Temperature dependence of the spin–spin relaxation time of NMR in ferromagnetic $CrBr_3$

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The temperature dependence of the spin–spin relaxation time of nuclei in the domain walls of ferromagnetic $CrBr_3$ has been studied by a two-pulse spin-echo method.

There are two reasons for the interest in the structure and dynamic properties of the domain walls in ferromagnetic materials. First, the widespread use of magnetic materials in computer data storage requires a predictable and controllable behavior of the domain structure, and the basic elements of this structure are the domain walls. Second, there is now a large body of theoretical work, which predicts various properties of domain walls. There is, however, considerable uncertainty regarding these questions, because sufficient experimental information has not been acquired. Even a passing acquaintance with magnetically ordered materials and the hyperfine interaction suggests that nuclear magnetic resonance (NMR) can provide information on the

structural details and the dynamics of domain walls of various scales. The NMR frequency in the hyperfine field is proportional to the absolute value of the magnetization in the domain walls (M_W). Such a parameter as the spin–spin relaxation time is determined in part by low-frequency dipole fields of elementary excitations of the magnetic system with which the nuclear magnetic moments are interacting. The temperature dependence of the NMR frequency was measured in domain walls [i.e., $M_W(T)$ was measured] in ferromagnetic $CrBr_3$ in Ref. 1. In this letter we are reporting a study of the temperature dependence of the spin–spin relaxation time, $T_2(T)$. The results found on the behavior of the magnetization in the domains and in the domain walls may be of assistance in reaching an understanding of the behavior of the spin–spin relaxation rate $\eta = 1/T_2$ and in learning more about spin–spin systems.

The compound $CrBr_3$ has a hexagonal crystal lattice of the BiI_3 type. It goes into a ferromagnetic state at 34.5 K, being one of only a few ferromagnetic insulators.[2] In its magnetically ordered state, it has an easy-axis anisotropy ($H_A = 6.15$ kOe). Its saturation magnetization is[3–5] $4\pi M = 3.38$ kG.

The test crystals in the present experiments were thin platelets (with a thickness on the order of 100 μm and an area on the order of 1 cm^2).[1)] The sixfold axis (the easy axis of the magnetic anisotropy) runs perpendicular to the plane of the platelets. We used a stack of (about 20) platelets in order to increase the signal-to-noise ratio in the measurements. The C_6 axes for all of these crystals were parallel, while the orientation in the plane was random. The sample was placed in the inductance coil of an LC circuit (whose resonant frequency could be varied over the range 32–73 MHz by adjusting the capacitor). The LC circuit, with the sample, was placed in a cryostat, in which the temperature could be stabilized and regulated. This circuit served as the absorbing cell of a Bruker MSL-300 NMR spectrometer.

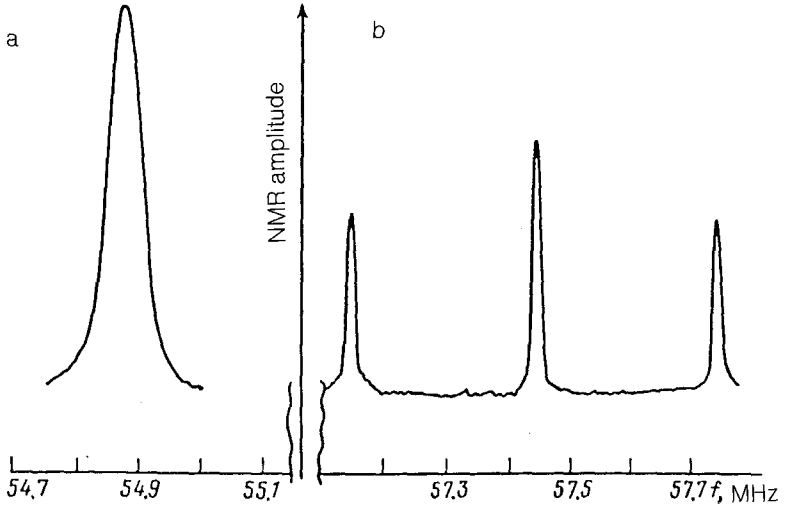

FIG. 1. NMR spectrum of Cr^{53} nuclei in ferromagnetic $CrBr_3$ at a temperature of 4.2 K in a zero external magnetic field. a—NMR line in the domain walls; b—quadrupole-split NMR in the domains.

