Nuclear magnetic relaxation in the domain and domain wall of pure iron

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The nuclear magnetic relaxation times in the domain and domain wall of pure natural iron have been measured separately. We have witnessed that the spin-spin relaxation time T_2 is longer than the spin-lattice relaxation time T_1 in the domain wall. The ratio T_2/T_1 increases and approaches 2 with decreasing rf pulse power level. This is due to the anisotropy in the fluctuation of the hyperfine field by single magnon processes or wall-type excitations. The measured spin-spin relaxation rate is well described by the sum of the nonsecular term and the dipole-dipole interaction term. [S0163-1829(97)03829-0]

I. INTRODUCTION

Since the first observation of the nuclear magnetic resonance (NMR) in a ferromagnet, investigators have reported various values for both the spin-lattice and spin-spin relaxation times of Fe^{57}. Weger attributed the spread of these values to a mixture of signals coming from the domain and domain wall.[1] He observed a nonexponential fast relaxation at low rf power levels and an exponential slow relaxation at high rf power levels, and suggested that they are the relaxations in the domain wall and the domain, respectively. Later, Stearns showed that the relaxations in the domain wall are nonexponential because both the enhancement factor and relaxation rates depend on the location inside the domain wall, being maximum at its center, due to the variation of the angle between adjacent electron spin directions.[2] In her case, most of the signal came from the domain wall, and therefore, she did not have to worry about the signal from the domain mixing into the total signal. The ratio of the signals coming from the domain and domain wall depends on the sample properties such as the enhancement factors and volumes of the domain and domain wall.

In this work, we clearly divided the total signal of the Fe^{57} NMR experiment in a multidomain state natural iron into the signal from the domain and that from the domain wall. The spin-spin and spin-lattice relaxation rates in the domain and domain wall were measured separately as a function of temperature from 20 K to room temperature. The most interesting result is that the spin-spin relaxation time (T_2) is longer than the spin-lattice relaxation time (T_1) in the domain wall. The ratio of the two relaxation times T_2/T_1 approached 2 in the limit of zero rf power levels. This is due to the anisotropy in the fluctuation of the hyperfine field by single magnon processes or wall-type excitations. The spin-lattice relaxation time in the domain is well explained by the hyperfine interaction with conduction electrons. The spin-spin relaxation rate is shown to be the sum of the nonsecular term and the dipole-dipole interaction term both in the domain and domain wall.

II. EXPERIMENT

The sample was 99% pure natural iron powder with the sizes varying from 1 to 4 μm. The picture taken by a scanning electron microscopy (SEM) shows that the particles are mostly spherical. The sample was annealed to remove the internal strain at 400 °C in the vacuum below 10^{-1} torr for 4 h. The spin-lattice relaxation time was measured by the saturation recovery method, where the amplitude of the echo was measured after a pair of pulses following the first $\Delta\tau$ pulse. The spin-spin relaxation was studied by measuring the amplitude of the echo after the application of a pulse sequence ($\Delta\tau - t - 2\Delta\tau -$). The linewidth obtained by the Fourier transform of the spin echo was 50 kHz. The pulse width $\Delta\tau$ was 1.5 μs which is short enough to excite the whole range of the spectrum.

III. RESULTS AND DISCUSSION

In the NMR of ferromagnets, the rf field experienced by a nuclear spin is enhanced due to the accompanying motion of the magnetization of electron spins,[3,4] and the NMR signal is enhanced by the same factor. The enhancement factor in the domain wall depends on the location,[5,6] and is usually order of magnitude larger than that in the domain. Therefore, the flip angles of nuclear spins are different for a given rf field. When the rf pulse is not strong enough to make the average flip angle of the nuclear spins in the domain walls 90°, the signal coming from the domain wall is dominant. If the rf pulse power is increased higher than the average 90° pulse power for the nuclear spins in the domain wall, the distribution of the flip angles in the domain walls tends to cancel out the resulting echo amplitude, while the signal from the domain keeps increasing until the flip angle of the nuclei in the domain reaches 90°. In this range, therefore, the contribution of the nuclear spins in the domain to the total signal increases with the rf field. Thus, the higher is the rf field then the larger is the contribution of the spins in the domain to the total signal.

