# Optical free-induction decay of laser-cooled $^{85}$Rb

K. Toyoda, Y. Takahashi, K. Ishikawa, and T. Yabuzaki
*Department of Physics, Faculty of Science, Kyoto University, Kyoto 606-01, Japan*
(Received 2 December 1996; revised manuscript received 28 April 1997)

Optical free-induction decay (FID) of laser-cooled $^{85}$Rb has been observed. Considerably good agreements are obtained between experiments and theoretical calculations in the condition of no inhomogeneous broadening. The possibility to apply optical FID measurements to the studies of phase relaxation in a dense atom trap is discussed. [S1050-2947(97)02808-4]

PACS number(s): 42.50.Md, 32.80.Pj

## I. INTRODUCTION

Most optical coherent transient experiments so far have been performed with samples which are subject to considerable inhomogeneous broadening [1]. Obtained signals are contributed from atoms with various resonance frequencies, and the behaviors of each homogeneous system are averaged. Coherent transient signals of a homogeneous sample [2] are, on the other hand, directly related to the behavior of the homogeneous system with great sensitivity, and this feature is advantageous for many purposes, including the study of optical dipole noise due to fluctuations of induced dipoles [3]. Also it is of great interest to study pulse propagation in a purely homogeneous sample. Since the area theorem [4] which describes the evolution of the pulse area is based on the assumption that the inhomogeneous width is larger than the natural width, the behavior of pulse propagation in a purely homogeneous sample [5] becomes different from that expected from the area theorem for an inhomogeneously broadened sample.

Recently, laser cooling and trapping technique has rapidly developed [6], and it has enabled us to prepare a sample of atoms which features both a negligibly small inhomogeneous width and a high density at the same time. These features are quite useful for optical coherent transient experiments, as well as many other applications. In this paper we report on the observation of optical free-induction decay (FID) signals of laser-cooled $^{85}$Rb atoms. The details of the experiment, as well as the theoretical explanations, are described. The possibility to apply optical FID measurements to the studies of phase relaxation in a dense atom trap is also discussed.

## II. BASIC EQUATIONS

We briefly review the basic equations which describe the interaction between an optical pulse and trapped atoms. We consider a trapped atom as a two-level system $|b\rangle,|a\rangle$ with energy eigenvalues of $\hbar\omega_0,0$, respectively. We assume that there is no inhomogeneous broadening, and that the atom density $N$ is homogeneous in space. In addition, spatial inhomogeneity of the excitation pulse is neglected.

The electric field $\mathbf{E}(z,t)$ of an optical pulse which propagates in the $z$ direction with an angular frequency $\omega$ are expressed as

$$\mathbf{E}(z,t)=2E_0(z,t)\mathrm{Re}\{\mathbf{e}e^{i[\omega t-kz+\phi(z,t)]}\}, \tag{1}$$

where $\mathbf{e}$ is a unit vector which expresses the polarization of the electric field, $\phi(z,t)$ is the additional phase, $k=\omega/c$ ($c$ is the velocity of light in vacuum). $E_0(z,t)$ is a slowly varying envelope of the field amplitude and is taken to be real.

A behavior of the two-level atom is described by the optical Bloch equation [7] as follows:

$$\dot{u}=-(\Delta\omega-\dot{\phi})v-\frac{u}{T_2'}, \tag{2a}$$

$$\dot{v}=(\Delta\omega-\dot{\phi})u+\kappa E_0 w-\frac{v}{T_2'}, \tag{2b}$$

$$\dot{w}=-\kappa E_0 v-\frac{w-(-1)}{T_1}, \tag{2c}$$

where $(u(z,t),v(z,t),w(z,t))$ is the Bloch vector, $\Delta\omega=\omega_0-\omega$ and $\kappa=2d/\hbar$ ($d=\mathbf{d}_{ab}\cdot\mathbf{e}$, where $\mathbf{d}_{ab}$ is the dipole matrix element between $|b\rangle$ and $|a\rangle$; $d$ is taken to be real by choosing an appropriate relative phase between the two states). $T_1$ is a radiative lifetime and $T_2'$ is a homogeneous phase relaxation time ($T_1=26.5$ ns for the $5\,^2S_{1/2}$–$5\,^2P_{3/2}$ transition of $^{85}$Rb [8]). When there is no additional phase relaxation $T_2'=2T_1$.

