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Angular dependence of nuclear spin echo decay in thin-film yttrium iron garnet

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Abstract

The angular dependence of the nuclear spin echo decay of ^{57}Fe in monocrystalline thin-film $Y_3Fe_5O_{12}$ was measured. The experimental results for the octahedral Fe^{3+} a-ions were explained by the orientation fluctuations of the electron magnetization about the local symmetry axis.

The nonexponential decay of the nuclear spin echoes of ^{57}Fe in magnetically ordered yttrium iron garnet $Y_3Fe_5O_{12}$ (YIG) has been observed by several workers [1–3]. This nonexponential decay was explained by Ghosh [1] on the basis of a model of fluctuations in the isotropic constant of the hyperfine magnetic field at the ^{57}Fe nucleus by the temperature-dependent spin fluctuations of the Fe^{3+} ions. In YIG the Fe^{3+} ions occupy both the tetrahedral (d) and octahedral (a) lattice sites [4]. For Fe^{3+} d-ions the hyperfine field at the ^{57}Fe nucleus is isotropic, but for Fe^{3+} a-ions the hyperfine field is anisotropic [5,6]. The fluctuations in the isotropic constant of the hyperfine magnetic field only must lead to the same rates of spin echo decay for the four magnetically nonequivalent Fe^{3+} a-ions. However, we observed in a polycrystalline thin film of YIG that the rates of nonexponential spin echo decay are different for the magnetically nonequivalent Fe^{3+} a-ions [3]. In this paper the nonexponential decay of the nuclear spin echoes of ^{57}Fe ions occupying a-sites is investigated in a monocrystalline thin film of YIG.

The NMR measurements were made at $T = 77$ K by the incoherent spin-echo method (Hahn two-pulse sequence [4]) on the thin-film sample of YIG, enriched to about 96% ^{57}Fe. The YIG films 8 μm thick were prepared by the method described in Ref. [3]. The normal to the film plane coincided with the crystallographic direction [111]. Spin-echo NMR measurements were made in the presence of an external magnetic field ($B > 1000$ G), when the sample of YIG was monodomain [2]. The external magnetic field $\boldsymbol{B}$ was applied in the film plane. The radio-frequency (rf) field $\boldsymbol{B}_1$ was also applied in the film plane and the orientation of $\boldsymbol{B}_1$ was perpendicular to $\boldsymbol{B}$ (Fig. 1). The width of the first rf pulse was 2 μs and the width of the second pulse was 4 μs.

The NMR resonance frequency (ν_I) of ^{57}Fe occupying a-sites of YIG is given by [5,6]

$$\nu_I = \left[B + B_0 + B_A(3\cos^2\theta - 1) \right](\gamma/2\pi), \qquad (1)$$

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where γ is the gyromagnetic ratio of the ^{57}Fe nucleus; B_0 and B_A ($B_A = -2500$ G [5]) are the isotropic and anisotropic constants, respectively, of the magnetic hyperfine field at the ^{57}Fe nucleus; B is the external magnetic field; and θ is the angle (Fig. 1) between the direction of the electron magnetization $\boldsymbol{M}$ and the direction of the local symmetry axis $\boldsymbol{n}_i$ (the axes of the types [111]: [111], $[\bar{1}1\bar{1}]$, $[\bar{1}\bar{1}1]$, $[1\bar{1}\bar{1}]$).

It follows from (1) that if we choose the axis [111] as the OZ axis and $[1\bar{1}0]$ as the OX axis (Fig. 1) then the resonance frequencies of the four magnetically nonequivalent ^{57}Fe nuclei are given by (a) the local symmetry axis [111] ($\cos\theta_1 = 0$):

$$\nu_I^{(1)} = [B + B_0 - B_A](\gamma/2\pi); \tag{2a}$$

(b) the local symmetry axis $[\bar{1}1\bar{1}]$:

$$\cos\theta_2 = -\sqrt{\frac{2}{3}}\cos\varphi + \frac{\sqrt{2}}{3}\sin\varphi,$$

$$\nu_I^{(2)} = \left[B + B_0 + \frac{B_A}{3}(1 + 2\cos 2\varphi - 2\sqrt{3}\sin 2\varphi)\right] \times\left(\frac{\gamma}{2\pi}\right); \tag{2b}$$

