle F'||\mu||F\rangle|^2(2F'+1)^{-1}\ , \tag{8}$$

with $\langle F'||\mu||F\rangle$ a reduced matrix element, and where the factor $1+s$ takes spontaneous transitions to other states into account (in the present case to the $F=1$ level of the Na ground state, which gives $s=\frac{1}{5}$).

The components of the dipole operators μ_{ge} and μ_{eg} can in our case be represented by 5×3 and 3×5 matrices, respectively, with matrix elements $\mu_{eg}=\langle F'M'|\mu|FM\rangle$. The operator $\mu_{ge}\rho_e(t)\mu_{eg}$ transfers a possible anisotropy from the excited state to the ground state by spontaneous decay processes. The stimulated contributions contain the intensity $I(\omega)$ and the polarization vector $\hat{\epsilon}$ of the laser light. $\hat{\epsilon}$ can conveniently be expressed in terms of spherical unit vectors $\hat{\mathbf{u}}_\sigma$ for circular ($\sigma=\pm1$) and linear ($\sigma=0$) polarization

$$\hat{\mathbf{u}}_{\pm1}=(\mp\hat{\mathbf{x}}-i\hat{\mathbf{y}})/2,\quad \hat{\mathbf{u}}_0=\hat{\mathbf{z}}\ . \tag{9}$$

We introduce the operators C_σ with dimensions $(2F'+1)\times(2F+1)$ defined via the Clebsch-Gordan coefficients for transitions between F and F',

$$\langle M'|C_\sigma|M\rangle\equiv\langle F'M'|FM;1\sigma\rangle\ . \tag{10}$$

For pure linearly or circularly polarized laser light we can then write

$$\hat{\mathbf{u}}_\sigma\cdot\mu_{eg}=C_\sigma\langle F'||\mu||F\rangle(2F'+1)^{-1/2} \tag{11}$$

according to the Wigner-Eckart theorem. Also we write the differential equations (4) and (5) in a dimensionless way by dividing by A and using the notation

$$r_e=\omega_e/A,\quad r_g=\omega_g/A,\quad \tau=At \tag{12}$$

for the reduced Larmor frequencies in the excited and ground state, respectively, and for the reduced time τ. Finally, we introduce the saturation parameter

$$b=\pi^2c^2I_{\text{eff}}/\hbar\omega^3=BI_{\text{eff}}/Ac\ , \tag{13}$$

with B, A the Einstein coefficients. Since we want to describe experiments in which pure polarized light is used we choose a coordinate frame with $\hat{\mathbf{z}}$ parallel to the polarization vector, such that $\hat{\epsilon}=\hat{\mathbf{u}}_0$ and we can use (11). Then the rate equations (4) and (5) become

$$\frac{d\rho_e}{d\tau}=-ir_e[F'_{||},\rho_e]-(1+s)\rho_e+3b(C_\sigma\rho_gC_\sigma^\dagger-\tfrac{1}{2}C_\sigma C_\sigma^\dagger\rho_e-\tfrac{1}{2}\rho_eC_\sigma C_\sigma^\dagger)\ , \tag{14}$$

$$\frac{d\rho_g}{d\tau}=-ir_g[F_{||},\rho_g]+\textstyle\sum_\sigma C_\sigma^\dagger\rho_eC_\sigma+3b(C_\sigma^\dagger\rho_eC_\sigma-\tfrac{1}{2}C_\sigma^\dagger C_\sigma\rho_g-\tfrac{1}{2}\rho_gC_\sigma^\dagger C_\sigma)\ . \tag{15}$$

Here $F'_{||}$ and $F_{||}$ are the components of $\mathbf{F}'$ and $\mathbf{F}$ in the direction of the magnetic field. In this reduced form the generalized rate equations can be used for numerical calculations in order to describe the time development of ρ_e and ρ_g.

B. Model calculations

Some insight regarding the competition between precession and optical pumping can be obtained by looking at a simple model case, for which analytical results are readily obtained. We consider the three-state system sketched in Fig. 5. The radiation field couples the states 1 and 2, and spontaneous emission occurs both to the state 0 and 1, in the ratio a_0 and a_1 (with $a_0+a_1=1$). Hence in the absence of Zeeman precession, state 0 acts as a trap. A magnetic field induces a precession frequency $\omega=rA$. This model describes the situation of a transition between two states with $F'=F=\frac{1}{2}$, pumped by circularly polarized light and with negligible Zeeman coupling in the excited state. The generalized rate equations (14) and (15) now take the form