The Maxwell equation for the electric field is given as

$$\left(\frac{\partial^2}{\partial z^2}-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\mathbf{E}=\frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{P}, \tag{3}$$

where $\mathbf{P}(z,t)=Nd\mathrm{Re}(\mathbf{e}[u(z,t)+iv(z,t)]\exp\{i[\omega t-kz+\phi(z,t)]\})$ is the polarization density. Applying a slowly varying envelope approximation to leave only the lowest-order derivatives, two equations for the components in phase and out of phase with the electric field are obtained

$$\kappa E_0\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)\phi=-\alpha' u, \tag{4a}$$

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)\kappa E_0=\alpha' v, \tag{4b}$$

where $\alpha'=2\pi N\omega d^2/\hbar c$. The absorption coefficient $\alpha$ is given as $\alpha=2\alpha' T_2'$. $\alpha L$ is equal to the sample optical density times ln10, where $L$ is the sample length. The whole

 

spatial and temporal evolution of an optical pulse can be obtained by solving Eqs. (2) and (4).

### III. EXPERIMENTAL SETUP

We trapped and cooled $^{85}$Rb atoms in a glass vapor cell by using the magneto-optical trap (MOT) method [9]. The background vapor pressure was about $1\times10^{-8}$ Torr and was dominated by the partial pressure of Rb. A quadrupole magnetic field for MOT was produced by an anti-Helmholz coil and its gradient at the trap position was 6 G/cm for the axial direction and 3 G/cm for the radial direction. Six beams for MOT (circularly polarized, intensity 18 mW/cm$^2$ per beam, linewidth$\leqslant$400 kHz) were applied to the vapor cell, frequency of which was detuned by 10 MHz below resonance of the $F=3$–$F'=4$ transition of $D_2$ line ($5\,^2S_{1/2}$–$5\,^2P_{3/2}$, 780 nm). To avoid optical pumping to the lower hyperfine state of the ground state ($F=2$), an additional beam (10–100 mW/cm$^2$) which was resonant to the $F=2$–$F'=3$ transition was applied. The diameter of the trapped atom cloud was typically 1.5 mm, which was determined from an image of a charge-coupled device array. The atom density was estimated from absorption of a weak probe beam and was typically $5\times10^{10}$ cm$^{-3}$. The typical number of the trapped atoms has been calculated from above values to be $2\times10^8$. $\alpha L$ could be varied up to about 10 by changing the shape or size of the trapped atom cloud, or the atom density. The temperature of the trapped atoms was approximately 200 $\mu$K, which was measured by the release and recapture method [10]. The inhomogeneous Doppler width of the $D_2$ line for this temperature is 360 kHz, which is almost negligible compared with the 6.0 MHz natural width.

Excitation pulses for optical FID (100ns–1 $\mu$s duration) were produced by an electro-optic modulator (EOM) (rise time 5 ns). In some cases, where phase rotation should be avoided, an acousto-optic modulator (AOM) (rise time 15 ns) was used instead. Since the observed transition must be a closed two-level system, we performed optical pumping of all atoms into the $F=3$, $m_F=+3$ state with a circularly polarized optical pulse resonant to the $F=3$–$F'=3$ transition (30 mW/cm$^2$, 10 $\mu$s duration) before the observation of optical FID. The completeness of optical pumping could be verified through the disappearance of fluorescence from the trapped atoms. In order to avoid the perturbation from the trapping beams, we turned them off during the optical FID measurement. The pulses transmitted through the trapped atoms were detected by an avalanche photodiode (bandwidth 1 GHz), and monitored with a digital storage oscilloscope (bandwidth 300 MHz).

