Reversal of the Direction of Population Transfer between Zeeman Sublevels in Optical Pumping

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We have observed a sudden reversal in the direction of population transfer between nearly degenerate Zeeman sublevels during optical pumping of the cesium $6S_{1/2}(F=4)\rightarrow 6P_{3/2}(F'=5)$ cycling transition. As a result of the reversal, the number of atoms in an atomic beam, with quantum numbers $6S_{1/2}$ ($F=4$; $m_F=-4$), is a function of laser frequency and has a sharp dispersive shape. A rate-equation model indicates that similar dispersive shapes for the population can be expected in the optical pumping of any nondegenerate system with more than two levels, if the level spacing is much smaller than the laser linewidth.

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Optical pumping (OP) is a fundamental tool in laser spectroscopy, and plays a central role in many quantum-optics phenomena, including laser cooling and trapping, orientation of atoms possessing hyperfine structure, laser "shelving" and quantum jumps, and atomic parity violation. OP of the cesium hyperfine levels, in particular, has received recent attention in connection with laser cooling of atoms [1-3] and with efforts to improve cesium atomic clocks [4,5]. The importance of OP has been dramatically demonstrated recently in a new laser-cooling mechanism, which relies on laser-polarization-dependent OP between degenerate Zeeman sublevels [3]. In spite of the awareness of the importance of OP among nearly degenerate levels, it has been implicitly assumed in the extensive research dealing with the subject that if the linewidth of the laser and/or transition is much greater than the frequency difference between a group of nearly degenerate transitions, the direction of OP (increasing or decreasing population of a particular state) is independent of laser detuning and is completely determined by laser polarization and atomic selection rules.

In this Letter, we show that such an approach is not correct in predicting the final population of the nearly degenerate Zeeman sublevels. The population of a Zeeman sublevel can be optically pumped or depleted, at the expense or gain of its nearly degenerate neighbors, with the direction of OP depending on the detuning of the laser from the center of the group of unresolved resonances. Most importantly, this behavior is present even if the degeneracy is lifted only slightly, as it is, for example, in almost all possible experiments, by ever-present miniscule residual magnetic fields.

The observation of the sudden reversal in the direction of OP for a particular Zeeman sublevel was made for the $F=4$ ground state of cesium. The linewidths of the transitions between Zeeman sublevels in our experiments are nearly 3 orders of magnitude wider than the frequency separation between these transitions. In spite of this, OP is able to "resolve" a single magnetic sublevel of the $F=4$ hyperfine state and pump atoms into or out of this state depending on the detuning of the laser. The reversal is also predicted by a rate-equation model, which treats each Zeeman sublevel individually. The model, which is far from providing satisfactory quantitative agreement, nonetheless shows that the reversal is a general result, valid for any simultaneously pumped, nondegenerate, multilevel (more than two) system. It predicts the correct functional form of the Zeeman sublevel populations as a function of laser frequency and polarization, but the size of the observed population changes are much larger than the predicted effect, a discrepancy for which we have no conclusive explanation.

The relevant energy levels of Cs are shown in Fig. 1. Each hyperfine level with total angular momentum F consists of $(2F+1)$ Zeeman sublevels, whose degeneracy is slightly lifted in the presence of a tiny magnetic field. Dipole-allowed transitions between these levels obey the selection rules $\Delta m_F=0$ for linearly polarized radiation parallel to the quantization axis (π polarization), and $\Delta m_F=\pm 1$ for linear polarization perpendicular to the quantization axis (σ polarization). The transition of interest to our experiment is the cycling transition, $6S_{1/2}(F=4)\rightarrow 6P_{3/2}(F'=5)$. When this transition is excited by a narrow laser, the sum of the upper- and lower-state populations stays constant, since selection rules allow the $F'=5$ state to decay back only to the $F=4$ state [4].

In the absence of OP, the populations of the Zeeman sublevels in the $F=4$ state are all equal. The relative population between the Zeeman sublevels after OP depends on the polarization of the pumping laser, and the relative transition probabilities of the nearly degenerate transitions [4]. We show in this Letter that in addition to these well-known effects, the population of the Zeeman sublevels is also critically dependent upon laser detuning from resonance. This comes about due to the slight imbalance in the stimulated pumping rates of the nearly degenerate transitions, which shifts from one extremum (the $m_F=-4$ state) to the other (the $m_F=+4$ state) as the laser is tuned across the hyperfine transition. The imbalance is caused by one transition always being slightly more in resonance with the "broad" laser than its neighbors. Although very small, such an imbalance will always lead to a dispersionlike population of the extremum Zeeman sublevels as a function of laser frequency, if the system is allowed to come to equilibrium. For the cycling

