Spin modulation of ^{57}Fe NMR frequency and relaxation in $BiFeO_3$

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Abstract

NMR spectra were measured in ^{57}Fe enriched ferroelectric-antiferromagnet $BiFeO_3$ with space modulated magnetic structure of cycloidal type. This structure leads to space modulation of local field on ^{57}Fe nuclei as well as space periodic spin-spin relaxation and specific NMR line shape with frequency variable local width. Line shape temperature behavior is described using the model of nuclei in Bloch walls. This effect is based on indirect Sule-Nakamura spin-spin interaction which becomes effective at high concentration of magneto-active nuclei as a consequence of excitement of spin waves.

Key words: spin-lattice relaxation; spin-spin interaction; NMR

1. Introduction

$BiFeO_3$ is a perovskite-like compound with magnetic and electrical long-range order with antiferromagnetic transition temperature $T_N = 670$ K. It has space modulated magnetic structure of cycloidal type with the period $\lambda = 620 \pm 20\,\mathring{A}$ incommensurate with the lattice parameter [1], [2]. The magnetic moments of the Fe^{3+} ions, while retaining local antiferro-magnetic G-type mutual orientation, are turned in the plane perpendicular to the hexagonal basal plane along the propagation direction of the modulated wave. The wave vector of such a cycloid is perpendicular to the threefold *c*-axis and lies in the plane of spin rotation. The existence of a spiral magnetic structure has been neither confirmed nor refuted by any alternative microscopic method yet. This structure leads to space modulation of local field on ^{57}Fe nuclei as well as space periodic spin-spin relaxation and specific NMR line shape with frequency variable local width. Therefore, NMR in hyperfine field could clarify this problem.

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2. Results and discussion

The experimental ^{57}Fe NMR spectrum of $BiFeO_3$ in zero external magnetic field is shown in Fig.1. Spin echo was exited by equal 10 μs pulses with 80 μs delay. The spectrum consists of two peaks of different heights. It holds the same shape at higher temperatures, but becomes more asymmetric with degraded low frequency peak [3]. The spectrum of a similar type with asymmetric lineshape is known to exist when the resonance frequency becomes a periodical function of a coordinate [4–6]. To describe the experimental spectrum we used the angular dependence of the resonance frequency due to anisotropic contribution into the hyperfine field at ^{57}Fe nuclei and the dependence of a spin rotation angle on the coordinates along the spiral direction [3]. The anisotropy of NMR frequency were defined in [7]:

$$\nu \approx \nu_{||} - (\nu_{||} - \nu_{\perp}) sin^2\theta = \nu_{||} - \delta\nu sin^2\theta \qquad (1)$$

where $\nu_{||}$ and $\nu_{\perp}$ are the NMR frequencies for the spin direction parallel (θ=0) and perpendicular (θ=π/2) to the *c*-axis, θ is the angle between magnetization $\vec{M}$ and *c*-axis. The spatial distribution of θ is determined by the elliptic function:

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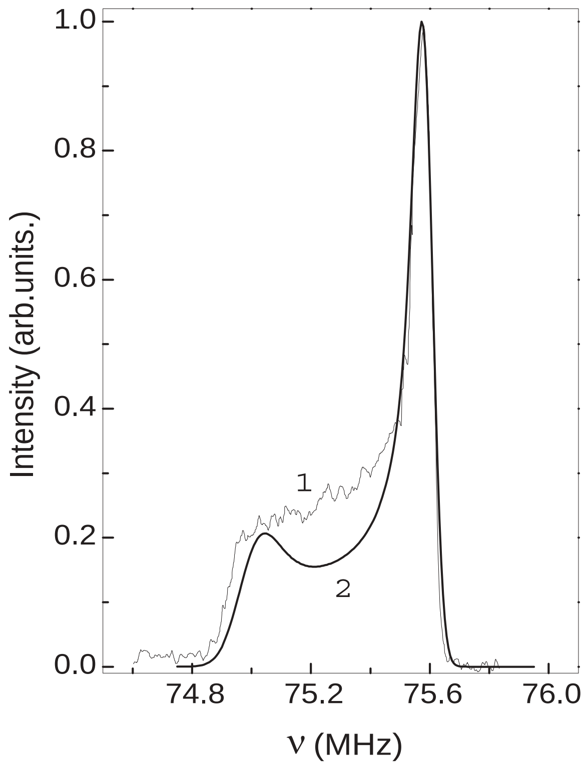