Figure 1 shows some typical relaxation curves of the echo amplitude as a function of echo time for various rf fields obtained at 20 K. As seen in Fig. 1(a), the relaxation curves at a rf field of $3H_{max}$ are almost single exponential, where H_{max} is the rf level which generates the maximum signal amplitude. In our sample, H_{max} is weakly temperature dependent over the range of the experiment. The single exponential decay means that a rf field of $3H_{max}$ is high enough to cancel out all the signal coming from the domain wall. In our

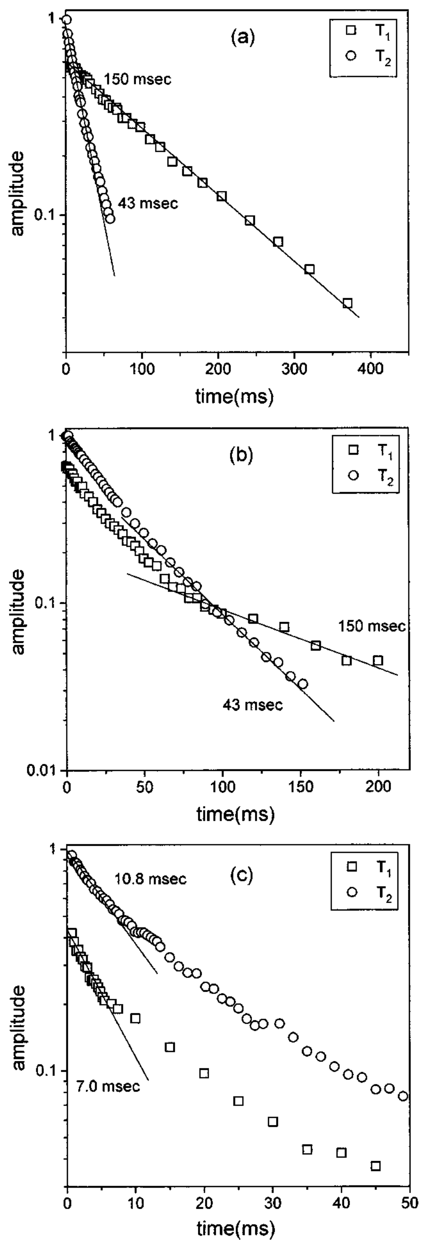

FIG. 1. The relaxation curves of the echo amplitude as a function of echo time for various rf fields obtained at 20 K. The rf fields are $3H_{\mathrm{max}}$ (a), H_{max} (b), and $H_{\mathrm{max}}/5$ (c). The lines represent the exponential fits in (a) and initial slopes in (c), respectively. The lines in (b) are single exponential curves with same relaxation times as (a).

previous study,[7] we showed that the spin echo amplitude at $3H_{\mathrm{max}}$ remained almost constant with increasing external dc field until all the domain walls are swept away at 7.5 kOe. Moreover, the relaxation rates at this rf level are close to those obtained at 8 kOe where a single domain is formed. Therefore, the NMR signal at this rf level is due mainly to the nuclear spins in the domain.

The spin-lattice relaxation rate in the domain increases almost linearly with temperature as seen in Fig. 2. Moriya showed that in ferromagnetic transition metals, the primary contribution to the relaxation comes from the thermal fluctuation of the orbital field at nuclei produced by the d-band electrons in the tight-binding approximation.[8] The relaxation by the $4s$ conduction electrons via the Fermi contact interaction is sometimes appreciable, while other mechanisms, such as the dipole-dipole interaction and spin-wave related mechanisms, were found to be negligible. The relaxation in pure iron due to the Fermi contact interaction and the orbital field of the d electron was estimated to be $1/T_1T \approx 0.046 \sim 0.30\ \mathrm{sec}^{-1}\ \mathrm{K}^{-1}$,[8,9] which is consistent with our experimental value $1/T_1T \approx 0.27\ \mathrm{sec}^{-1}\ \mathrm{K}^{-1}$.

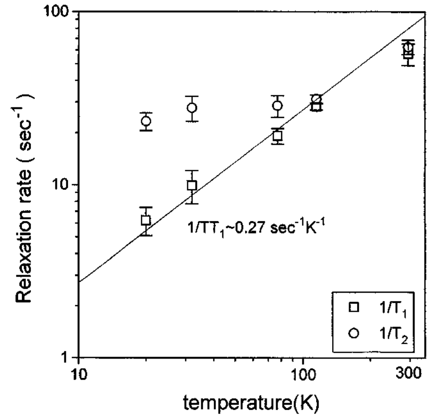

FIG. 2. The spin-lattice and spin-spin relaxation rates in the domain obtained at $3H_{\mathrm{max}}$ as a function of temperature. The line represents the best fit of T_1T=constant curve.