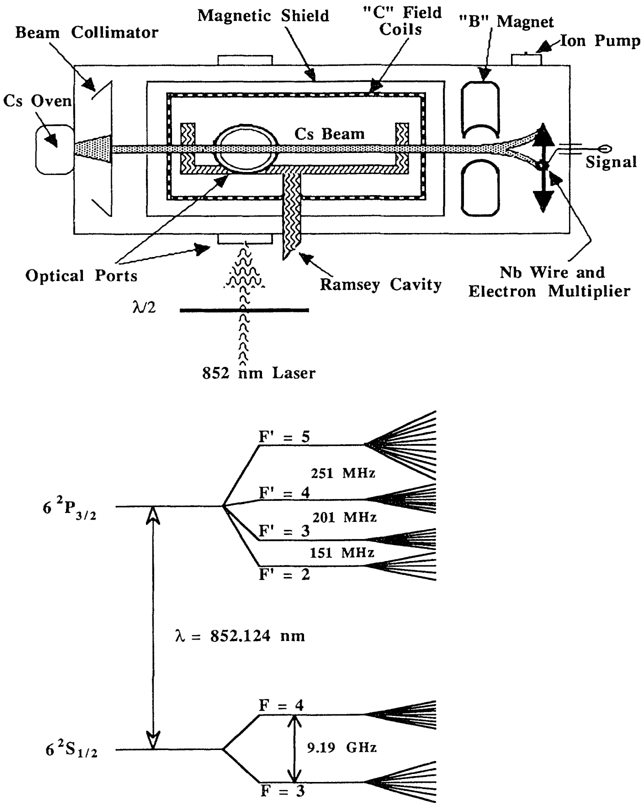

FIG. 1. Modified Cs atomic-clock tube and relevant Cs energy levels. The lifting of the degeneracy of each hyperfine level is shown as a function of a small magnetic field that was applied by the C-field Helmholtz coils.

transition, equilibrium is reached quickly due precisely to the cycling nature of the OP and, thus, the imbalance is the cause for important and observable effects. As the lifting of the degeneracy grows, due, for example, to an increasingly stronger magnetic field, the imbalance also grows, resulting in dispersive shapes with larger peak-to-valley amplitudes.

The experimental apparatus, Fig. 1, consists of a specially modified Cs atomic-clock tube [6]. The conventional state-selection magnet A was removed and four Pyrex viewports provided optical access to the Cs beam in what is normally the low-field, 6×10^{-6}-T, "C-field"-Ramsey section. (This Ramsey cavity, though present in the tube, was not used in our experiment.) The C-field was produced by two variable-current Helmholtz coils, and its direction determined the quantization axis for the atoms. The oven operated at a temperature of $\sim 80\,°\mathrm{C}$, resulting in an atomic beam with $\sim 2\times10^{12}$ atoms/sec, with an $\sim$8-mrad divergence, as measured at the detector end of the tube. The atomic beam was state separated by magnet B ($\sim$1.2 T), into two beams [7]: one with $m_J=+\frac{1}{2}$, and thus referred to as the "plus" beam, having quantum numbers $\{F;m_F\}=\{4;-3,\ldots,+4\}$, and the other with $m_J=-\frac{1}{2}$, the "minus" beam, with quantum numbers $\{F;m_F\}=\{3;-3,\ldots,+3\}$ *and* $\{F;m_F\}=\{4;-4\}$. Either of these beams could be detected by a movable hot niobium wire, which ionized the incident Cs beam, and produced ions which were detected by an electron multiplier [8].

As the laser source we used a single-frequency (linewidth $\sim$2 MHz) dye laser operating with Styril-9M dye, near 850 nm. The laser propagated perpendicular to the atomic beam and parallel to the detector wire, and its polarization could be adjusted to be either π or σ. Laser intensity was kept below 2.0 mW/cm^2, so as to avoid strong saturation and nonlinear processes. The typical 6×10^{-6}-T field in the laser-atom interaction region lifted the Zeeman degeneracy and resulted in a 21-kHz splitting for the $6S_{1/2}(F=4)$ Zeeman sublevels and a 33-kHz splitting for the $6P_{3/2}(F'=5)$ sublevels.

