Optical Nutation in Cold ^{85}Rb Atoms

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We have observed optical nutation in cold ^{85}Rb atoms with a negligible Doppler broadening. The optical nutation of a two-level atom arranged by optical pumping has been studied as a function of detuning frequency and Rabi frequency. The change of the nutation signal caused by magnetic substate degeneracy has also been observed for σ and π excitations. This can be explained by optical nutation beatings from different transition probabilities among magnetic sublevels. Absolute transition probabilities with σ and π transitions and a branching ratio between them have been measured.
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KEYWORDS: rubidium, nutation, trap, transition probability, cold atom, branching ratio

1. Introduction

A two-level atom that is irradiated by a resonant or nearly resonant electromagnetic field will undergo transient oscillations between its ground and exited states. Prior to reaching a steady state the oscillations arising from successive atomic absorptions and stimulated emissions, are known as the "optical nutation" and are characterized by the generalized Rabi frequency Ω given by

$$\begin{aligned}\Omega &= [(4\pi^2/h^2)[(\mu_{12}\cdot E_0)^2 + (\omega-\omega_0)^2]]^{1/2} \\ &= [\Omega_R^2 + \delta^2]^{1/2}\end{aligned} \tag{1}$$

where μ_{12} is the matrix element of the electric dipole moment, E_0 the amplitude of the incident electric field, and $\delta = (2\pi/h)(\omega-\omega_0)$ the detuning between the frequency of the exciting field and the atomic transition frequency. In zero detuning, Ω equals the Rabi frequency

$$\Omega_R = (2\pi/h)(\mu_{12}\cdot E_0).$$

The reason this transient oscillation is often called the "optical nutation" is that it is the optical analogy of the spin nutation in nuclear magnetic resonance (NMR).

Hocker and Tang[1)] observed the first optical nutation of SF_6 gas using a Q-switched CO_2 laser tuned to the 10.57 µm line. They noticed that an optical nutation can be a very good tool for measuring transition probabilities. It does not require the estimation of population density which is different from the conventional method based on the measurement of absorption coefficient. To obtain more accurate transition probabilities, Brewer and Shoemaker[2,3)] used a Stark switching technique. Shoemaker and van Stryland[4)] obtained NH_2D molecular transition probabilities with a 5–8% accuracy in the microsecond time range using the Stark switching technique. Golub *et al.*[5)] have seen an optical nutation in Yb atomic beam experiments. They have shown that atomic fluorescence is maximized when Yb atoms are purely in their excited states rather than in a superposition of the ground and excited states using optical nutation and optical spin locking,[5)] which enable the preparation of the atoms for selective dressed states.[6,7)] Wännström *et al.* observed an optical nutation in a fast-ion-beam (Ba II) laser experiment by a rapid Doppler-shift technique. They measured absolute transition probabilities in the Ba II ion and observed frequency beats in an optical nutation due to the effects of magnetic substate degeneracy.[8–10)] Wei *et al.*[11)] observed a dressed-state nutation by a Raman heterodyne technique using nuclear magnetic transitions in diamond. The reduced Rabi frequencies associated with the dressed states for a three-level system were measured for the first time. Wu *et al.*[12,13)] studied the transient excitation of dressed atoms in two-level atoms using a bichromatic field consisting of a strong resonant component and a weak nonresonant component. Toyoda *et al.*[14)] observed optical free induction decay (FID) instead of the optical nutation of laser-cooled ^{85}Rb atom in their study of optical coherent transient effects.

To observe an optical nutation, the Rabi frequency of the optical field should be higher than those of the excited state relaxation rate and dipole moment decay rate. Otherwise, the nutation would be damped out before it could show one complete period. Most previous nutation experiments have been performed on systems with hyperfine levels consisting of degenerate magnetic sublevels with unequal electric dipole matrix elements. Frequency beatings due to these different nutation frequencies can be analyzed using complicated theoretical equations.[8–10)] In our nutation experiment, we prepared a two-level system with a homogenous sample prepared in a magneto optical trap (MOT) and a circularly polarized optical pumping pulse.

