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Optical Free Induction Decay in Cold ^{85}Rb Atoms

Unyob SHIM*, Sidney CAHN, Anantharaman KUMARAKRISHNAN, Tycho SLEATOR and Jin-Tae KIM†

Department of Physics, New York University, New York, NY 10003, U.S.A.
†*Department of Photonic Engineering, Chosun University, Gwangju 501-759, Korea*

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We have observed optical free induction decay (FID) in cold ^{85}Rb atoms. The shortening of the FID lifetime with increasing optical depth of the trap has been found. This shortening is explained by an anomalous absorption for FID propagating in a homogeneous and resonant sample. Theoretical calculations are consistent with experimental results. The Rabi oscillations of FID amplitude have been observed by increasing the pulse area. [DOI: 10.1143/JJAP.41.3688]

KEYWORDS: MOT, FID, Rb, Rabi, cooling, lifetime

1. Introduction

Recent dramatic developments in laser cooling and trapping techniques for neutral atoms have opened up various atomic experimental areas. Coherence transient experiments are refocused on the laser cooled atoms due to their negligible Doppler width, high density and locality in space. Coherent transient effects of the cold atoms involving atomic ground and excited states[1–5] as well as ground and ground state levels[6,7] have been recently performed. In particular, echo schemes creating coherence between ground-state sublevels are used to measure precise values of h/m_e,[6,8] gravitational acceleration,[6,9] and rotation.[10]

When a resonant or near-resonant excitation laser pulse interacts with two-level atoms, the wave radiated freely by the atoms induced by the excitation pulse is extinguished following the excitation pulse. This radiation, which comes from coherently coupled dipole moments along the propagation direction of the excitation pulse, is known as free induction decay (FID).[11,13] In a homogeneously broadened ensemble of atomic systems, the FID signal decays exponentially with the dipole moment decay time T_2. In the case of an inhomogeneously broadened sample, such as a room temperature vapor, groups of atoms with different velocities have different resonant frequencies. Dephasing from different frequencies reduces the lifetime of the FID to below T_2. If the Rabi frequency of the excitation pulse is higher than the inhomogeneous broadening $\Delta\omega_{\mathrm{inh}}$, the FID will decay with a characteristic time of $(\Delta\omega_{\mathrm{inh}})^{-1}$. In the trapped atom, whose inhomogeneous broadening (approximately 0.5 MHz) is much smaller than homogeneous broadening (5.9 MHz), the exponential lifetime of FID is expected to be T_2. As Toyoda *et al.*[2] pointed out, the study of pulse propagation with laser-cooled atoms is interesting due to their high density and homogeneity. Shortening of FID according to the optical depth has been observed. We attempt to explain this shortening by the propagation of an exponential pulse having a small pulse area through a resonant media and a classical abnormal absorption for pulse widths smaller than the decay time of dipole moment.

Another experiment will be presented involving the Rabi oscillations[14,15] of FID when the pulse area is gradually increased to the order of π. When a two-level atom interacts with a resonant electromagnetic field, the probability for each population undergoes transient oscillations between its ground and excited states with the Rabi frequency $\mu E/\hbar$. This Rabi oscillation[11,13] in an optical interaction is clearly observed by using a short (compared with the excited state lifetime), coherent and intense light pulse and measuring the fluorescence after the excitation pulse. Because the fluorescence is proportional to the excited state population, the integrated fluorescence as a function of the pulse area (θ) should show Rabi oscillations. This observation has been made by Gibbs[14] with a ^{87}Rb atomic beam under a high magnetic field involving three levels. Similar oscillations are expected with coherent radiation of atomic polarization. Many attempts to observe the Rabi oscillations by probing the population as a function of initial field have been made. In the latter part of this paper we will discuss the Rabi oscillations of FID which is one of the coherent radiations of atomic polarization.