Laser-induced resonances could be detected by either the Cs beam fluorescence at $\sim$852 nm, which was monitored through a separate optical port, or by the changes in the number of atoms (ion current) in one of the two beams (the plus beam in our experiment) emerging from the B magnet. A unique aspect of our apparatus is that the measurement of the plus- (or minus-) beam current *yields the population of a single Zeeman sublevel.* This is because the narrow laser linewidth guarantees that the $6S_{1/2}(F=3)$ states do not interact with the laser and so there is no significant radiative leakage to the $F=3$ state via the $6P_{3/2}(F'=4)$ excited state [4]. Thus the plus beam contains only, and all, atoms with $F=4$, with the exception of atoms with $\{F;m_F\}=\{4;-4\}$. Changes in the plus-beam current near the cycling transition frequency thus represent changes in the population of a single Zeeman sublevel, the $6S_{1/2}(F=4;m_F=-4)$.

In Figs. 2(a) and 2(b) we show the plus-beam current as the laser frequency is scanned over $6S_{1/2}(F=4)\rightarrow 6P_{3/2}(F'=3,4,5)$ transitions for π and σ polarizations. As expected, the current decreases when the laser is in resonance with a transition to the $6P_{3/2}(F'=3,4)$ states, since the atoms can decay from these excited states into the $F=3$ state, and thus end up in the minus beam [4,9]. The smaller, incompletely resolved feature at the $6S_{1/2}(F=4)\rightarrow 6P_{3/2}(F'=5)$ transition is the subject of this Letter. Since this is the cycling transition, the plus-beam current cannot be affected, unless the laser selectively populates or depopulates the $(F=4;m_F=-4)$ level. Not only was this Zeeman sublevel optically pumped, but the direction of pumping depends on laser detuning and polarization. This asymmetric, detuning-dependent pumping of a nearly degenerate Zeeman sublevel is particularly surprising, since the laser linewidth ($\sim$2 MHz) was much greater than the separation of the transitions between Zeeman sublevels ($\sim$10 kHz).

The dispersive shape of the ion current is shown in higher resolution in Figs. 2(c) and 2(d). For σ polarization the $(F=4;m_F=-4)$ state gains population for red detuning and loses population for blue laser detuning, the

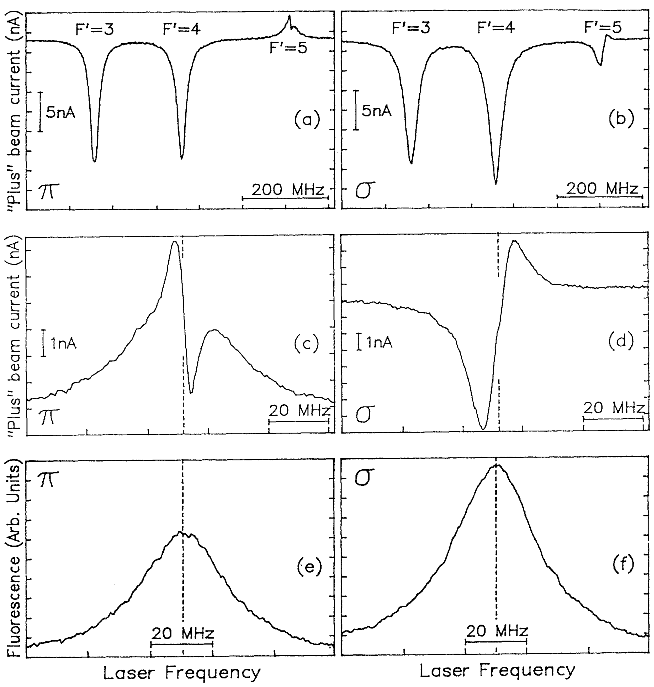

FIG. 2. The "plus"-beam current as a function of laser frequency near the $6S_{1/2}(F=4) \to 6P_{3/2}(F'=3,4,5)$ transitions for the laser with (a) π and (b) σ polarization. The plus-beam current for the $(F=4) \to (F'=5)$ cycling transition on an expanded scale for (c) π and (d) σ polarization. (e),(f) The fluorescence signal of the excited Cs atoms monitored simultaneously with the plus-beam current. The peak of the fluorescence and the reversals of the dispersive shapes in the atomic beam population occur at the same laser frequency as indicated by the dashed lines. The laser power in all the above was 1.4 mW/cm^2 and the C field was 60 mG.