### IV. EXPERIMENTAL RESULTS AND THEORETICAL ANALYSES

Figure 1 shows a typical transmitted pulse. The frequency of the excitation pulse was just resonant and its intensity was much weaker than the saturation intensity. This optical FID signal of laser cooled atoms is analogous to superradiance [11] in that a large fraction of absorbed energy is emitted as coherent radiation. At the very beginning of the transmitted pulse ($t=0$–10 ns) the absorption is relatively small. At this period $v$ which reduces $\kappa E_0$ through Eq. (4b) is not induced considerably. On the contrary, as the time passes, $v$ comes to take a large negative value driven by the second term of Eq. (2b) with $\kappa E_0>0$ and $w\sim-1$. This large $v$ reduces $\kappa E_0$ through Eq. (4b), resulting in a sizable absorption. At $t\sim70$ ns, $v$ is subject to relaxation from the third term of Eq. (2b) to become a constant value, which results in a constant absorption. When the incident excitation pulse amplitude falls to zero ($t\sim80$ ns), the contribution from the incident excitation pulse suddenly disappears and only the contribution from $v$ ($v<0$) remains [Eq. (4b)]. $\kappa E_0$, which has been a positive value up to this time, suddenly crosses zero and becomes a large negative value. Finally $v$ decays to zero with the time constant $T_2'$ according to the Eq. (2b).

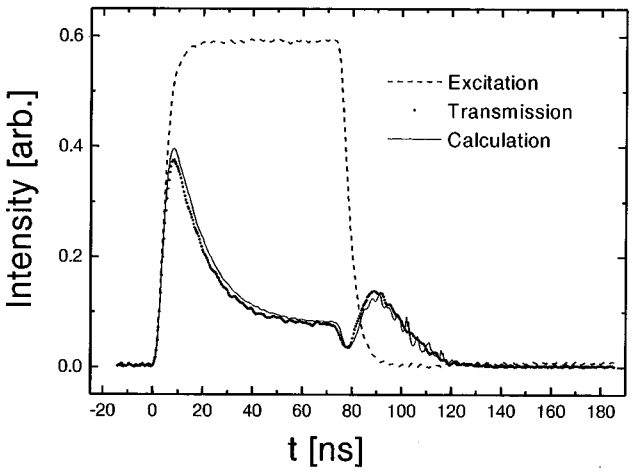


FIG. 1. A transmitted pulse is shown as dots. The frequency is just resonant and the intensity is sufficiently weak compared with the saturation intensity. $\alpha L$ is approximately 5. The decay from $t\sim90$ ns is an optical FID signal. The dashed line is the excitation pulse without trapped atoms. The solid line is a theoretical result calculated in the condition where $T_2'=2T_1=53$ ns and $\alpha L=4$.

For a quantitative study of the whole transmitted pulse shape, we compared it with theoretical calculation with Eqs. (2) and (4). The detail of calculation is as follows. The sample was considered as an assembly of thin layers sliced perpendicularly to the propagation direction. A real experimental incident pulse was used as the boundary condition for the electric field at the first layer, and it was assumed that initially all atoms were in the ground state ($u,v=0$ and $w=-1$). At first, the time evolution of $(u,v,w)$ in the first layer was calculated with Eqs. (2). By using this $(u,v,w)$ the time evolution of the electric field at the next layer was calculated with Eqs. (4a) and (4b). The same procedure was repeated for all layers along the propagation direction, and finally the transmitted pulse shape was obtained. In this calculation, back scattering of the excitation pulse by the atoms was neglected, so that such a successive calculation with sliced layers became possible. Thus calculated transmitted pulse shapes were compared with the experimental transmitted pulse shape with various values of parameters of $T_2'$ and $\alpha L$. The solid lines in Fig. 1 shows the intensity of a calculated transmitted pulse in the case of $T_2'=2T_1=53$ ns and $\alpha L=4$. In order to incorporate the imperfect matching between the excitation beam cross section and the atom density distribution, 10% of the intensity of the excitation pulse was assumed to be transmitted without interacting with the

