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Spin-phonon coupling in MnZn ferrites

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Abstract

Ultrasonic attenuation and dispersion are studied in polycrystalline MnZn ferrites. Both factors are strongly influenced by the coupling of sound waves to the spin system. This coupling is governed by the λ_{111} magnetostriction constant only, and not by λ_{100}. A simple model can account for all the observed results in a quantitative fashion. Using this model, values of λ_{111} can be determined from acoustic and susceptibility measurements which show good agreement with λ_{111} data from the literature on static torque measurements of single crystals.

1. Introduction

Of all the ferrites, MnZn ferrites are the most commercially important class of materials. They have come to be used in video heads, power transformers and many other electronic components [1]. It is ever-important to study the coupling of mechanical excitations of the crystal lattice to the spin system of these materials; this process is what ultimately determines, for example, the rubbing noise of tapes on the video head. In this paper we address this issue in an unconventional way by studying the influence of the spin system on ultrasonic attenuation. As a result, only shear waves couple to the spin system and longitudinal waves do not participate in this coupling. The results are described in terms of a simple model. The results will be applied to experiments, details of which will be presented in a subsequent paper. Central to the results is the finding that ultrasonic attenuation is basically determined by the magnetostriction constant and the imaginary part of the magnetic susceptibility. Additional effects do not seem to have a significant influence.

2. The model

An expansion of the Free energy of a cubic spinel ferrite about its thermodynamic equilibrium yields, to the lowest order in the magnetization m and strain tensor $u_{ij}=(\partial_i u_j+\partial_j u_i)/2$ [2]

$$F=\int\Big\{\frac{1}{2}c_{11}(u_{xx}^2+u_{yy}^2+u_{zz}^2)+c_{12}(u_{xx}u_{yy}+u_{xx}u_{zz}+u_{yy}u_{zz})$$

$$+2c_{44}(u_{xy}^2+u_{xz}^2+u_{yz}^2)-6\lambda_{111}c_{44}\frac{1}{\bar{m}}(u_{xz}m_x+u_{yz}m_y)$$

$$+\frac{\mu_0}{2\chi}(m_x^2+m_y^2)\Big\}\,dV \tag{1}$$

Here, we have assumed that the equilibrium magnetization is pointing in the z-direction and that $\bar{m}=(m_x^2+m_y^2+m_z^2)^{1/2}$ is the saturation magnetization. To within this order of expansion, only λ_{111} contributes to the magnetostriction while the λ_{100} component enters in third order approximation. As a consequence only shear strain couples to the spin system, whereas compressional strain is expected to be only weakly dependent on magnetization. A tacit assumption behind eqn. (1) is that the susceptibility χ is determined mainly by rotational permeability and not by domain wall permeability [3].

The dynamics of the material is modeled by a set of coupled sound equations and Landau–Lifschitz equations [4]

$$\rho\ddot{u}_i=-\frac{\delta F}{\delta u_i} \tag{2}$$

$$\dot{m}_x(t)=\gamma\bar{m}\frac{\delta F}{\delta m_y(t)}-\int dt'/\lambda(t-t')\frac{\delta F}{\delta m_x(t')} \tag{3}$$

$$\dot{m}_y(t)=-\gamma\bar{m}\frac{\delta F}{\delta m_x(t)}-\int dt'\lambda(t-t')\frac{\delta F}{\delta m_y(t')} \tag{4}$$

Here, $\gamma\mu_0$ is the gyromagnetic constant and $\lambda(t)$ represents a spectrum of relaxation times. Ignoring, for a while, the magnetostrictive coupling between eqns. (2)–(4), eqn. (2) represents the well known sound