The spin-spin relaxation rate in the domain is relatively constant at low temperature. The Redfield theory[10] gives the spin-spin relaxation rate

$$\frac{1}{T_2}=\frac{1}{T_2'}+\frac{1}{2T_1}, \tag{1}$$

where the first and second terms represent the secular and nonsecular broadening terms, respectively. Two candidates which result in the temperature-independent secular broadening in ferromagnets are the dipole-dipole and Suhl-Nakamura (SN) interactions.[4] The secular broadening due to the SN interaction in 100% Fe^{57}-rich iron was estimated to be about 60 sec^{-1}.[2] Since this broadening is expected to be proportional to Fe^{57} concentration, it is about 1.5 sec^{-1} in natural iron (2.25% Fe^{57}) which is an order of magnitude smaller than the experimental value. The second moment M_2 due to the dipole-dipole interaction for 100 % Fe^{57}-rich iron of bcc structure is 1.45×10^4. The second moment is proportional to the fraction of active nuclei,[11] and therefore, the secular broadening due to the dipole-dipole interaction for a Gaussian line shape $1/T_2' \approx (M_2/2)^{1/2} = (1.45\times 10^4 \times 0.025/2)^{1/2} = 13.5\ \mathrm{sec}^{-1}$ for natural iron. Equation (1) with $1/T_2' \approx 13.5\ \mathrm{sec}^{-1}$ qualitatively explains the experimental results shown in Fig. 2.

The relaxation processes, especially T_1 process, become nonexponential at an rf field of H_{max} [Fig. 1(b)]. Both the spin-spin and spin-lattice relaxation rates at long echo times are pretty much the same with those at $3H_{\mathrm{max}}$ (solid lines). Therefore, the slower single exponential decay is attributed to the nuclear spins in the domain, while the faster decay is attributed to those in the domain wall. In fact, the faster relaxation processes are not single exponential themselves, since there are spreads in relaxation rates in the domain wall. This nonexponential decay in the domain wall is more obvi-

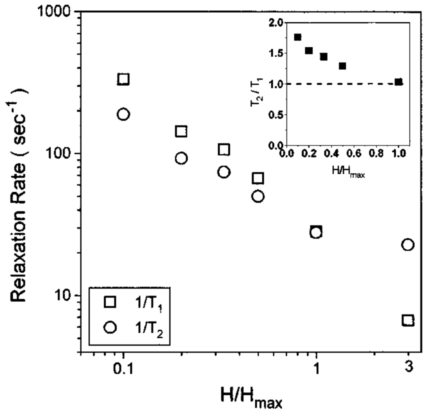

FIG. 3. The spin-lattice and spin-spin relaxation rates, and the ratio T_2/T_1(inlet) in the domain wall obtained from the initial slopes of relaxation curves at 20 K.

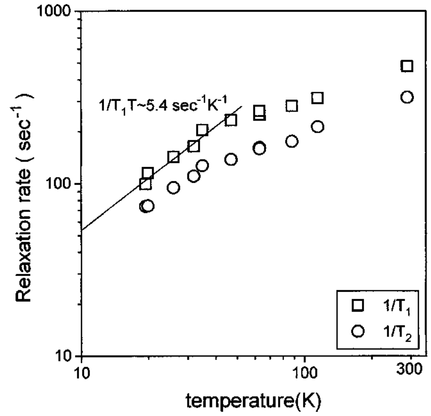

FIG. 4. The spin-lattice and spin-spin relaxation rates at the domain wall center plotted as a function of temperature. The line represents the best fit of T_1T=const curve to the low temperature data.

ous in Fig. 1(c) obtained at $H_{max}/5$. Since the relaxations are much faster than those in the domain, the signal from the domain is rarely mixed at this rf field level. Both the relaxation rates and enhancement factor increase to their maximal values as approaching the centers from the wall boundaries. Therefore, the weaker the rf field is, the more the nuclear spins with larger enhancement factors and short relaxation times contribute to the total signal. The initial slopes of relaxation curves obtained in the limit of zero rf field give the maximal relaxation rates. To our surprise, the initial slope of the spin-spin relaxation curve is smaller than that of the spin-lattice relaxation curve in the domain wall, meaning $T_2 > T_1$. The ratio T_2/T_1, as well as $1/T_2$ and $1/T_1$ obtained from the initial slopes, increases with decreasing rf field as shown in Fig. 3. The ratio approaches 2 in the limit of zero rf field.

This factor of 2 reminds us an anisotropic random fluctuating field which generates only the nonsecular broadening term in Eq. (1). One of the mechanisms suggested to explain the spin-lattice relaxation in ferromagnetic materials is the interaction with magnons. At low temperature, spin waves generate fluctuating local fields at nuclei via the hyperfine interaction

$$\delta\mathcal{H}_{hyp} = A(I^+ \delta S_- + I^- \delta S_+). \qquad (2)$$

Since the fluctuation of local fields due to magnons is transverse as seen in this equation, and the spin-lattice relaxation rate has elements from both transverse components while the spin-spin relaxation rate has only one component, it is expected that $1/T_2 = 1/2T_1$ if this anisotropic fluctuating field is the principal relaxation source.