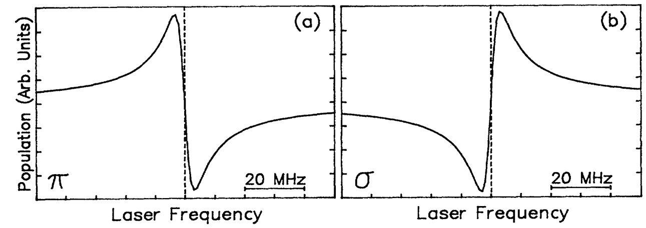

FIG. 3. Calculated population in the plus beam as a function of increasing laser frequency near the cycling transition for (a) π and (b) σ polarization, with a magnetic field of 60 mG and laser power of 0.3 mW/cm^2. The calculated π-polarization line shape was scaled by $\sim 5\times 10^3$ relative to the σ-polarization line shape.

switchover taking place over a very narrow frequency range centered at the $(F=4) \to (F'=5)$ resonance. The simultaneously measured fluorescence signals, shown in Figs. 2(e) and 2(f), on the other hand, look perfectly normal, as they should, since the fluorescence does not state select a specific Zeeman sublevel. (The fluorescence looks somewhat broad due to Doppler broadening from the imperfectly collimated atomic beam which gets better collimated as it propagates down the tube.)

We investigated the dependence of the $(F=4;m_F=-4)$ population on laser polarization, power, and magnitude of the magnetic field responsible for removing the Zeeman degeneracy. The results of these experiments can be summarized as follows: (i) The asymmetry in the population change is centered on line center and is very sharp, with a peak-to-valley width ≤ 10 MHz. (ii) The sign of the asymmetry reverses as the polarization is changed from π to σ. (iii) The shape of the dispersive feature is insensitive to incident laser power over a wide range, 0.3 to 14 mW/cm^2. (iv) The line shape is very sensitive to magnetic field, and changing the field in the range from 0 to $+33$ μT can change the sign of the asymmetry. The signal strength is also a function of magnetic field, and at 33 μT, with σ-polarized light detuned to the red of resonance, we were able to almost completely eliminate the plus-beam current, indicating a total depopulation of the $(F=4;m_F=-3,\ldots,+4)$ sublevels.

The rate-equation model used to analyze these results treats each Zeeman sublevel as a separate state, even if its degeneracy is lifted by only a minute fraction of the laser or natural linewidths. The model requires the solution of eleven equations for the $6P_{3/2}(F'=5)$ Zeeman sublevels of the form

$$\partial N_{5,m}/\partial t = N_{4,m-q}W_{4,m-q\to 5,m} - N_{5,m}W_{5,m\to 4,m-q} - N_{5,m}(A_{5,m\to 4,m} + A_{5,m\to 4,m+1} + A_{5,m\to 4,m-1}), \tag{1}$$

and nine equations for the $6S_{1/2}(F=4)$ sublevels,

$$\partial N_{4,m}/\partial t = -N_{4,m}W_{4,m\to 5,m+q} + N_{5,m+q}W_{5,m+q\to 4,m} + N_{5,m-1}A_{5,m-1\to 4,m} + N_{5,m}A_{5,m\to 4,m} + N_{5,m+1}A_{5,m+1\to 4,m}, \tag{2}$$

where N_{F,m_F} is the population of the $\{F;m_F\}$ state, q can have values of $(+1,-1,0)$ and is the index of the polarization vector along the σ^+, σ^-, and π polarization, and $A_{F,m_F\to F',m_{F'}}$ and $W_{F,m_F\to F',m_{F'}}$ are the spontaneous and induced transition rates derived from the convolution of the laser line shape and the line shape for the transition between the Zeeman sublevels:

$$W_{F,m_F\to F',m_{F'}} = (3\lambda^3/8\pi hc)\left[\int_{-\infty}^{\infty} g_{F,m_F\to F',m_{F'}}(\nu)\rho_L(\nu)\,d\nu\right] I_L A_{F,m_F\to F',m_{F'}};$$

λ is the wavelength of the laser, h Planck's constant, c the speed of light, I_L the laser intensity in W/m^2, $g(\nu)$ the line shape of the atomic transition, $\rho_L(\nu)$ the laser line shape, and units mks. In the numerical calculations we took both $g(\nu)$ and $\rho_L(\nu)$ to be normalized Lorentzians.

Inherent in a rate-equation model are certain assumptions. It is valid only for laser intensities well below the saturation intensity, a restriction which is approximately satisfied in our experiments. It also assumes that the OP is Doppler free, which is appropriate to our experiment. In addition, the induced transition rate is assumed to be time independent, an assumption strictly valid only for a uniform laser intensity profile, which our Gaussian laser profile satisfies only approximately. The model does not include inhomogeneities in the magnetic field, which in the experiment are known to be small ($\sim 10^{-7}$ T). The most severe shortcoming of the model as applied to our experiment is that rate equations are strictly valid only when the laser linewidth is greater than the transition linewidth [10] and, thus, coherences can be ignored. In our experiments these linewidths are nearly equal, and coherent effects are certain to be important, thus requiring the solution of the Bloch equations for the full set of Zeeman sublevels [4,11].