2. Experimental Setup

Cold ^{85}Rb atoms are prepared in a magneto-optical trap (MOT).[16] The Doppler width and temperature of the cold atoms are measured by a new technique which measures the ground-ground state coherent transient effect[17,18] and found to be approximately 0.5 MHz and 200 μK, respectively. At this temperature, the sample can be considered a homogeneous medium. Optical pumping and an excitation pulse are applied 3 or 4 ms after the trapping laser beams are turned off. To prevent an unwanted residual magnetic field from the anti-Helmholtz coil, it is turned off 12 ms before the excitation pulse. A small ($\approx$300 mG) 8-ms-long homogeneous magnetic field beginning 3 ms before the optical pumping and excitation pulses is applied along their propagation direction. To eliminate the ambient magnetic field at the trap during the trapping period, we used three pairs of Helmholtz coils with active cancelling circuits. Applying a 6-μs-long optical pumping pulse with circular polarization (σ^+) pumps atoms into a single magnetic ground substate ($m_F = 3$). Several micro-second later, a 40-ns-long excitation pulse whose pulse area is close to π is applied with the same circular polarization as that of the optical pumping pulse. These two optical pulses are realized by passing the light from a CW Ti:sappire laser through a pair of acoustic optic modulators (AOMs). The two pulses are tuned to the $[5S_{1/2}\,F = 3]$–$[5P_{3/2}\,F' = 4]$ transition.

The basic experimental apparatus is shown in Fig. 1. The FID signal is obtained with a heterodyne method. In this

*Present address: Thermawave Korea Branch Co., Suwon 442-070, Korea.
†E-mail address: kimjt@chosun.ac.kr

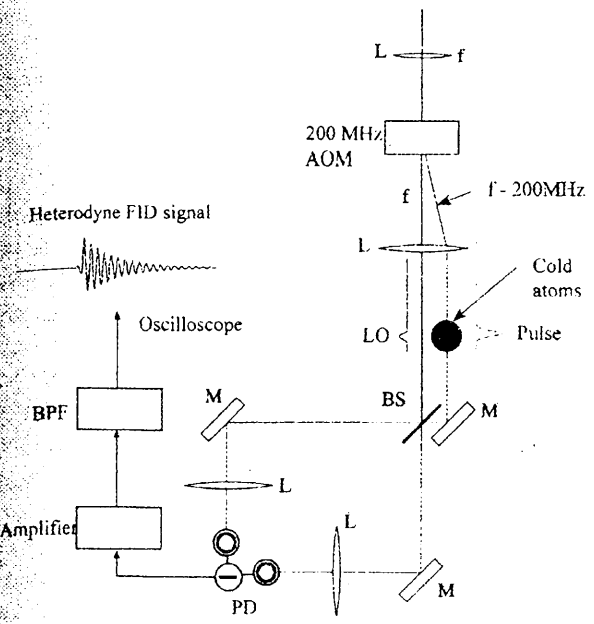

Fig. 1. Experimental setup: LO = optical local oscillator, M = mirror, BS = beam splitter, PD = high-speed photodiode, AOM = acousto-optic modulator, L = lens, f = frequency of CW laser, BPF = band-pass filter.

technique, the CW local oscillator from the undiffracted beam passing through the AOM is mixed with the small FID signal on a photodiode. To increase the signal-to-noise ratio, we used a balanced detection scheme with two high-speed photodiodes (risetime is smaller than 1 ns) and a beam splitter. The spatial diameter of electric field amplitude is $\approx$2 mm. Because the diameter of the trapped atom cloud, monitored by a calibrated charge-coupled device (CCD) camera, is less than 0.4 mm, most of the atoms are under the same electric field. Care was taken to ensure that the center portion of the beam hit the trapped atoms with a pinhole for correct alignment. The optical depth was measured by scanning a weak probe beam slowly through the resonance and measuring the absorption. The sequence of the experiment shown in Fig. 2 is the same as that of FID except for a very long (4 ms) and weak excitation pulse whose frequency is scanned slowly with ramping. The heterodyne FID signal is amplified by two AC amplifiers and filtered by 150–300 MHz band-pass filters. Transient and absorption spectra were acquired using a Tektronix digitizing oscilloscope with 2 G sample/s.