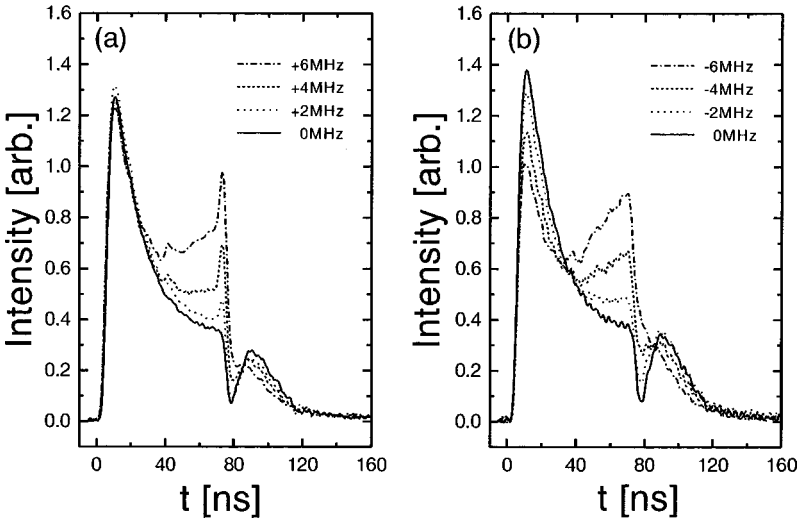


FIG. 2. Transmitted pulses for (a) red-detuned ($\Delta\omega>0$) or (b) blue-detuned ($\Delta\omega<0$) excitation pulses. The numbers in the figure represents the value of $\Delta\omega$. $\alpha L$ is considered to be smaller than that in the case of Fig. 1. As for the case of red detuning, steep peaks are observed at the end of the excitation pulse.

atoms. An excellent agreement with experimental results was achieved for the calculated pulse shape obtained in such a way. This complete coincidence implies that the effect of additional phase relaxation, due to cold collisions for example, is not significant at the present atom density of $5\times10^{10}$ cm$^{-3}$.

Figure 2 shows transmitted pulses for detuned excitation. As the detuning of the excitation pulse becomes larger, the magnitude of the induced $v$ becomes smaller, which leads to smaller absorption and, hence, a smaller optical FID signal. For red-detuned excitation pulses ($\Delta\omega>0$), steep peaks can be seen near the end of the excitation pulses. The appearance of such steep peaks are explained by taking account of the phase rotation of the excitation pulse, as follows. When we produce optical pulses by using an EOM, it is difficult to avoid the rapid rotation of the optical phase during the rising and falling period of the pulse envelope. As for the EOM which we have used, the phase $\phi(z,t)$ of the pulse takes nonzero values during the rising and falling period [$\phi(z,t)>0$ and $\phi(z,t)<0$, respectively]. Now we consider the weak-excitation case ($u,v\sim0$ and $w\sim-1$). Just before the steep peak appears, the system has reached the steady state, and $v$ takes a negative value. Since $\Delta\omega>0$, $u$ takes a positive value through Eq. (2a). During the falling period of the excitation pulse, a sudden phase shift $\Delta\phi(z,t)<0$ occurs, which leads to a sudden decrease in $|v|$ through Eq. (2b) [$\Delta v=-\Delta\phi(z,t)u>0$]. This decrease in $|v|$ leads to an increase in the pulse envelope through Eq. (4b), resulting in a steep peak in the transmitted signal. In a similar way, in the case of $\Delta\omega<0$ the phase shifts result in a decrease in the pulse envelope, although this decrease cannot be well distinguished from the sudden decrease of the original excitation pulse envelope. In the case of $\Delta\omega=0$, $u$ almost always equals zero and such a steep structure will not appear. This type of steep structure might average out and not become apparent in the case of Doppler broadened samples. The appearance of such steep peaks is a notable consequence of the great sensitivity of coherent transient signals of laser cooled atoms to the phase of the electric field.

We have also performed numerical calculations for detuned excitation. The EOM which we have used has two crystals in series, in order to compensate a temperature dependence of refraction index. A slight time difference between the applied voltages for two crystals results in a sudden phase shift. Figure 3 shows calculated transmitted pulses incorporating such phase shifts, where the time difference is assumed to be 2 ns. The characteristic features of the experimental pulse in Fig. 1, such as the overall shape and the appearance of steep peaks in the red-detuned cases, are explained with this calculation.