An idea known as the Stark shift technique[2,3,15,16)] using a narrow continuous wave (CW) laser has been proposed to reduce the inhomogeneity of a gas sample. Another method is to use an atomic beam as a sample and observe a nutation signal in the fluorescence of the atoms. In this experiment we avoid the huge velocity distribution of the atoms at room temperature by applying laser cooling techniques. Until now, previous researchers have only used a dressed state nutation in the microwave range[11)] or in two-level atomic systems.[12,13)] Cold atoms are localized in space, unlike an atomic beam which has a tremendous velocity in one direction. Thus, we can obtain a signal not only from the atomic fluorescence but also from the coherent radiation from the atomic polarization in the same propagation direction as the excitation pulse. We used this signal to study the characteristics of an optical nutation, the nutation frequency beatings and to measure the transition probabilities from $[5S_{1/2}F = 3\ (m_F = 3)]$–$[5P_{3/2}F' = 4\ (m'_F = 4)]$

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and $[5S_{1/2}F = 3\ (m_F = 3)]$–$[5P_{3/2}F' = 4\ (m'_F = 3)]$ with a carefully prepared two-level atom and branching ratios.

2. Theory

We use a semiclassical density matrix formula to describe an optical nutation in a two-level system.
The set of density matrix equations becomes the optical Bloch equations in the rotating frame[17] given as

$$\begin{aligned} \frac{du}{dt} &= -\delta v - \frac{u}{T_2} \\ \frac{dv}{dt} &= \delta u - \frac{v}{T_2} + \Omega_R w \\ \frac{dw}{dt} &= -\frac{w - w_{eq}}{T_1} - \Omega_R v \end{aligned} \tag{2}$$

where δ is the detuning, w the single-atom population difference (inversion), u and v the atomic dipole moments in-phase and in-quadrature with the applied field, Ω_R the Rabi frequency and T_1, T_2, and w_{eq} are the longitudinal and transverse homogeneous lifetimes and -1 in thermal equilibrium, respectively. An optical nutation can be illustrated graphically using a vector model. The Bloch vector M has components (u, v, w). In the absence of atomic relaxation, the above three equations become

$$\frac{dM}{dt} = M \times \Omega \tag{3}$$

where $\Omega = (\Omega_R, 0, \delta)$. This equation shows that the Bloch vector M processes at approximately the effective field Ω. Suppose that a sample is initially in thermal equilibrium. Then u and v are zero, so that M points straight down along the w axis. When an exactly resonant optical field is turned on, Ω points along the u axis and causes M to precess in the w–v plane. As M precesses, the population difference w changes, showing that we are driving the system up to the excited state and down to its original state. In addition, the v component of M, which is proportional to the out-of-phase component of the oscillating dipole moment induced in the samples, changes its sign depending on whether the M is moving upward or downward. This sign determines whether the samples are absorbing or emitting radiation. In the case of an off-resonant optical field, Ω lies somewhere in the u–w plane. M precesses at approximately Ω with a frequency of $\Omega = [(4\pi^2/h^2)[(\mu_{12} \cdot E_0)^2 + (\omega - \omega_0)^2]]^{1/2}$. We solve the optical Bloch equations numerically by assuming that the excitation pulse has a Gaussian rise time.

The steady-state solutions $(t = \infty)$ of eq. (2) for each component of the Bloch vector can be easily obtained by inserting the differential terms zeros.[17]

$$\begin{aligned} u(\infty, \delta) &= w_{eq} \frac{\delta T_2 \Omega_R T_2}{1 + T_2^2\delta^2 + T_1 T_2 \Omega_R{}^2} \\ v(\infty, \delta) &= -w_{eq} \frac{\Omega_R T_2}{1 + T_2^2\delta^2 + T_1 T_2 \Omega_R{}^2} \\ w(\infty, \delta) &= w_{eq} \frac{1 + \delta T_2^2}{1 + T_2^2\delta^2 + T_1 T_2 \Omega_R{}^2} \end{aligned} \tag{4}$$

Where, w_{eq}, T_1 and T_2 are -1, 27 ns and 54 ns, respectively. We will use these equations to plot the atomic polarization related to u and v as a function of detuning and Rabi frequency in the steady state.