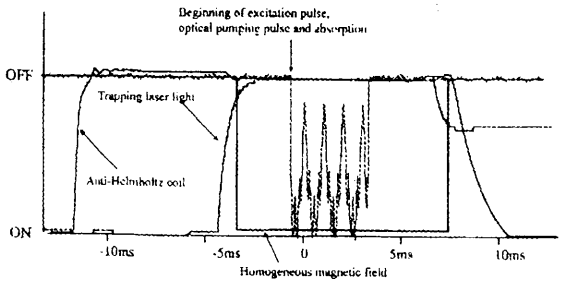

Fig. 2. Experimental sequences.

3. Theory for the Shortening of FID

When a two-level atom, which has upper level $|a\rangle$ and lower level $|b\rangle$, interacts with a monochromatic field $\boldsymbol{E}$, the Hamiltonian and the field are given by

$$H = E_a|a\rangle\langle a| - E_b|b\rangle\langle b| - \mu_{ab}\cdot\boldsymbol{E}, \tag{1}$$

$$\mu_{ab} = \mu(\rho_{ab} + \rho_{ba})\epsilon, \tag{2}$$

$$\boldsymbol{E} = (E^+ + E^-)\epsilon, \tag{3}$$

where E_a and E_b are the upper and lower level energies. μ_{ab} is the quantum-mechanical average value of the oscillating dipole moment and μ is the transition dipole matrix element. We have assumed that the field and dipole have the same spatial orientation ϵ. The field E^+ can be written as

$$E^+(x,t) = \frac{1}{2}|\varepsilon_o|e^{-i(\omega t - kx - \phi(x,t))}, \tag{4}$$

where the field E^- can be described as $E^- = (E^+)^*$, ε is complex amplitude and $\phi(x,t)$ is the slowly varying phase of the electric field. Based on the fact that the time evolution of the density matrix ρ obeys the Heisenberg equation, three equations known as optical Bloch equations can be obtained using rotating-wave approximation and defining $W \equiv \rho_{aa} - \rho_{bb}$, $\rho_{ab} \equiv \tilde{\rho}_{ab}e^{i(kx-\omega t+\phi)}$ and $\tilde{\rho}_{ab} \equiv (U - iV)/2$:

$$\begin{aligned}\dot{U} &= -(\Delta + \dot{\phi})V - \frac{U}{T_2},\\ \dot{V} &= (\Delta + \dot{\phi})U + \Omega W - \frac{V}{T_2},\\ \dot{W} &= -\frac{(W+1)}{T_1} - \Omega V,\end{aligned} \tag{5}$$

where the Rabi frequency $\Omega = \mu\varepsilon_o/\hbar$.

Consider a collection of dipoles with uniform density through a small region. This assumption is normally achieved by a cold sample in MOT having more than 4×10^4 atoms.[19] The atomic polarization $\boldsymbol{P}(x,t)$ of the homogeneous sample can be written as

$$\begin{aligned}\boldsymbol{P}(x,t) &= N\mu_{ab}\epsilon = 2\mu\epsilon\mathrm{Re}\left[\tilde{\rho}_{ab}e^{i(kx-\omega t+\phi)}\right]\\ &= N\mu\,\mathrm{Re}(U(x,t) - iV(x,t))\epsilon e^{i(kx-\omega t+\phi)}.\end{aligned} \tag{6}$$

The electric field amplitude $\boldsymbol{E}$ is related to the macroscopic polarization $\boldsymbol{P}$ by Maxwell's wave equation:

$$\left(\frac{\partial^2}{\partial x^2} - \frac{\partial^2}{c^2\partial t^2}\right)\boldsymbol{E}(x,t) = \frac{4\pi}{c^2}\frac{\partial^2\boldsymbol{P}(x,t)}{\partial t^2}. \tag{7}$$

In general, the microscopic electric field in eq. (5) is not the same as the macroscopic field in eq. (7), which contains all local fields for a dense sample.[20] In our case, the density ($\approx10^{10}\,\mathrm{cm}^{-3}$) is sufficiently low to neglect the difference between them.