The relaxation rates due to single magnon processes are linearly dependent on temperature, while those due to three magnon processes are proportional to $T^{7/2}$.[4] Stearns suggested that in ferromagnetic materials the principal mechanism of nuclear relaxations is through interactions with real bulk magnons, because she observed the spin-lattice relaxation rate in the domain wall linearly depends on temperature.[2] Though the energy conservation law forbids the real processes of emission or absorption of one magnon by a nucleus, the damping of magnons due to their interaction with one another, with the lattice, defects, or impurities and the finite magnon lifetime introduce some uncertainty in the magnon energy and single magnon processes become possible. She roughly estimated the spin-lattice relaxation rate by single magnon processes in iron and gave $1/T_1T \approx 5-700$ $\sec^{-1}$ K^{-1} at the domain wall center. On the other hand, Winter suggested magnon processes due to wall-type excitations as the principal relaxation mechanism in the domain wall.[12] This mechanism predicts that the relaxation rate at the center of the wall does not linearly depend on temperature. Weger[1] observed that the relaxation rate in the domain wall depended on temperature weakly and expressed the relaxation mechanism as thermal fluctuations of the domain wall. In Fig. 4, experimentally measured spin-spin and spin-lattice relaxation rates in the domain wall are plotted for various temperatures. The relaxation rates in this plot were obtained from the initial slope at a rf field of $H_{max}/3$. The spin-spin relaxation time is always longer than the spin-lattice relaxation time over the whole experimental temperature range. Both relaxation rates appear to be linear at low temperature, but the slope decreases at high temperature. At low temperature, our experimental data gives $1/T_1T \approx 5.4$ $\sec^{-1}$ K^{-1} which is consistent with Stearns' estimation considering the fact that the initial slope in the limit of zero rf field would give larger relaxation rates. The relaxation rate increasing slower than kT at high temperature seems to imply that the wall-type excitations or fluctuations play a role. Whatever the dominant relaxation mechanism is in the domain wall, T_2 is expected to be longer than T_1 because they are common in predicting that the fluctuation of the hyperfine field is anisotropic. The spin-spin relaxation rate data follows the spin-lattice relaxation data with constant discrepancy in the log-log plot of Fig. 4, which means that the secular term is negligible compared to the nonsecular term in the experimental temperature range. This is consistent with the idea that the dipole-dipole interaction, which gives $1/T_2 \approx 13.5$ $\sec^{-1}$, is the main spin-spin relaxation mechanism. The ratio T_2/T_1 is less than 2 in Fig. 4 because the data is not the values in zero field limit.

Other mechanisms predicting a linear temperature dependence are the nuclear interactions with electron spins or orbital angular momentums and phonons.[4] The former was estimated to be one order of magnitude smaller than the relaxation rate in the domain, and the latter is also expected to give an order of magnitude smaller values. Since the experimentally observed spin-lattice relaxation rate increases not faster than the linear temperature dependence in the whole temperature range, three magnon processes are not dominant relaxation mechanisms in the domain wall.

This unusual phenomenon of $T_2 > T_1$ was predicted by some authors[2,12] and in fact also observed in some works on iron and other samples, though the authors did not pay attention to this fact. It has been observed in the Pt NMR of UPt_3,[13] where $1/T_1$ is anisotropic because the antiferromagnetic fluctuation is slightly stronger in the basal plane than that in the c axis of UPt_3. By careful examination, we can find that T_2 is longer than T_1 at turning angles of $2 \sim 2.3$ rad in Stearns' report[2] on Fe NMR. As far as we know, Robert and Winter's short report[14] on garnet Yttrium is the only one which asserts that $T_2 = 2T_1$ is experimentally verified. This paper was not noticed much because they did not provide enough experimental data to persuade readers, we think.

Intuitively, it seems to be impossible to observe $T_2 > T_1$ because the magnetization vector of an individual spin is fixed length. Suppose that a nuclear magnetization is on the x-y plane after a 90° pulse. The magnetization $M(t)$ should be always less than or equal to the thermal equilibrium value M_0, that is,

$$M(t) = M_0 \sqrt{(e^{-t/T_2})^2 + (1 - e^{-t/T_1})^2} \leqslant M_0 . \qquad (3)$$

Using $x = \exp(-t/T_1)$, this condition can be rewritten as

$$x^{2a} + (1 - x)^2 \leqslant 1, \qquad (4)$$

where $a = T_1 / T_2$. It is trivial to show that this relation can be satisfied for $0 \leqslant x \leqslant 1$ if $a \geqslant 1/2$, that is, $T_2 \leqslant 2T_1$.

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