3. Experiment

In our experiments, cold ^{85}Rb atoms are prepared in a MOT from room-temperature vapor.[18,19] The temperature of the atoms is approximately 100 µK measured by a new ground-ground state coherent transient effect method.[20] The optical depth of the trapped atoms measured by the scanning frequency of the very weak probe beam is ≈1 except for the transition probability measurement. To reduce the effects due to the use of a dense sample and to obtain an accurate transition probability, we prepared trapped atoms with ≈30% absorption depth. 12 ms after the trapping laser beams and gradient magnetic field are turned off, two optical pulses are applied with a small magnetic field (≈300 mG) along the quantization axis. The first pulse which has an on-resonant intense σ^+ circular polarization with a 5–10 µs duration optically pumps the atoms into the $F = 3$, $m_F = 3$ magnetic sublevel, while the second pulse is the nutation probe pulse with a 500–1000 ns duration. To measure the absolute transition probability between $5S_{1/2}F = 3$ $(m_F = 3)$–$5P_{3/2}F' = 4$ $(m'_F = 4)$, the polarization of this probe pulse is the same as that of the optical pumping pulse with σ^+ circular polarization as shown in Fig. 2. When we measure the transition probability between $5S_{1/2}F = 3$ $(m_F = 3)$–$5P_{3/2}F' = 4$ $(m'_F = 3)$, a linearly polarized probe pulse instead of a circular polarized probe pulse is applied perpendicular to the direction of the optical pumping pulse (π transition). The optical nutation signal is observed with two high-speed photodiodes whose rise times are shorter than 1 ns. Two laser pulses should arrive at the same time on the photodiodes to avoid an unwanted artificial oscillation at the onset of the signal. Even if the pulses arrive at the same time, the different rise times of the photodiodes can induce artificial oscillation. We used photodiodes with nearly the same rise times and subtracted each balanced signal without atoms from each balanced signal with atoms. Figure 1 shows the experimental data (dotted line) and a least-squares fit (solid line) obtained by solving the two-level Bloch

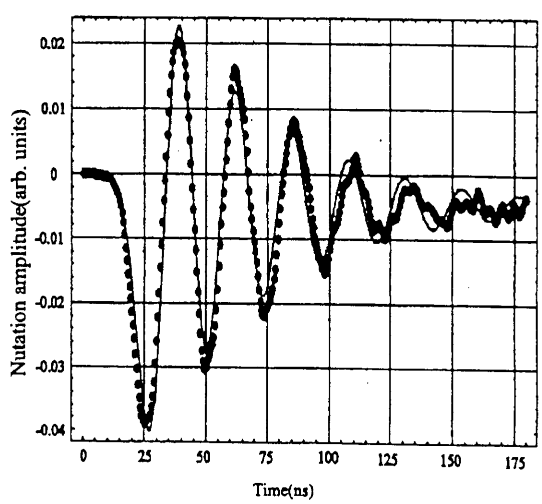


Fig. 1. Experimental data (dotted line) and theoretical fit (solid line) for nutation amplitude with respect to time. The experiment was performed at a 40.8 MHz detuning and a 5.35 mW laser power which correspond to a 13.4 MHz Rabi frequency.

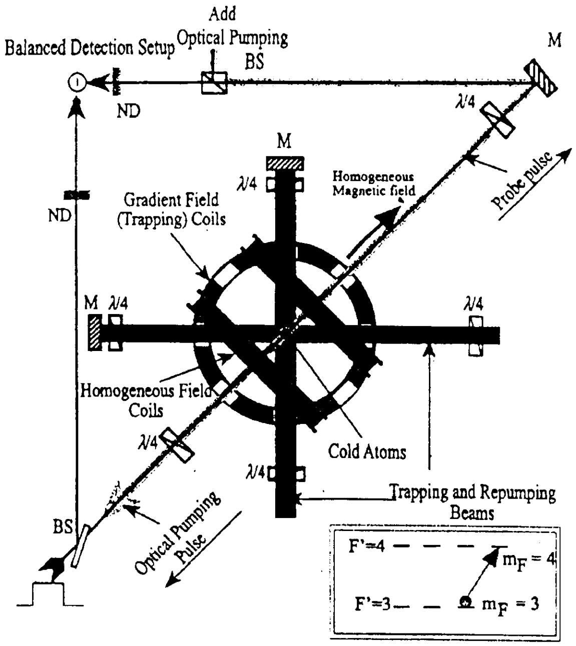


Fig. 2. Nutation experimental setup for σ^+ transition. The polarization of the probe pulse is the same as that of the optical pumping pulse with σ^+ circular polarization.

equation. The experimental results agree well with the fitted results. The experiment was performed with a 40.8 MHz detuning and a 5.35 mW power.