This wave equation can be simplified even further by the slowly varying envelope approximation, which means the electric field, polarization amplitudes and phase vary slowly in time compared to the optical frequency and slowly in space compared to an optical wavelength. We use this approximation to simplify the equation as

$$\left(\frac{\partial}{\partial x} + \frac{\partial}{c\partial t}\right)\varepsilon_o(x,t)e^{i\phi(x,t)} =$$

$$\frac{2\pi N\mu\omega i}{c}(U(x,t)-iV(x,t))e^{i\phi(x,t)}. \tag{8}$$

Resonant pulse propagation can be described by solving eqs. (5) and (7) simultaneously.

According to Crisp's work,[21] small-area pulse approximation linearizes optical Bloch equations (5) for small-area pulse propagation through resonant media in the case that the pulse width is comparable to or shorter than T_2. For an exponential pulse whose area is small, the final shape of the pulse after the medium is obtained as

$$\varepsilon_o(x,t)e^{i\phi(x,t)} = e^{-\frac{(t-x/c)}{T_2}}\eta(t-x/c)J_0\left[2\left(\alpha_o x\left(t-x/c\right)\right)^{1/2}\right], \tag{9}$$

where $\eta(t)$ is the unit step function and J_0 is the zeroth-order Bessel function. The shape of the optical FID of laser-cooled atoms is exponential. Normally, the input pulse area for this experiment is slightly less than π. Contrary to our expectations, the FID signal is large even with that pulse area. Because the pulse width ($\approx$40 ns) is larger than the excited state lifetime (27.0 ns), spontaneous emission plays an important role in realizing a large signal where the smallest signal is expected for a very short pulse compared to the excited lifetime. Also, the size of the FID heterodyne signal is normally 100 or 1000 times smaller than the initial pulse amplitude in a small αL, which the small-area approximation satisfies. Total FID measured by the photodiode is determined by adding all of the exponential FID signals occurring at every position of the sample with consideration of the fact that the dimensions of the sample are very small ($\lesssim$0.4 mm), and the shape of the total FID can be written as

$$e^{-\frac{t}{T_2}}\eta(t)\int_0^L J_0\left[2(\alpha_o x t)^{1/2}\right]dx. \tag{10}$$

4. Results and Discussion

A heterodyne FID signal and the square of its Fourier transform are shown in Fig. 3. This is a clear observation of optical FID immediately after the initial pulse in the cold two-level atoms. As we expected, the FID in the homogeneous media shows exponential decay modulated with beating frequency. A Lorentzian fit to the square of the Fourier transform of the heterodyne FID gives the beat frequency and the lifetime for FID. The atomic frequency can be determined very precisely with an accuracy of less than 0.5 MHz from the beat frequency and the RF applied to AOM. While the frequency of the local oscillator is fixed, the radiation of FID has the atomic transition frequency. The difference between the beat and AOM frequency provides the exact detuning. Figure 4 clearly shows the changes of beat frequency when the frequency of the local oscillator is varied by changing the frequency of the CW incoming beam while the AOM frequency is fixed. This confirms that FID radiates with the atomic frequency. As the optical length increases, the lifetime of FID decreases. This shortening of FID is quite different from sudden termination of FID causing the bandwidth of partially excited atoms in an inhomogeneous sample.[11,12] This is related to radiation damping,[22,23] which occurs in an optically thick sample.