The results of the numerical steady-state solution are shown in Figs. 3(a) and 3(b) for π and σ polarizations, for a magnetic field of 6 μT, and a laser intensity of $\sim$0.3 mW/cm^2 (corresponding to the lowest intensities used in the experiment). The shapes of the dispersionlike features resemble the experimental results, and the model predicts the correct symmetry and reversal of the dispersive shape for the two different laser polarizations. We also obtain qualitative agreement between the calculation and experimental dispersive shape as a function of magnetic field. The model, however, completely fails to quantitatively predict the effect, which is in some cases more than 3 orders of magnitude larger than predicted, especially for π polarization.

To account for the large discrepancy between the experiment and calculation we examined the nonequilibrium (time-dependent) solution of the rate equations. The numerical results indicate that for our experimental conditions, steady-state solutions are reached for interaction times of $\sim$50 μsec, as long as one is concerned only with the region within $\pm$20 MHz of line center. In a wider region of $\pm$100 MHz from line center, steady-state solutions are not reached even for a 1-msec atom-laser interaction time. The experimental interaction time in our apparatus is on the order of 40 μsec, and the main features of the dispersive shape are within $\pm$15 MHz of line center. Steady-state solutions for this feature are therefore appropriate, and the discrepancy cannot be ascribed to transient behavior [12]. Other possible causes for the discrepancy, such as nonadiabatic (Majorana) transitions were also calculated and found to be of no importance [8]. Only the full Bloch equation calculation will elucidate if this discrepancy can be ascribed to coherences between the Zeeman sublevels.

In conclusion, we demonstrated that the population of a single nondegenerate but unresolved sublevel has a dispersive shape as a laser is tuned across the resonance. The dispersive shape is very sharp, centered at line center, and its sign and magnitude depend on laser frequency, polarization, and the magnitude by which the degeneracy is lifted. A model based on rate equations qualitatively predicts the effect, but completely fails in predicting the size of the observed dispersive shapes.

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[1] A. Aspect *et al.*, Phys. Rev. Lett. **57**, 1688 (1986); C. Salomon *et al.*, Europhys. Lett. **12**, 683 (1990).
[2] C. Monroe *et al.*, Phys. Rev. Lett. **65**, 1571 (1990).
[3] S. Chu *et al.*, in *Atomic Physics 11*, edited by S. Haroche, J. Gay, and G. Grynberg (World Scientific, Singapore, 1989), p. 636; J. Dalibard *et al.*, *ibid.*, p. 199. For a recent review see C. Cohen-Tannoudji and W. Phillips, Phys. Today **43**, No. 10, 33 (1990).
[4] M. Arditi and J. Picque, J. Phys. (Paris), Lett. **41**, 379 (1980); E. de Clercq *et al.*, J. Phys. (Paris) **45**, 239 (1984); G. Avila *et al.*, Phys. Rev. A **36**, 3719 (1987); D. Wang *et al.*, Chin. Phys. Lett. **5**, 97 (1988).
[5] D. Sesko and C. Wieman, Opt. Lett. **14**, 269 (1989); C. Monroe *et al.*, Opt. Lett. **16**, 50 (1991).
[6] The tube is manufactured by TFL Ltd., Jerusalem, Israel.
[7] N. Ramsey, *Molecular Beams* (Clarendon, Oxford, 1969).
[8] Y. Nafcha, M.S. thesis, Bar-Ilan University, Ramat Gan, Israel, 1990 (unpublished).
[9] V. Giordano *et al.*, Metrologia **25**, 17 (1988); G. Theobald *et al.*, Opt. Commun. **71**, 256 (1989).
[10] C. Cohen-Tannoudji, in *Atomic Physics IV*, edited by G. Zu-Putlitz, E. Weber, and A. Winnacker (Plenum, New York, 1975), p. 589.
[11] E. de Clercq and P. Mangin, in Proceedings of the Third European Time and Frequency Forum, Besancon, France, 21–23 March 1989 (unpublished), pp. 281–292.
[12] The reason why the experimental dispersion seems to sit atop a broader structure is that at larger than $\pm$15-MHz detuning the population has not reached steady-state equilibrium.