$$\begin{aligned}
\frac{d}{d\tau}\rho_{00}&=a_0\rho_{22}-\tfrac{1}{2}ir(\rho_{10}-\rho_{01})\ ,\\
\frac{d}{d\tau}\rho_{11}&=a_0\rho_{22}+\tfrac{1}{2}ir(\rho_{10}-\rho_{01})-3a_1b(\rho_{11}-\rho_{22})\ ,\\
\frac{d}{d\tau}\rho_{22}&=-\rho_{22}+3a_1b(\rho_{11}-\rho_{22})\ ,\\
\frac{d}{d\tau}\rho_{10}&=-\tfrac{3}{2}a_1b\rho_{10}-\tfrac{1}{2}ir(\rho_{00}-\rho_{11})\ .
\end{aligned} \tag{16}$$

It is now an easy matter to calculate the stationary population of the excited state 2, by assuming the limit $d\rho/dt=0$. This stationary population is

$$\bar{\rho}_{22}=3r^2a_1b/[r^2(2+9a_1b)+9a_1^2a_0b^2]\ . \tag{17}$$

One notices that the excited-state population $\bar{\rho}_{22}$ is an in-

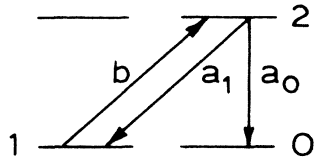

FIG. 5. Schematic level diagram for the system assumed in the model calculations of Sec. IV B.

creasing function of the reduced Zeeman precession frequency r. On the other hand, for a fixed value of r, this excited-state population increases as a function of the stimulated transition rate b for low intensities, but when b is larger than r, $\bar{\rho}_{22}$ becomes a decreasing function of b. In this region, the stimulated radiative transitions cause an effective inhibition of the Larmor precession, and prevent the atoms from escaping from the trapped state 0. This inhibition is due to the damping of the Zeeman coherence at the rate $3a_1b/2$. It is this damping of the coherence by stimulated radiative transitions which prevents an appreciable buildup of ρ_{10}, and thereby of a repopulation of the state 1 by Larmor precession. The basic mechanism in the model system is obviously equally present in the Na transitions under discussion.

V. COMPARISON BETWEEN CALCULATIONS AND OBSERVATION

Equations (14) and (15) allow us to trace the populations of the various magnetic sublevels as a function of time when they travel through the laser spot. Also we can numerically determine the average number of photons that are emitted in each time interval. As an example we have calculated the time-dependent sublevel populations of the $F=2$ ground state for the case of a magnetic field $B=0.65$ G and saturation parameters $3b=10$ and $3b=0.8$. An initial population of 20% for each of the five sublevels was assumed. Figure 6 shows the results. Whereas the $M=0,\pm1$ levels are quickly depopulated, the $M=2$ population initially increases and then slowly decreases. For $3b=10$ the trapping in $M=2$ is very effective and after 6 μs still 30% of initial $F=2$ population is in each of the $M=\pm2$ levels (the other atoms are "lost" to the $F=1$ level of the ground state). Consequently only $\bar{N}=2.6$ photons/atom have on the average been emitted in this time interval. For the lower light intensity ($b=0.8$), however, the trapping is less effective; after 6 μs only 2.5% of the atoms are left in each of the $M=2$ levels and an average number of $\bar{N}=5.7$ photons/atom has been emitted.

For a comparison with experimental data we have calculated the average number of photons $\bar{N}$ emitted from an atom on its way through the laser spot as a function of the laser intensity and for different magnetic fields B. The saturation parameter b defined in (13) is numerically given by $b=26.3I$, with I the laser intensity specified in units of W/cm^2. The calculations were performed for the passage time through the laser spot of $t=8.4$ μs and for the magnetic field strengths used in the experiment. The result of the calculations is shown in Fig. 7. For $B=0$ the fluorescence increases with increasing laser intensity until the limiting value of $\bar{N}=1.2$ is reached [Eq. (3)]. For increasing B an initial increase of the fluorescence is followed by a decrease which is obviously due to an inhibition of the Larmor precession, caused by the laser radiation. For sufficiently high laser intensities the number $\bar{N}=1.2$ is always reached again, corresponding to a perfect trapping in the levels $M=\pm2$. In between we find relative maxima, and for magnetic field $B\geq1$ G the maximum possible value of $\bar{N}=6$ [Eq. (2)] is reached. This clearly demonstrates that (i) a sufficiently high magnetic field is necessary for optimal excitation conditions and (ii) an appropriate laser intensity has to be chosen, *not* an intensity as high as possible.