(c) the local symmetry axis $[\bar{1}\bar{1}1]$:

$$\cos\theta_3 = -\frac{2\sqrt{2}}{3}\sin\varphi,$$

$$\nu_I^{(3)} = \left[B + B_0 + \frac{B_A}{3}(1 - 4\cos 2\varphi)\right]\left(\frac{\gamma}{2\pi}\right); \tag{2c}$$

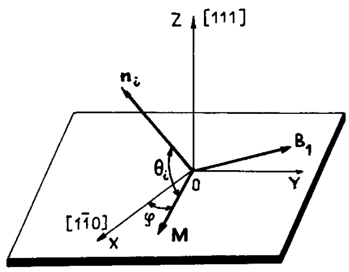

Fig. 1. Orientations of the coordinate frame XYZ and $\boldsymbol{M}$, $\boldsymbol{B}_1$, $\boldsymbol{n}_i$ relative to the plane of the thin-film YIG.

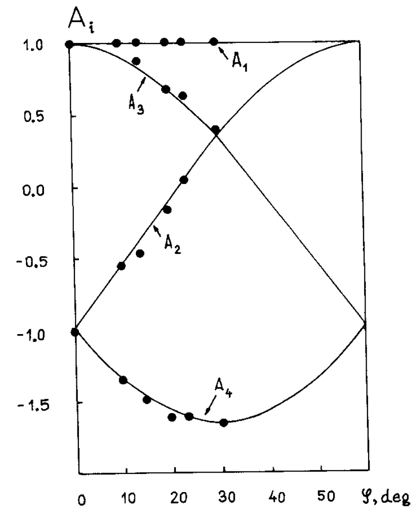

Fig. 2. Angular dependences of $A_i = (1 - 3\cos^2\theta_i)$ for the four magnetically nonequivalent ^{57}Fe nuclei in thin-film YIG.

(d) the local symmetry axis $[1\bar{1}\bar{1}]$:

$$\cos\theta_4 = \sqrt{\frac{2}{3}}\cos\varphi + \frac{\sqrt{2}}{3}\sin\varphi,$$

$$\nu_I^{(4)} = \left[B + B_0 + \frac{B_A}{3}(1 + 2\cos 2\varphi + 2\sqrt{3}\sin 2\varphi)\right] \times\left(\frac{\gamma}{2\pi}\right). \tag{2d}$$

It follows from Eqs. (2) that the values A_i ($i = 1, 2, 3, 4$)

$$A_i(\varphi) = 1 + \frac{2\pi}{\gamma}\frac{\left(\nu_I^{(1)} - \nu_I^{(i)}\right)}{B_A} = 1 - 3\cos^2\theta_i \tag{3}$$

are not dependent on B. The angular dependences of A_i are shown in Fig. 2.

Measurements of the spin echo decay as a function of the time separation of the two rf pulses (Fig. 3) were made for all well resolved NMR lines. The angle θ was re-established with the help of Eq. (3).

In order to explain the observed angular dependence of the spin echo decay we consider the spectral diffusion model [7–9] and assume that the non-exponential decay of the spin echo signal in YIG is due to the time fluctuations in the resonance frequency ν_I of the ^{57}Fe nucleus. For the Markov–Lorentzian process of the resonance frequency–time fluctuations, the echo amplitude at $t = 2\tau$ is given by [7,8]

$$V(2\tau) = V(0)\exp\left[-2\sigma\left(\tau - \tau_c \ln\left(2 - \exp\left(-\frac{\tau}{\tau_c}\right)\right)\right)\right], \quad (4)$$

where τ_c is the correlation time of the time fluctuating frequency ν_I (τ_c^{-1} is the mean frequency of ν_I time fluctuations); τ is the time interval between the first and second pulses; and σ is the width of the Lorentzian shape of the inhomogeneously broadened NMR spectrum ($\tau_c \to \infty$).

The solid lines in Fig. 3 represent the theoretical curves obtained from the best fit to the observed values of $V(2\tau)$ by Eq. (4) with the same τ_c and different σ. The fitting value of the correlation time is $\tau_c = 5 \times 10^{-4}$ s. From Fig. 3 we may conclude that our experimental results are well explained by the Markov–Lorentzian spectral diffusion model [7,8].