Figure 1 shows recordings of the NMR lines of Cr^{53} nuclei in chromium tribromide at 4.2 K in a magnetic field $H = 0$. The triplet with the central frequency of 57.434 MHz is a quadrupole-split NMR signal of the domains. The isolated line (54.87 MHz) is the NMR line of the domain walls. The domain structure in $CrBr_3$ platelets in a zero magnetic field is labyrinthal[3] with a domain width on the order of 10 μm. The width of the domain wall amounts to several lattice constants ($\sqrt{H_E/H_A} \approx 6$) because of the fairly strong crystalline anisotropy and the low exchange field. The ratio of the volume of the domains to the volume of the walls is on the order of several hundred. The total intensity of the NMR from the domain walls, on the other hand, is significantly higher than the intensity of the NMR from the domains, apparently because of a difference in the gain values for the NMR in the domains and in the walls. A difference between the NMR frequencies of domains and domain walls was linked in Ref. 1 with differences in the saturation magnetization in these formations and with differences in the temperature dependence of these properties. One problem which has yet to be explained is the absence of a quadrupole splitting of the NMR lines in the domain walls.

The width of the NMR lines in the domains (the triplet in Fig. 1) is close to 10 kHz, while that in the domain walls (the single line in Fig. 1) is about 50 kHz. To determine whether the broadening of the NMR lines is homogeneous or inhomogeneous, we measured the spin–spin relaxation time T_2, using the two-pulse spin-echo technique.

The spin-echo signal is easily observable in the domain walls. It differs somewhat from the standard Hahn echo, and it is observed slightly earlier (in time). We did not make a detailed study of this difference, but the scale of the effect suggests a time shift

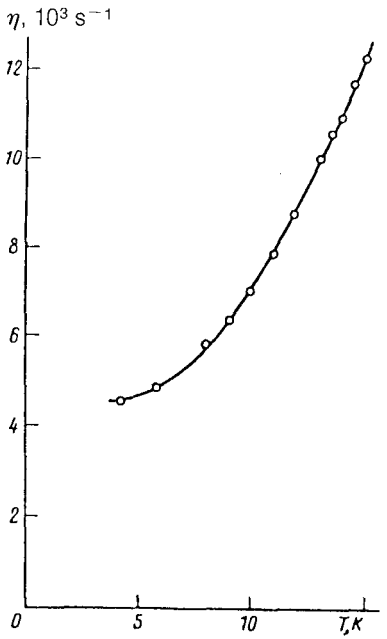

FIG. 2. Spin–spin relaxation rate of the NMR in the domain walls as a function of the temperature.

TABLE I.

Temperature (K)	4,2	6	8	9	10	11	12	13	13,5	14	14,5	15
T_2 (μs)	220	205	172	158	143	128	113	100	95	92	86	82

of the echo amounting to a few microseconds at a delay time on the order of hundreds of microseconds. This difference causes no problems in measurements of the spin–spin relaxation time. We studied the temperature dependence from 4.2 to 15 K. Figure 2 shows the results, plotted as the relaxation rate, $\eta = 1/T_2$. The use of this quantity simplifies the interpretation, since when several processes are operating in parallel, the overall relaxation rate is simply the sum of the rates of the individual processes. Table I gives an idea of the change in T_2 itself as a function of the temperature.

A mathematical analysis of the experimental results yields the expression

$$\eta = 1/T_2 = (4189 + 7.36T^{2.59})\ [\mathrm{s}^{-1}]. \qquad (1)$$

If we assume that the power to which the temperature is raised in (1) is simple, namely, 2.5, then the relaxation rate is described by

$$\eta = (4094 + 9.43T^{2.5})[\mathrm{s}^{-1}].$$

We can work from this value of the relaxation rate to estimate the homogeneous width of the NMR line in the domain walls. At 4.2 K we find

$$\Delta f = \eta/2\pi \approx 1\ \mathrm{kHz}\ .$$

This figure is significantly smaller than the width (10 kHz) of the NMR line in the domains, for which we have assumed a homogeneous broadening. We accordingly measured the spin–spin relaxation time of the NMR in the domains. The results of these measurements are less reliable than those of the preceding measurements. The reason is that the NMR lines have a lower total intensity, there is a quadrupole splitting, and there is a significant deviation of the time dependence of the echo amplitude from an exponential shape. The measurements show that the time T_2 in the domains is 30–40% shorter than T_2 in the domain walls. It is nevertheless greater than the reciprocal of the line width; in other words, an inhomogeneous broadening is occurring in the domains. (The results of a study of the NMR of ferromagnetic $CrBr_3$ in a magnetic field will be published in a detailed paper.) Suladze and Khutsishvili[6] have calculated the rate of nuclear spin–spin relaxation of a ferromagnet, taking into account the interaction of the nuclear magnetic system with the spin waves of the domain walls (Winter modes[7]). Our results do not agree with that calculation. The reason for the discrepancy may be the high frequency of these spin waves in $CrBr_3$ (Ref. 8).

That component of the relaxation rate (and of the corresponding spin–spin relaxation time), which is independent of the temperature, is determined by an indirect interaction of nuclear spins through the ferromagnetic system.[9]

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[1)]The $CrBr_3$ crystals were grown by a gas-transport method by L. A. Klinkova of the Institute of Solid State Physics, Russian Academy of Sciences.

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