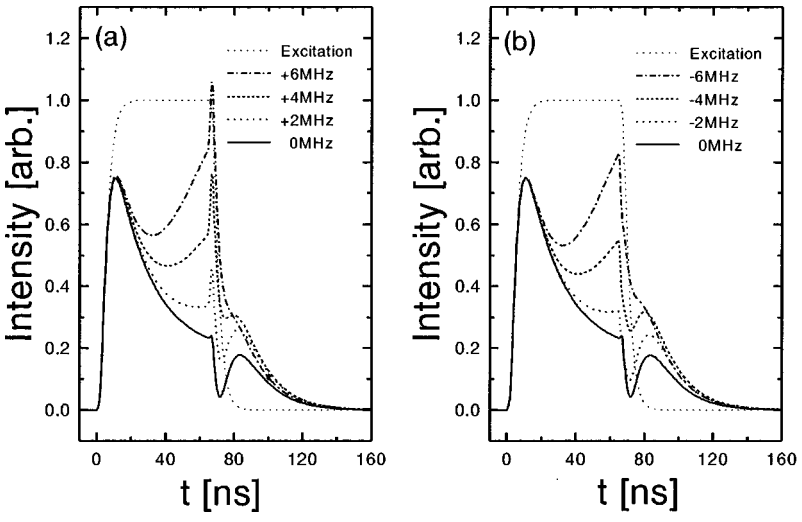


FIG. 3. Calculated transmitted pulses for (a) red-detuned or (b) blue-detuned excitation pulses, in the case where the excitation pulse is sufficiently weak compared with the saturation intensity, and $\alpha L=2.5$.

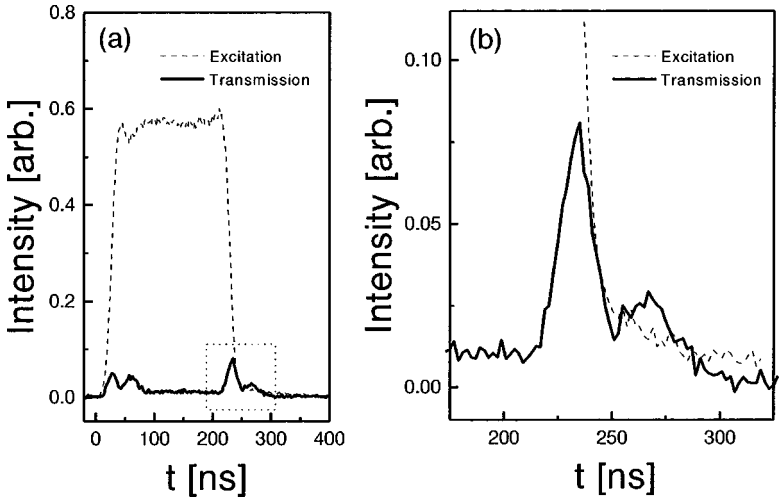


FIG. 4. (a) A transmitted pulse in the case where $\alpha L$ is approximately 9. The excitation pulse is weak enough compared with the saturation intensity. A splitting of optical FID into two parts can be seen. (b) A magnified scale is used to examine the detailed feature in the dashed region in (a).

In the case of nearly the highest $\alpha L$ in our experiment ($\sim 9$) and sufficiently weak excitation compared with the saturation intensity, a novel feature that an optical FID splits into two parts has been observed (Fig. 4). In order to avoid the ambiguity associated with the phase rotation of the excitation pulse, an AOM, instead of an EOM, was used for producing the excitation pulse. Also, we checked that the second peak did not come from an electrical mismatch by inserting an attenuator in the signal path. Such a splitting of the optical FID, or in other words an appearance of double peaks, is considered to correspond with ''anomalous classical absorption'' [7,12]. In the classical weak-excitation regime, the pulse area follows Beer's law, and it approaches zero as $\alpha L$ becomes fairly large, while the total energy of the pulse needs not. In fact, an excitation pulse with a duration much shorter than $T_2'$ [13] can be transmitted through a sample with a fairly large $\alpha L$ in the shape of a damped oscillation. In our experimental result, this type of oscillation is considered to have resulted in the double peaks. Figure 5 shows a theoretical result in the case of $\alpha L = 9$, which shows a similar type of splitting as in Fig. 4.