Fig. 4 shows the angular dependence of the constant σ obtained from the experimental curves $V(2\tau)$ with the help of Eq. (4) for $\tau_c = 5 \times 10^{-4}$ s. As can

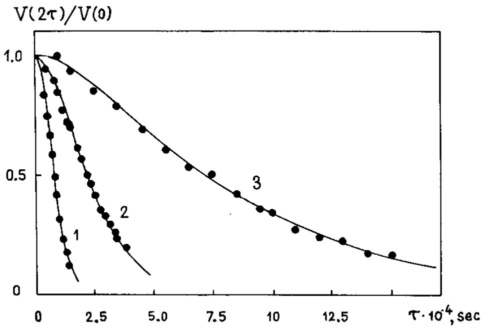

Fig. 3. Angular dependence of the nuclear spin echo decay of ^{57}Fe in thin-film YIG. The solid lines show the least-squares fits to Eq. (4). 1, $\theta = 48°$ ($\sigma = 32$ kHz); 2, $\theta = 76°$ ($\sigma = 5$ kHz); 3, $\theta = 90°$ ($\sigma = 0.78$ kHz).

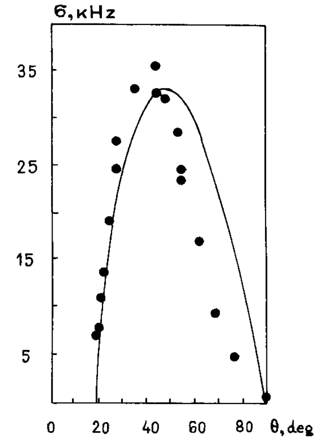

Fig. 4. Angular dependence of the constant σ. The solid line shows the least-squares fit to Eq. (7).

be seen from Fig. 4, the width σ of the Lorentzian shape of the inhomogeneously broadened NMR spectrum exhibit the characteristic angular (θ) dependence and $\sigma(\theta)$ takes a maximum value for $\theta \approx 40°$. To explain the observed angular dependence of σ we assume that the Lorentzian shapes of the inhomogeneously broadened NMR spectra are caused by the spread in the values of the angle θ as a result of a spread in the directions of the electron magnetization $\boldsymbol{M}$. In a thin film the spread in the directions of $\boldsymbol{M}$ may be only in the plane of the thin film. According to Eqs. (2) the spread ($\delta\varphi$) in the values of the angle φ leads to the spreads ($\delta\nu_I^{(i)}$) in the values of the resonance frequencies $\nu_I^{(i)}$ of the ^{57}Fe nuclei

$$\delta\nu_I^{(i)} = \left(\frac{d}{d\varphi}\nu_I^{(i)}\right)\delta\varphi. \quad (5)$$

The expressions for $\delta\nu_I^{(i)}$ ($i = 1, 2, 3, 4$) can be calculated easily from Eqs. (2)

$$\delta\nu_I^{(i)} = -\frac{\gamma}{\pi}B_A \cos\theta_i\sqrt{|8 - 9\cos^2\theta_i|}\,\delta\phi. \quad (6)$$

Assuming that the value of σ is proportional to $\delta\nu_I$, and omitting the index i in Eq. (6), we can write

$$\sigma(\theta) = \sigma_0 \cos\theta\sqrt{|8 - 9\cos^2\theta|}. \quad (7)$$

where

$\sigma_0 = \gamma/\pi \mid B_A \mid \sqrt{\langle \delta\varphi^2 \rangle}$

and $\langle \delta\varphi^2 \rangle$ is the mean-square distribution of the angle φ.

We then fitted our experimental data for $\sigma(\theta)$ with Eq. (7) using the method of least squares (Fig. 4). From such a fit, we obtain $\sigma_0 = 25$ kHz and $\langle \delta\varphi^2 \rangle^{1/2} \approx 2°$.

Reasonable agreement between experiment and theory suggest that the model of the orientational time fluctuations of the electron magnetization $\boldsymbol{M}$ gives a fair description of the angular dependence of the nuclear spin echo decay for a-ions Fe^{3+}.

In conclusion, we point out that investigations of nuclear spin echo decay in YIG with various contents of ^{57}Fe nuclei demonstrate that the rate of spin echo decay increases with increasing ^{57}Fe content [2,3]. The concentration dependence of the nuclear spin echo decay indicates that the Suhl–Nakamura interaction [4] between nuclear spins of ^{57}Fe plays an important role in the nuclear spin echo dynamics in YIG. In order to have a better understanding of the physical mechanism of the orientational fluctuations of the electron magnetization $\boldsymbol{M}$ and their relationship to the Suhl–Nakamura interaction, further experiments under different conditions (temperature, various contents of ^{57}Fe nuclei) are necessary.

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