When we measure transition probability, all the atoms should observe the same Rabi frequency. The center portion of the Gaussian beam with a full width half maximum (FWHM) of 4 mm diameter is aligned carefully to hit the trap of approximately 0.4 mm in diameter. To prevent too much expansion of the trap after turning off the trapping laser and anti-Helmholtz coil, the trapping laser was turned off 10 ms after turning off the anti-Helmholtz coil. The optical pumping and excitation pulses are applied only 2 ms after turning off the laser and 12 ms after turning off the anti-Helmholtz coil. For the frequency stabilization of the pump and probe lasers, frequency modulation (FM) spectroscopy is used. We found that the frequency jitter is less than several hundreds kHz. The optical pumping and excitation pulses are generated by an acousto-optic modulator (AOM) diffracting a single-mode CW Ti:sapphire laser (Coherent 899 ring laser) pumped by an argon ion laser (Coherent Innova 400).

4. Observations and Discussion

The frequency of an optical nutation in a two-level system is related simply to the detuning and Rabi frequency which is proportional to the electric field of the excitation pulse and dipole moment matrix element. Figure 3 shows the nutation frequency and steady-state value of atomic polarization as functions of various detunings at the same Rabi frequency. We used the following equation for atomic polarization (P) which is related to the atomic dipole moments u and v.

$$P(\Omega_R, \delta) = \frac{a\Omega_R{}^2 T_2}{1 + T_2^2\delta^2 + T_1 T_2 \Omega_R{}^2} \quad (5)$$

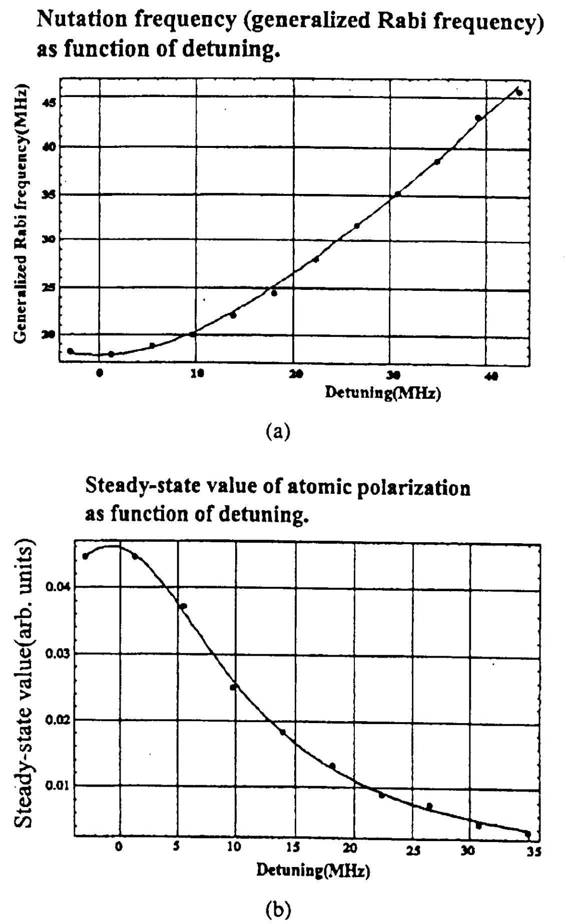


Fig. 3. (a) Nutation frequency and (b) steady-state value of atomic polarization as functions of detuning at the same laser power (1.13 mW). The solid curves are fits to the experimental data (Rabi frequency is 17.98 MHz).

Where, a is arbitrary amplitude.

As we can see from the figure, when the detuning is much higher than the Rabi frequency, the steady state values of atomic polarization are ~12 times smaller than those at 35 MHz detuning. Thus, nutation frequency can be obtained using the square of the Fourier transform of the nutation signal. However, when the detuning is close to zero, the steady state value of atomic polarization increases, preventing us from obtaining the right frequency through Fourier transformation. In this case, it is better to determine the positions of the maxima and minima of a nutation in the time domain. In Fig. 3, the solid lines are theoretical fits of the nutation frequency and steady state value as functions of detuning when the Rabi frequency is fixed. We have nearly the same Rabi frequency from the fits, assuming that T_1 is 27 ns. The intensity dependences of the nutation frequency and steady state value for detuning 1.3 MHz (close to resonance) are shown in Fig. 4.

