Measurement of spin-spin relaxation of 0.05-wt.% ruby

Takasi Endo and Tsuneo Hashi
Department of Physics, Faculty of Science, Kyoto University, Kyoto 606, Japan

Takao Muramoto
Faculty of Education, Shiga University, Otsu 520, Japan
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The intraline electron spin-spin relaxation in the ground state $^4A_2$ in 0.05-wt.% ruby was measured with a time resolution higher than that reported previously. A much faster decay of 20–30 μs ($1/e$ decay time) was found, which is to be assigned to the spin-spin relaxation.

In a previous paper,[1] hereafter referred to as paper I, we reported an optical labeling technique which enables us to measure an intraline electron spin-spin relaxation (SSR). A $\sim$500-μs decay of an optical hole (center hole[1]) was found for 0.05-wt.% ruby at 0.35 kOe and we interpreted it to be due to SSR. The time resolution was 50 μs.

Szabo and Heber measured photon-echo decay times of the R_1 line and found homogeneous line narrowing due to depletion of the population of the ground Zeeman sublevel $(+\frac{1}{2})^4A_2$. They interpreted the line-narrowing effect under the assumption that the homogeneous line broadening of the R_1 line comes from the SSR between $(\pm\frac{1}{2})^4A_2$, and suggested a few μs as the SSR time. Although their experiment was performed at a higher magnetic field ($H=3$ kOe), there is a clear discrepancy between their SSR times and ours.

We improved the time resolution of our method (to about 1 μs) and performed essentially the same measurement as in paper I. The same optical transitions as those in paper I were examined. As a result, we found a much faster decay of $\sim 10\ \mu$s, which is to be assigned to SSR among sublevels of the electron spins in the ground state $^4A_2$. From a careful examination of the temperature dependence it was clarified that the slow decay reported in paper I was due to the spin-lattice relaxation between the excited states $(\pm\frac{1}{2})E(^2E)$. The sample temperature was a little higher than that estimated in paper I, probably because of the heating by the pumping laser light.

In order to avoid optical heating and pumping effects a cw ruby laser was chopped by an acousto-optic modulator as shown in Fig. 1. The Stark switching technique was not used in the present experiment. The time evolution of the optical hole burned by the first light pulse was monitored by measuring the transmission of the second light pulse. The intensity of the transmitted light was detected by a PIN photodiode and accumulated by a boxcar integrator with a gate as shown in Fig. 1(b). The difference of the transmission in the presence and absence of the optical hole was measured by turning the first light pulse on and off. In order to improve the time resolution, the width of the first light pulse was reduced to 1 μs, and the signals were much weaker than those in paper I.

Figure 2 shows the change of the transmission or the decay of the center hole in a time shorter than $\sim 100\ \mu$s at

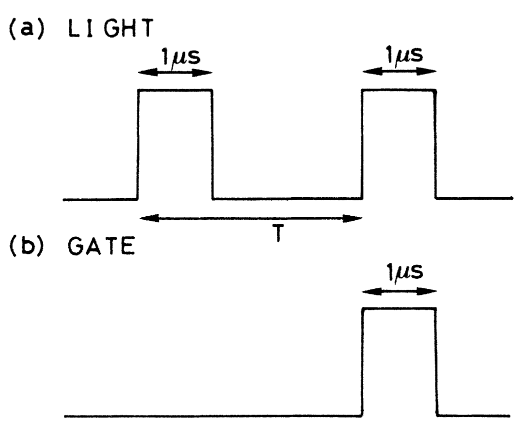

FIG. 1. (a) Light pulse sequence and (b) timing of the gate of the boxcar integrator for the measurement of the decay of optical holes.

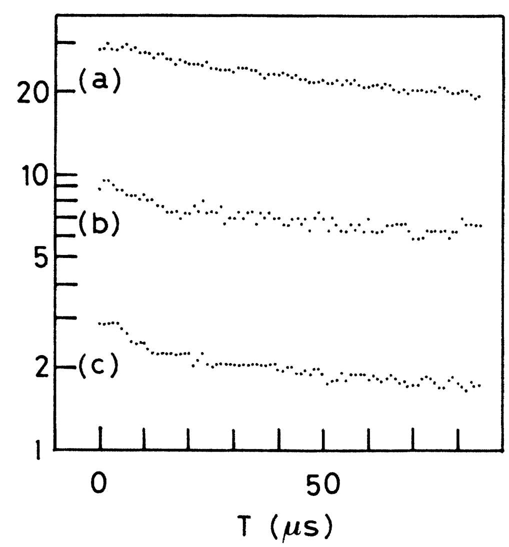

FIG. 2. Decay curves of optical holes. (a) and (b) are those for 0.0034- and 0.05-wt.% ruby at 0.35 kOe, respectively. (c) is that for 0.05-wt.% ruby at 3 kOe.

$H = 0.35$ kOe. In a dilute 0.0034-wt. % ruby [trace (a)] no fast decay was observed. However, in a 0.05-wt. % ruby a fast decay with a $1/e$ decay time of 10 μs can be recognized as shown in trace (b). This fast decay was not found in Fig. 4(a) of paper I because of the poor time resolution. By the rate equation analysis on the assumption that SSR times between sublevels in $^4A_2$ ($m_s \leftrightarrow m_s+1$ where $m_s = -\frac{3}{2}, -\frac{1}{2}, \frac{1}{2}$) are equal, the (average) SSR time ($1/e$ decay) of 20–30 μs is obtained.

The side hole was observed but the time constant of its development was not measured because of the poor signal-to-noise ratio.

We also observed the transmission change [trace (c)] at the same magnetic field ($H = 3$ kOe) as in Ref. 2. The decay time is nearly the same (about 10 μs).

Therefore, the SSR time in the ground state $^4A_2$ of 0.05-wt. % ruby should be 20–30 μs. These values are still much longer than the optical dephasing time (T_2) of 0.45 μs.[2] This fact indicates that the mechanisms of homogeneous broadening cannot be understood fully by the SSR in the ground state.

[1]T. Endo, T. Hashi, and T. Muramoto, Phys. Rev. B 30, 2983 (1984).

[2]A. Szabo and J. Heber, Phys. Rev. A 29, 3452 (1984).