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equation for undamped longitudinal and transverse sound, while eqns. (3) and (4) describe the damped precession of the spin system. In particular, the dynamic response functions $\chi_x(\omega) = \partial m_x(\omega)/\partial h_x$ and $\chi_y(\omega) = \partial m_y(\omega)/\partial h_y$ ($m_x(\omega)$ and $m_y(\omega)$ are the Fourier components of $m_x(t)$, $m_y(t)$ and h_x, h_y, external fields in the x- and y-directions, respectively) are nothing but the complex susceptibilities. For $\omega = 0$, $\chi(\omega)$ is identical to the static susceptibility of eqn. (1). Equations (2)–(4) are linear equations which can be evaluated in a straightforward way. From the resulting dispersion relations it can be deduced that the sound propagation is modified according to

$$\tilde{c}_{44}(\omega) = c_{44}\left(1 - 9c_{44}\lambda_{111}^2 \frac{\chi(\omega)}{\mu_0 \bar{m}^2}\right) \tag{5}$$

for transverse sound modes with a displacement vector $\boldsymbol{u}$ perpendicular to the magnetization direction. However, c_{44} remains unchanged for displacements parallel to the magnetization and c_{11} c_{12} are not modified. The structure of the modification is apparent from eqn. (1) when determining the free energy. In a polycrystalline material which shows random distribution of the magnetization direction, the effective elastic constant is obtained by averaging over the three directions.

$$\tilde{c}_{44}^{\rm eff}(\omega) = c_{44}\left(1 - \frac{2}{3}\, 9c_{44}\lambda_{111}^2 \frac{\chi(\omega)}{\mu_0 \bar{m}^2}\right) \tag{6}$$

From eqn. (6), the magnetization dependent modifications of the sound velocity v and the attenuation α read

$$\frac{\delta v}{v} = -3c_{44}\lambda_{111}^2 \frac{1}{\mu_0 \bar{m}^2} \operatorname{Re}(\chi(\omega)) \tag{7}$$

$$\alpha = \frac{\omega}{v}\, 3c_{44}\lambda_{111}^2 \frac{1}{\mu_0 \bar{m}^2} \operatorname{Im}(\chi(\omega)) \tag{8}$$

Equations (7) and (8) are the main results of this work, we tested them in the following ways.

3. Experimental details and results

The experiments were performed on a polycrystalline ferrite with composition $Mn_{0.74}Zn_{0.18}Fe_{2.08}O_4$. Susceptibility measurements were done on small toroidal rings using a commercial impedance analyzer. For the acoustic measurements, pieces 5 mm × 5 mm × 5 mm were cut and $LiNbO_3$ transducers were attached to the polished surfaces. A pulse echo technique with an automatic phase comparison method was employed. The acoustic measurements were always done in two consecutive runs, one without a field and the second one in presence of an external field ($\boldsymbol{B}\|\boldsymbol{k}$) of 0.5 T. At such a field strength the magnetization saturates and so the susceptibility decreases to nearly zero. This implies that the spin system can be decoupled from the crystal lattice allowing separation of the magnetization-induced effects on the ultrasound attenuation.

In a first experiment, longitudinal sound was investigated as a function of the applied field. Virtually no change in sound velocity and attenuation was measured when a field was applied; as suggested by the theoretical

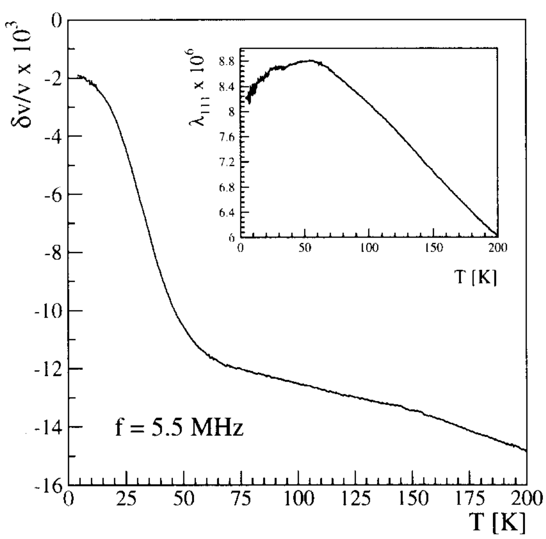

Fig. 1. Magnetization dependent shift of the transverse ultrasound velocity $\delta v/v = [v(0) - v(B)]/v(0)$. The magnetic field of 0.6 T was applied parallel to the propagation direction. The frequency was 5.5 MHz. Inset shows the magnetostriction constant calculated from $\delta v/v$ Re_χ.