Also, the observation of frequency beatings in an optical nutation due to magnetic sublevel degeneracy is an interesting subject. These types of beatings have been observed by Kastberg *et al.*[10] in a Ba II ion beam. The optical nutation obtained was compared with full quantum mechanical calculations including the degeneracy of energy

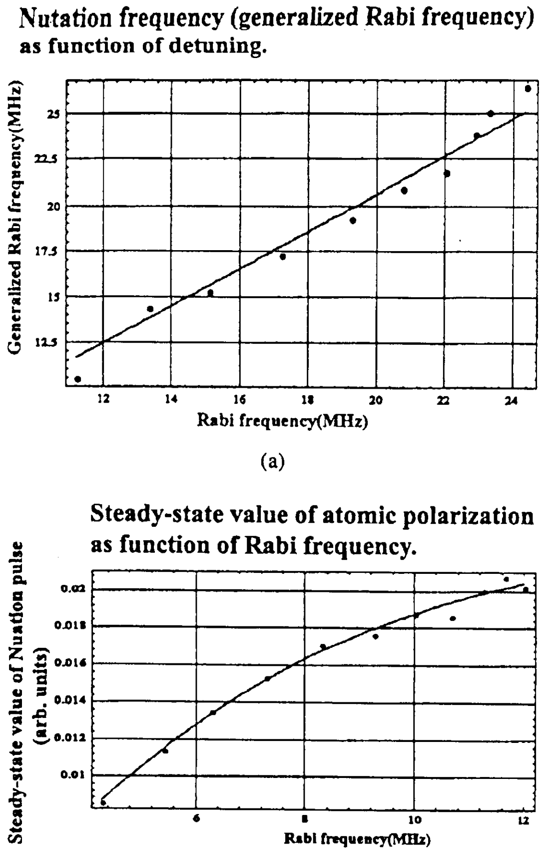


Fig. 4. (a) Nutation frequency and (b) steady-state value of atomic polarization as functions of Rabi frequency by maintaining 1.3 MHz detuning. The solid curves are fits to the experimental data.

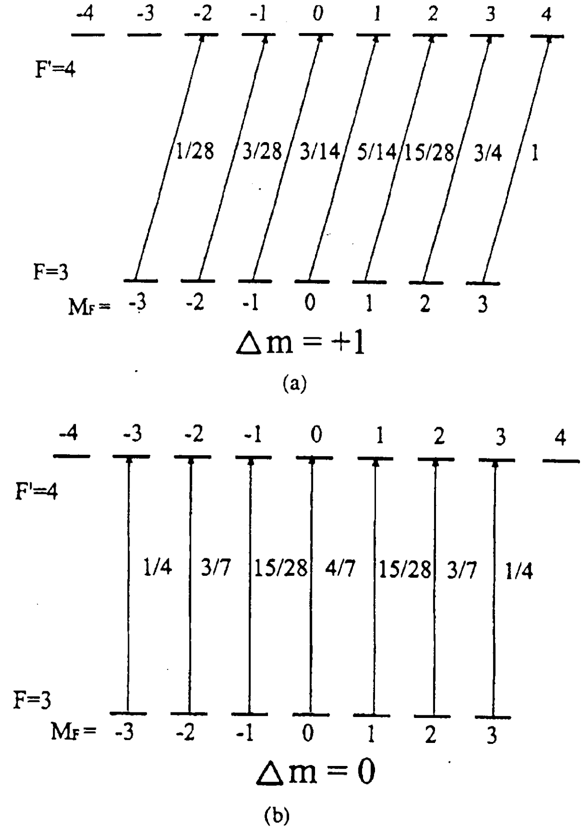


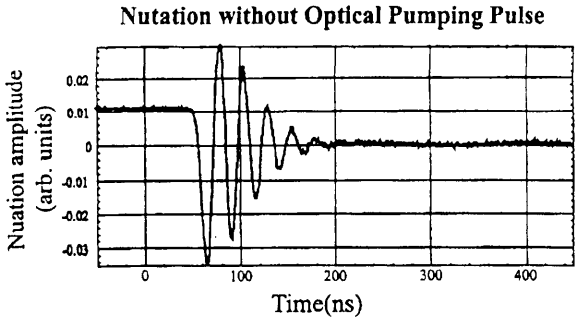
Fig. 5. Relative transition probabilities of $5S_{1/2}F = 3$–$5P_{3/2}F' = 4$ for (a) σ^+ excitation pulse ($\Delta m = +1$) and (b) π excitation pulse ($\Delta m = 0$).