Fig. 1. (1) ^{57}Fe NMR spectrum of $BiFeO_3$ in zero external magnetic field at 4.2 K. (2) Line shape simulation according to the equation (3)

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$$cos\theta(\overrightarrow{r}) = sn(\pm\sqrt{\frac{K_u}{mA}}x, m) \quad (2)$$

where m is the anharmonic parameter [7].

The detailed measurements of the spin-spin relaxation at $\nu_{||}$ and $\nu_{\perp}$ revealed the frequency dependence of T_2 (Fig.2) which was taken into account in line shape simulation:

$$P(\nu) \propto \int_0^{\pi} \frac{(m^{-1} - 1 + sin^2\theta)^{-1/2}}{\triangle_{||} + \delta_1 sin^2\theta} B d\theta \quad (3)$$

where

$$B = exp\{-\frac{2[\nu - (\nu_{||} - \delta\nu sin^2\theta)]^2}{(\triangle_{||} + \delta_1 sin^2\theta)^2}\}, \quad (4)$$

local Gaussian linewidth $\triangle(\theta) = \triangle_{||} + \delta_1 \sin^2\theta$; δ_1 - additional broadening at $\nu_{\perp}$. The best fit is shown as a solid line in Fig.1 and corresponds to parameter m = 0.95. Line shape temperature behaviour is described using the model of nuclei in Bloch walls. This effect is based on indirect Sule-Nakamura spin-spin interaction which becomes effective at high concentration of magneto-active nuclei as a consequence of excitement of spin waves [8,9]. The details of the calculations are published in [9].

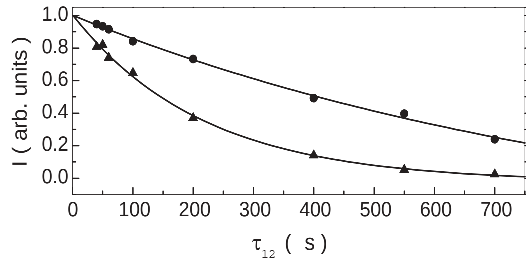

Fig. 2. Echo decay curves as a function of time delay t_{12} between pulses in spin-echo experiment at $\nu_{||}$ (circles) and $\nu_{\perp}$ (triangls).

3. Conclusion

The existence of spin modulated magnetic structures at Fe-sites in $BiFeO_3$ has been confirmed by nuclear magnetic resonance. The structure has the cycloidal type with the period $\lambda = 620 \pm 20$ $\mathring{A}$ and anharmonic parameter $m = 0.95$. This structure results in specific line shapes of ^{57}Fe NMR spectra as well as in space modulation of transverse relaxation rate and local linewidth.

References

[1] F. Kubel,H. Schmid, Acta Cryst.B **46** (1990) 698.

[2] I. Sosnowska,T. Peterlin-Neumaier,E. Steichele, J.Phys.C: Solid.State Phys. **15** (1982) 4835.

[3] A.V. Zalessky,A.K. Zvezdin,A.A. Frolov,A.A. Bush, JETP Letters **71** (2000) 682.

[4] I.P. Alexandrova, Incommensurate Phases in Dielectrics, ed. by R. Blinc and A.P. Levanyuk (1986) 277.

[5] M. Horvatic *et al.*, Physica B **22** (1998) 246-247.

[6] A.V. Zalessky,A.K. Zvezdin, Phys. Status Solidi B **73** (1976) 317.

[7] A.V. Zalessky *et al.*, Europhysics Letters **50** (2000) 547.

[8] M.A. Bulter, Int.J.Magnetism **4** (1973) 131.

[9] A.V. Zalessky *et al.*, JETP July (2002).