Comparing Fig. 4 with Fig. 7 we see that the decrease of the fluorescence intensity with increasing laser intensity

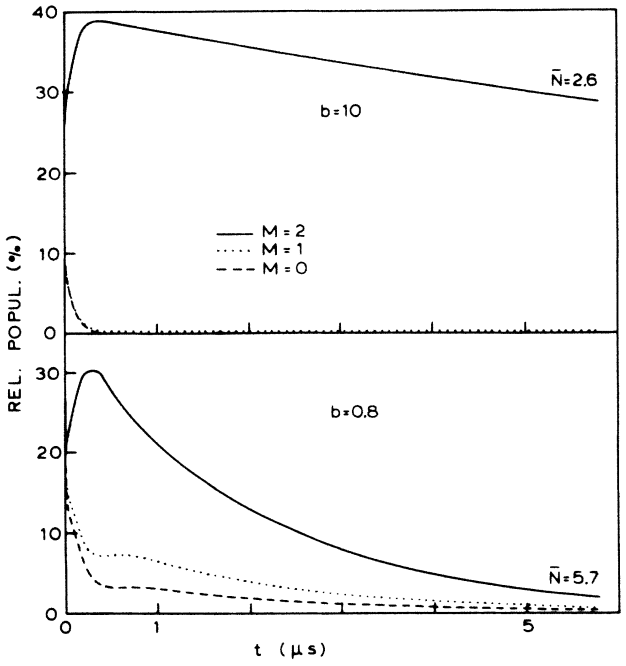

FIG. 6. Time-dependent sublevel populations for the $F=2$ ground state, as calculated by means of Eqs. (14) and (15). A magnetic field of 0.65 G was assumed and a resonant light intensity corresponding to $b=10$ and 0.8, respectively [see Eq. (12)].

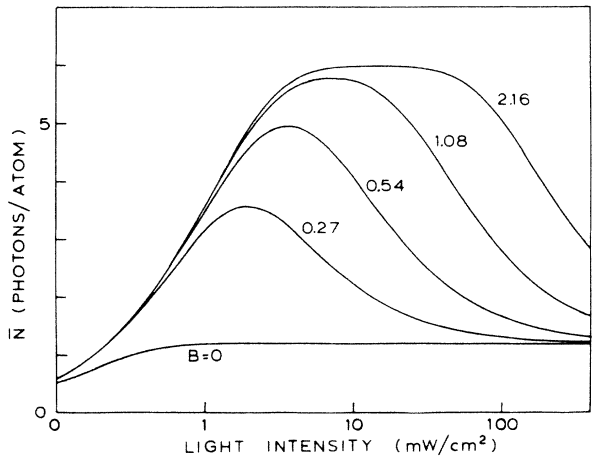

FIG. 7. Average number of photons $\bar{N}$ that are emitted from an atom, calculated by means of Eqs. (14) and (15) as a function of laser intensity. Interaction time with the laser and magnetic field strengths is chosen in accordance with the experimental situation (see Fig. 4).

is experimentally much less pronounced than expected from the calculations. At laser intensities above $I \simeq 20$ mW/cm^2 even a further increase of the fluorescence intensity is observed in the experiment. This is probably caused by saturation broadening of the absorption line which leads to a considerable excitation of the $F=2 \rightarrow F'=2$ transition which has a resonance frequency only $\Delta\nu = 190$ MHz from the $F=2 \rightarrow F'=1$ transitions. However, as an important agreement between measured and calculated curve we find (i) by means of the magnetic field the fluorescence intensity, and thus the number density of excited atoms in the interaction region, can be increased by a factor of 5 and (ii) an *increase* of the laser intensity can lead to a *decrease* of fluorescence intensity. The optimal laser intensity depends on the magnetic field and is correctly predicted by the calculations.

VI. CONCLUSION

We have shown that the Larmor precession can effectively be inhibited by optical pumping. The removal of magnetic sublevel traps by means of a magnetic field is therefore effective only at sufficiently low light intensities. For the transition $(3s)^2S_{1/2}$ $(F=2) \rightarrow (3p)^2P_{1/2}$ $(F'=1)$ in Na we have found that laser light with an intensity of 150 mW/cm^2 is too strong to allow effective Larmor precession in the earth's magnetic field. In order to obtain optimal excitation conditions one has to choose carefully the correct laser intensity, which depends on the magnetic field.

The general description of optical pumping—also in external fields—has of course been given earlier (see Refs. 5 and 6 and references therein). Also an influence of the light intensity of Hanle effect signals has been discussed.[6] However, a decrease of excited state population with increasing light intensity has to our knowledge not yet been discussed and quantitatively been compared with theory.

ACKNOWLEDGMENTS

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