levels and spontaneous emission. In our case, when a ^{85}Rb atom is in thermal equilibrium, many magnetic sublevels are involved for $5S_{1/2}F = 3$–$5P_{3/2}F' = 4$ transitions. Without solving the equations of motion for density matrix elements including every possible transition between magnetic sublevels, we can roughly estimate the effects from the different dipole moments of the magnetic sublevels. The decay mechanism of a nutation is not simply associated with the excited state or dipole moment relaxation like that for a FID. Even an optical FID having a simple decay mechanism in homogeneous media shows a dependence on the density of trapped atoms. According to Schenzle and Brewer,[21] for an isolated two-level system consisting of excited and non-decaying ground states in homogeneous samples, the Bloch vector component $v(t)$ is given by

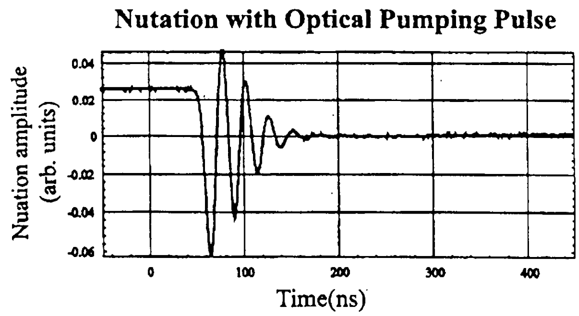
$$v(t) = \Omega_R \omega_0 \exp\left[-\left(\frac{1}{T_2} + \frac{\Omega_R^2}{2T_2(\delta^2 + \Omega_R^2)}\right)t\right] \times \frac{\sin(\delta^2 + \Omega_R^2)^{1/2}t}{(\delta^2 + \Omega_R^2)^{1/2}} \qquad (6)$$

Where Ω_R is the Rabi frequency. This shows that an optical nutation has a Rabi-frequency-dependent damping term. In Fig. 5, the relative transition probabilities of σ^+ [Fig. 5(a)] and π excitations [Fig. 5(b)] for $5S_{1/2}F = 3$–$5P_{3/2}F' = 4$ including all magnetic sublevels are shown. If an atom is optically pumped by a σ^+ circularly polarized optical pumping pulse to $m_F = 3$, a nutation coming from a pure two-level system could be obtained. We checked the absorption signals of a pure two-level system using a scanning laser near the resonance frequency. Without optical pumping, we obtained absorption signals due to different magnetic sublevels except for that of a pure two-level transition.

Nutations from optically pumped atoms and thermally equilibrated atoms are shown in Fig. 6. Note that the decay of a nutation with the optically pumped atoms is shorter than that with the thermally equilibrated atoms. This is due to the Rabi-frequency-dependent nutation damping when $\delta \ll \Omega_R$. The nutation for a transition with a high transition probability decays fast due to the high Rabi frequency. Because a nutation lasts on the order of T_2, we can establish these individual nutations from 7 different transitions that act independently, by considering a nutation from atoms in thermal equilibrium. The total nutation signal is assumed to be obtained by adding each nutation from different Rabi frequencies. Most of them (6 out of 7) have smaller transition probabilities. A nutation from a smaller Rabi frequency contributes to a greater signal at a later time. A nutation frequency difference between optically pumped and thermally equilibrated atoms is also expected. Figure 7 shows this difference in nutation frequency between with and without optical pumping for (a) an excitation pulse with circular polarization (σ^+) and (b) an excitation pulse with linear polarization (π). In the top figure, the nutation

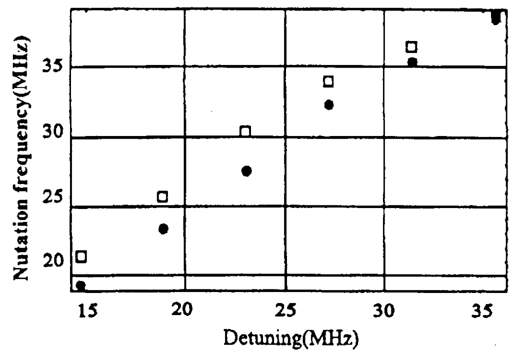


(a)