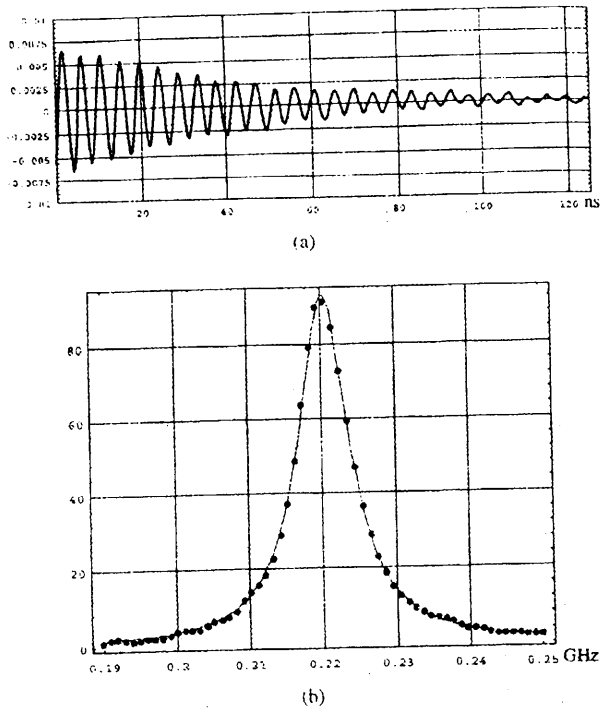

Fig. 3. (a) Heterodyne signal of FID and (b) square of its Fourier transform. From the Lorenztian fit, the center frequency is 220.3 MHz and the decay time is 37.9 ns.

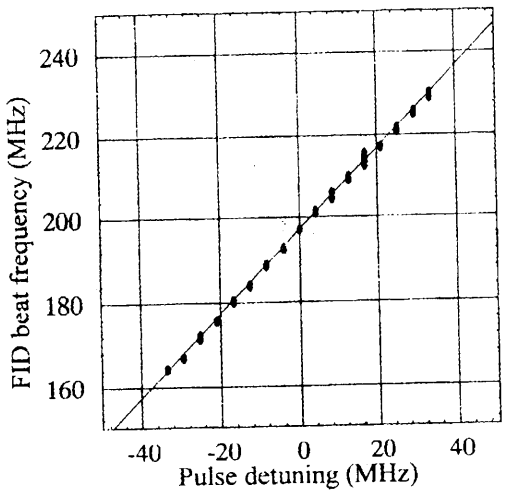

Fig. 4. Beating frequency of the heterodyne FID signal as a function of frequency of the CW laser before AOM while RF of AOM is fixed.

The optical thickness was obtained by dividing an absorption spectra with trapped atoms by one without atoms and taking the logarithm as

$$\log[I/I_0] = -\alpha L. \tag{11}$$

From the modified absorption spectrum, we can determine the value of $\alpha(0)L$, where $\alpha(0)$ is the highest absorption coefficient at atomic resonant frequency. The weak probe (much smaller than the saturation intensity) is scanned $\approx$84 MHz around the resonance. The trap size was controlled by adjusting the size of the repumping laser beam. After changing the trap size, we wait to ensure that the

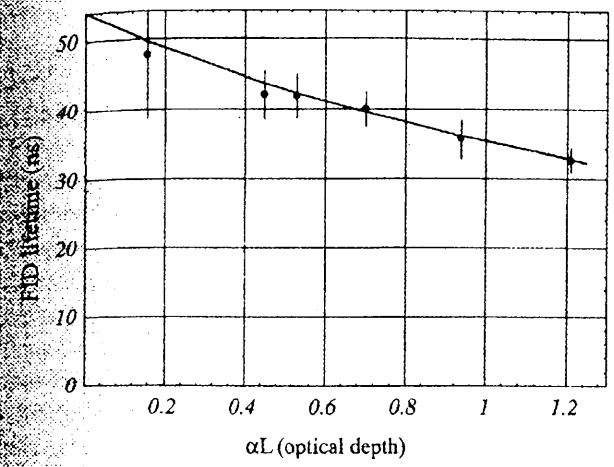

Fig. 5. Decay time of FID as a function of $\alpha(0)L$.

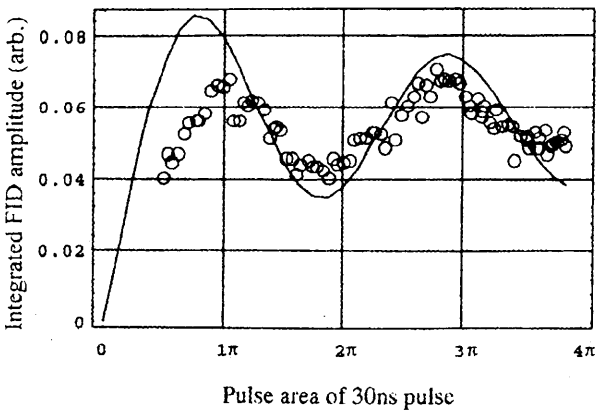

Fig. 6. Integrated FID amplitude shows oscillations as the pulse area increases. Circles show experimental results and the solid line is the simulated curve.