## V. DISCUSSION AND CONCLUSION

Since optical FID signals of laser cooled atoms are sensitive to optical coherence, it is expected that phase relaxation in a laser trap can be investigated by observing optical FID signals. We will consider here two types of broadening processes which may contribute to phase relaxation, one of which is a ''static'' type and the other a ''dynamic'' type.

At first, let us consider the possibility of observing the effect of a ''static'' broadening which comes from the interatomic distance distribution. A pair of identical ground-state and excited-state atoms is subject to a line shift due to the $C/R^3$ potential [14], so that a variety of interatomic distance results in inhomogeneous broadening, which leads to dephasing among induced dipoles. The atom density at which the resonance frequency shift due to $C/R^3$ potential ($C = 10.1$ in atomic units for $^{85}$Rb [15]) becomes equal to the natural width is calculated as about $n = 6 \times 10^{14}$ cm$^{-3}$. In such a high atom density the line shift can be distinguished from the natural width. Such a high density can be obtained through the evaporative cooling method [16].

We also argue the possibility of studying a ''dynamic'' broadening process due to interatomic collisions by using optical FID measurements. It appears unlikely that ordinary elastic collisions lead to any significant phase relaxation for the typical atom density of a usual magneto-optical trap ($1 \times 10^{10}$–$1 \times 10^{11}$ cm$^{-3}$), since the expected elastic collision interval for the above condition (typically $\sim 1$ s) [17] is by far longer than the duration of an optical FID signal

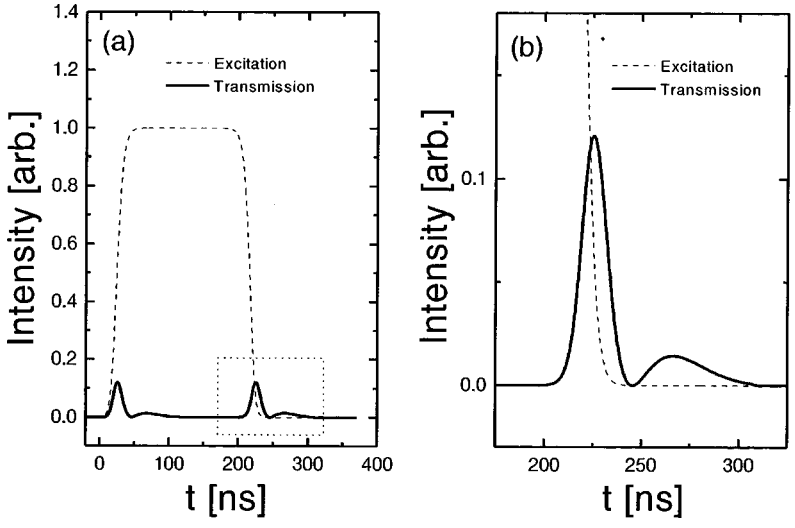


FIG. 5. (a) A calculated transmitted pulse in the case where the excitation pulse is sufficiently weak compared with the saturation intensity, and $\alpha L = 9$. (b) The dashed region in (a) with a magnified scale.

($\sim$50 ns). However, if there is any kind of enhancement effect on the collision rate due to acceleration of the interatomic motion by the $C/R^3$ interaction potential, as is the case in the trap loss mechanism in a laser trap [18], collisional phase relaxation effects may be observed in optical FID signals. While we used a simple semi-classical picture, it no longer gives a sufficient description for this type of argument, and fully quantum-mechanical analyses will be required.