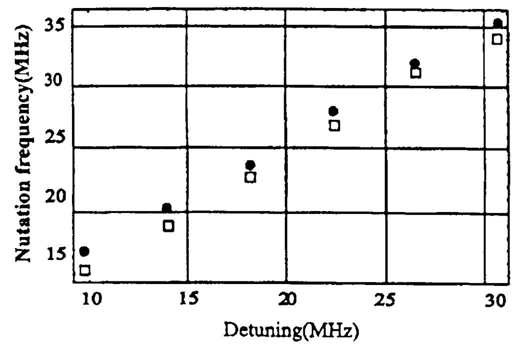


(b)

Fig. 6. Optical nutation amplitude with respect to time (a) without and (b) with optical pumping pulse.



(a)



(b)

Fig. 7. Nutation frequencies with (square) and without (circle) optical pumping as functions of detuning (a) when σ^+ excitation pulse is applied and (b) when π excitation pulse is applied.

frequency difference with a circularly polarized excitation pulse is displayed as a function of detuning at the same intensity. When an atom is polarized by optical pumping, the highest Rabi frequency at the same electric field is obtained. We expect a higher nutation frequency with polarized atoms than that with equally populated atoms at magnetic sublevels. The coherent excitation of degenerate two-level atoms can be regarded as a collection of independent non-degenerate two-level systems. A total nutation signal is obtained by adding each nutation at different Rabi frequencies. The frequencies of the total nutation signal are attained using the Fourier transform of the added signal in the time domain. Figure 7(b) shows nutation frequency with a linearly polarized light (π transition) as a function of detuning at the same intensity. As one can see, the nutations from the polarized atoms have lower oscillation frequencies. This is because the transition probability of π transition for the polarized atoms is smaller than the average transition probability of π transitions for the equally populated atoms at magnetic sublevels. Another difference between the nutation signals of the polarized and thermally equilibrated atoms is observed in their signal amplitude which is proportional to Rabi frequency. Nutation frequencies with [Fig. 8(a)] and without [Fig. 8(b)] optical pumping are shown as functions of detuning. Squares denote experimental data and circles theoretically calculated frequencies. In Fig. 8, the measured nutation frequencies (squares) agree with theoretically calculated frequencies (circles) very well.

5. Transition Probabilities

Three different methods can be used to obtain accurate transition probability. One is to evaluate the Fourier transform of the data. However, this method is only possible when the detuning of the excitation pulse is large. To obtain an accurate value, the experiments should be performed at approximately zero detuning. The second technique is to locate the maxima and minima of the nutation data, but this does not give an accurate value. The third approach is to make a fit to the theoretical model with nutation frequency as a free factor. We used the theoretical fitting without including the effect of the dense atoms. Thus we probed the absorption of well-balanced trapped atoms in space with a very weak CW beam and a small pinhole, and obtained a ≈30% absorption depth to avoid those problems. Detunings can be determined using the absorption method by scanning several calibrated periods or the beat frequency of the FID heterodyne fixed local oscillator and coherent radiation from dipole moments. We determined a detuning of less than 0.5 MHz.

The measured transition probability for $5S_{1/2}F = 3$–$5P_{3/2}F' = 4$ $(m'_F = 4)$ is $3.758 \pm 0.179 \times 10^7$ (s^{-1}) and that for $5S_{1/2}F = 3$–$5P_{3/2}F' = 4$ $(m'_F = 3)$ is $9.764 \pm 1.24 \times 10^6$ (s^{-1}). The ratio of the two transition probabilities from our measurements is 3.85, which agrees well with the theoretical value 4.