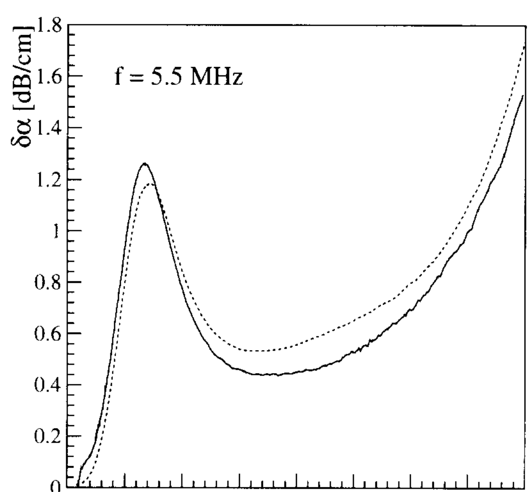

Fig. 2. Magnetization dependent ultrasonic attenuation, $\delta\alpha = \alpha(0) - \alpha(B)$, for transverse sound mode (full line). The magnetic field of 0.6 T was and applied parallel to the propagation direction. The frequency was 5.5 MHz. The dashed line represents the theoretical prediction using λ_{111} data from Fig. 1 and experimental values of Im_χ. Absolute height was scaled down by a factor of two.

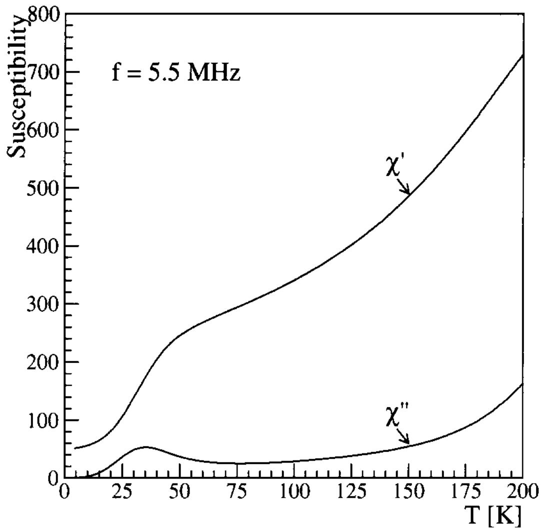

Fig. 3. Real and imaginary part of the complex susceptibility *vs.* temperature.

model. However, transverse sound exhibits very distinct dependence on magnetic field. In Fig. 1, the velocity shift is shown for 5.5 MHz. In Fig. 2, the corresponding attenuation data are presented. The kink in the velocity and the associated maximum of the attenuation at about 30 K is probably related to a cubic to orthorhombic phase transition which was thoroughly studied in the case of pure magnetite by Verwey [5]. This interpretation is supported by resistivity measurements which show a sharp rise at about the same temperature.

In Fig. 3, the real and imaginary parts of the complex susceptibility are presented. Again, the transition at 30 K is clearly recognizable. The results are evaluated in the following way: using eqn. (7) and the measured values for $\delta v/v$ and Re_χ the magnetostriction constant λ_{111} can be determined (see Fig. 1, inset) where we have assumed constant values for $\mu_0\bar{m} = 0.5$ T; $v = 2850$ ms^{-1}, and $\rho = 4570$ kgm^{-3}. The resulting λ_{111} value correlates surprisingly well with the values given in the literature [6]. Using the λ_{111} value and the experimental values of Im_χ given in Fig. 3, the attenuation can be predicted without any adjustable parameter (eqn. (8)). The result is presented in Fig. 2 (dashed line). Apart from an overall scaling factor of the order of two, the theoretical curve correlates well with the experimental curves. We consider this as a direct proof of the validity of our model which connects ultrasound with susceptibility data. This connection was missing from previous work on the subject where the attenuation was either attributed to domain wall motion [7] or internal stress [8]. The discrepancy factor of two between theory and experiment is probably a result of the geometry effects which cause deviation from plane wave propagation of sound.

In conclusion we have demonstrated that ultrasound propagation is directly linked to susceptibility data. This link is provided by the λ_{111} magnetostriction constant. Hence, the method provides an access to magnetostriction constant values at high frequencies even in polycrystalline materials. More details of the present work will be the subject of a forthcoming paper.

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