trap is stabilized by monitoring the absorption. Absorption spectra are recorded before and after the FID experiments. More than 10 FID signals are taken at each $\alpha(0)L$. The error bar of Fig. 5 is the standard deviation of the mean value. The lowest $\alpha(0)L$ shows a large error bar because the FID signal is very small. The solid line is the simulated lifetime of the FID obtained from the exponential fitting of the generated waveform by equation (10). Fairly good agreement with data is shown.

We observed that FID decays faster than T_2 even in the homogenous two-level system and depends on the optical thickness of the trap. This kind of shortenings which has been observed even in small $\alpha(0)L$ (less than 1), can be explained by the propagation effect of the small FID exponential envelope through the media.

5. Rabi Oscillation of FID

Consider a two-level system with no relaxation interacting with a resonant electric field. The optical Bloch equation becomes one simple vector equation,

$$\frac{d\boldsymbol{M}}{dt} = \boldsymbol{M} \times \Omega, \tag{12}$$

where $\Omega = (k\varepsilon_o, 0, \Delta)$ and $\boldsymbol{M} = (U, V, W)$ is called a Bloch vector. This equation shows that the Bloch vector $\boldsymbol{M}$ precesses about the effective field Ω. On resonance frequency, because Ω is in the direction of the u axis, $\boldsymbol{M}$ precesses in the W–V plane. W (associated with the population inversion) and V (associated with the atomic polarization) are expected to oscillate at the Rabi frequency.

In a homogeneous sample, the FID amplitude is simply proportional to V. With the laser-cooled ^{85}Rb atoms, the $[5S_{1/2}\,F = 3\,(m_F = 3)]$–$[5P_{3/2}\,F' = 4\,(m'_F = 4)]$ transition makes an ideal two-level system. We have used a heterodyne and homodyne detection scheme to obtain the FID amplitude. Because pulses are created by AOM with CW Ti-sappire laser, a 40-ns-long pulse, larger than the excited state lifetime (27.0 ns), is used to observe Rabi oscillations. In this case, spontaneous emission plays a significant role. If the pulse is short and sufficiently strong to observe the oscillations, the maxima and minima of Rabi oscillation can be estimated using a simple Bloch scheme. Maxima are expected for $\theta = \pi/2,\ 3\pi/2,\ 5\pi/2, \cdots$ and the minima for $\theta = \pi,\ 2\pi,\ 3\pi, \cdots$. However, the positions of the maxima and minima are expected at a larger pulse area, due to spontaneous emission, when the width of the incoming pulse (τ) increases.

Simulation has been carried out by numerically integrating the optical Bloch equations with longitudinal (T_1) and transverse (T_2) relaxation time and a Gaussian excitation pulse. Because optically thin samples ($\alpha(0)L \approx 0.5$) are used, the effect of optical thickness is taken into account in this simulation only by reducing T_1 and T_2, which are obtained from the FID signal. The homodyne signal is obtained by mixing the heterodyne FID with a reference (220 MHz) from the RF oscillator in a quadrature demodulator. The integrated amplitude of homodyne FID is measured as a function of the pulse area θ. Figure 6 shows oscillations of the integrated FID and simulated curve. Slight disagreement between experimental and simulated result is believed to be caused by simple treatment of the effect of dense media in the simulation.

6. Conclusions

In conclusion, we have observed optical free induction decay (FID) in homogeneously broaden ^{85}Rb atoms, which is difficult to observe in fast decaying inhomogeneously broadened samples. We found that the FID lifetime is sensitively dependent on the optical thickness of the sample and the shortening of the FID lifetime is explained by anomalous FID absorption in resonant media. FID amplitude is expected to show Rabi frequency oscillation with increasing excitation pulse area. We can not get clear Rabi oscillation due to excitation pulse width comparable to T_1. Experimental oscillation of the integrated FID signal is found to be fairly consistent with the simulation result by taking spontanous emission into account.

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