6. Conclusions

We observed an optical nutation in a homogeneous broaden medium composed of ^{85}Rb atoms confined in a MOT. This MOT sample can be prepared as an ideal two-level system with the optical pumping and polarization of an

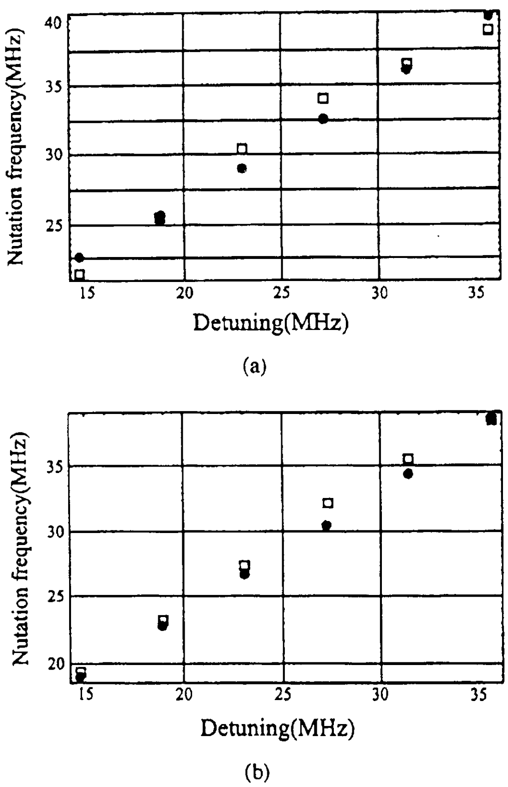


Fig. 8. Nutation frequencies (a) with and (b) without optical pumping as functions of detuning. Squares denote experimental data and circles theoretically calculated frequencies.

excitation pulse, which is fairly difficult to satisfy in most systems due to magnetic sublevels and Doppler broadening. A nutation coming from a purely two-level system has been achieved. Nutation beating involving atoms from many different magnetic sublevels has been obtained and studied as a function of Rabi frequency and detuning. We obtained the absolute transition probabilities for $5S_{1/2}F = 3$–$5P_{3/2}F' = 4$ $(m'_F = 4)$ and $5S_{1/2}F = 3$–$5P_{3/2}F' = 4$ $(m'_F = 3)$ using an optical nutation signal from a carefully prepared two-level system.

1) G. Hocker and C. Tang: Phys. Rev. Lett. **21** (1968) 591.
2) R. Brewer and R. Shoemaker: Phys. Rev. Lett. **27** (1971) 631.
3) R. G. Brewer and R. L. Shoemaker: Phys. Rev. A **6** (1972) 2001.
4) R. Shoemaker and E. Stryland: J. Chem. Phys. **64** (1976) 1733.
5) J. Golub, Y. Bai and T. Mossberg: Phys. Rev. A **37** (1988) 119.
6) Y. Bai, A. Yodh and T. Mossberg: Phys. Rev. Lett. **55** (1985) 1277.
7) Y. Bai, T. Mossberg, N. Lu and P. Berman: Phys. Rev. Lett. **57** (1986) 1692.
8) A. Wännström, A. Kastberg, A. Arnesen, R. Hallin, C. Nordling and O. Vogel: Phys. Rev. A **41** (1990) 440.
9) A. Kastberg, P. Villemoes, A. Arnesen, F Heijkenskjöld, A. Langereis, P. Jungner and S. Linnaus: J. Opt. Soc. Am. B **10** (1993) 1330.
10) A. Kastberg, P. Villemoes, A. Arnesen, F Heijkenskjöld, A. Langereis, P. Jungner and S. Linnaus: Opt. Commun. **101** (1993) 25.
11) C. Wei and N. Manson and J. Martin: Phys. Rev. Lett. **74** (1995) 1083.
12) Q. Wu, D. Gauthier and T. Mossberg: Phys. Rev. A **50** (1994) 1474.
13) Q. Wu, D. Gauthier and T. Mossberg: Phys. Rev. A **49** (1994) R1519.
14) K. Toyoda, Y. Takahashi, K. Ishikawa and T. Yabuzaki: Phys. Rev. A **56** (1997) 1564.
15) R. Brewer: Phys. Today **30** (1977) 50.
16) R. Brewer and R. Smith: *Very High Resolution Spectroscopy* (Academic Press, New York, 1976).
17) L. Allen and J. Eberly: *Optical Resonance and Two-Level Atoms* (Dover, New York, 1975).
18) E. Raab, M. Prentiss, A. Cable, S. Chu and D. E. Pritchard: Phys. Rev. Lett. **59** (1987) 2631.
19) B. Dubetsky and P. Berman: Laser Phys. **4** (1994) 1017.
20) B. Dubetsky and P. R. Berman: Appl. Phys. B **59** (1994) 147.
21) A. Schenzle and R. Brewer: Phys. Rev. A **14** (1976) 1756.