In conclusion, we have observed optical FID signals of laser cooled $^{85}$Rb atoms. The observed signals have been compared with theoretical calculations in the case of no pure phase relaxation ($T_2'=2T_1=53$ ns), and fairly good agreements have been achieved. In the case of slightly red-detuned excitation, we have observed a steep peak near the end of the excitation pulse, which is a consequence of the great sensitivity of optical FID signals of laser cooled atoms to the phase of the excitation pulse. We have also observed a splitting of an optical FID signal into two parts in the case where the excitation pulse is sufficiently weak compared with the saturation intensity and $\alpha L$ is large ($\sim 9$). The possibility to apply optical FID measurements to detect phase relaxation effects in a dense atom trap has been discussed, and it has been predicted that in such a high atom density as more than $6\times10^{14}$ cm$^{-3}$ ''static'' phase relaxation effects due to line shift from the interatomic interaction potential can be observed.

## ACKNOWLEDGMENTS

We would like to thank M. Fujimoto and T. Isayama for their experimental assistance. This work was partially supported by the Grant-in-Aid for Scientific Research. One of the authors (K.T.) acknowledges financial support from the Japan Society for the Promotion of Science for Young Scientists.

[1] As for coherent transient experiments with solid samples, see, for example, R. M. Macfarlane and R. M. Shelby, in *Spectroscopy of Solids Containing Rare Earth Ions*, edited by A. A. Kaplyanskii and R. M. Macfarlane (North-Holland, Amsterdam, 1987), p. 51. As for experiments with gas samples, see, for example, R. E. Slusher and H. M. Gibbs, Phys. Rev. A **5**, 1634 (1972).

[2] In the work of Golub, Bai, and Mossberg, an atomic beam sample with a reduced Doppler width is used, although it is still about ten times as large as the natural width J. E. Golub, Y. S. Bai, and T. W. Mossberg, Phys. Rev. A **37**, 119 (1988).

[3] A. M. Bacon, H. Z. Zhao, L. J. Wang, and J. E. Thomas, Phys. Rev. Lett. **75**, 1296 (1995).

[4] S. L. McCall and E. L. Hahn, Phys. Rev. Lett. **18**, 908 (1967); S. L. McCall and E. L. Hahn, Phys. Rev. **183**, 457 (1969).

[5] G. L. Lamb, Jr., Phys. Lett. **25A**, 181 (1967); Rev. Mod. Phys. **43**, 99 (1971).

[6] See special issue on laser cooling and trapping, edited by S. Chu and C. Wieman [J. Opt. Soc. Am. B **6**, No. 11 (1989)].

[7] L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975).

[8] A. A. Radzig and B. M. Smirnov, *Reference Data on Atoms, Molecules, and Ions* (Springer-Verlag, Berlin, 1985).

[9] E. L. Raab, M. Prentiss, A. Cable, S. Chu, and D. E. Pritchard, Phys. Rev. Lett. **59**, 2631 (1987).

[10] P. L. Gould and H. J. Metcalf, Phys. Rev. Lett. **61**, 169 (1988).

[11] R. H. Dicke, Phys. Rev. **93**, 99 (1954).

[12] M. D. Crisp, Phys. Rev. A **1**, 1604 (1970); O. Kinrot and Y. Prior, *ibid.* **51**, 4996 (1995).

[13] Although this condition is not fulfilled for our experimental pulse, its spectral width at the rising or falling period temporarily becomes large compared with the inverse of $T_2'$.

[14] G. W. King and J. H. Van Vleck, Phys. Rev. **55**, 1165 (1939).

[15] P. S. Julienne and J. Vigue, Phys. Rev. A **44**, 4464 (1991).

[16] H. F. Hess, Phys. Rev. B **34**, 3476 (1986); H. F. Hess *et al.*, Phys. Rev. Lett. **59**, 672 (1987).

[17] Here, for simplicity it is assumed that the cross section for elastic collisions does not change even when one of the atoms is in the excited state. This assumption, oversimplified as it would be in some cases, might be appropriate for the first estimation of the approximate order of the collision interval.

[18] A. Gallagher and D. Pritchard, Phys. Rev. Lett. **63**, 957 (1989).