Magnetoelasticity in $SmFe_2$

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The temperature and magnetic field dependence of the elastic moduli of $SmFe_2$ Laves phase compound has been investigated by means of an ultrasonic-pulse technique at a frequency of 10 MHz. The spin reorientation from the high-temperature easy direction of magnetization [111] to the low-temperature easy axis directed along [110], was observed to occur at $T_{sr} = 195$ °K. The behavior of the elastic moduli near T_{sr} suggests a first-order phase change. Applied magnetic fields strongly affect the temperature dependence and the absolute values of the elastic moduli of $SmFe_2$. The elastic moduli increase significantly with increased magnetic fields, yielding a low and temperature-independent adiabatic compressibility. A remarkably high (42%) ΔE effect was observed in $SmFe_2$ at room temperature, 10-MHz ultrasonic frequency, and an applied magnetic field of 25 kOe. The magnetostriction of $SmFe_2$ is high and negative. For a field of 25 kOe at room temperature and 77 °K values are -2100×10^{-6} and -3340×10^{-6}, respectively. The limiting value of the Debye temperatures were found to be 200 and 212 °K, for the unmagnetized and at 25 kOe, respectively.

INTRODUCTION

The cubic rare-earth–iron Laves phases (type $MgCu_2$) form an interesting group of compounds the magnetic properties of which are strongly influenced by magnetoelastic phenomena. Several of these compounds have been found to exhibit the highest known magnetostrictions at room temperature.[1,2] Their magnetic properties have been investigated by a variety of experimental techniques.[3–6] The heavy-rare-earth–iron Laves compounds are magnetically ordered at room temperature with an antiparallel coupling between the moments of the two sublattices.[7,8]

Recent Mössbauer-effect studies have shown that the one-ion model is remarkably successful in accounting for the complex magnetic anisotropy behavior of the heavy-rare-earth–iron binary RFe_2,[9] and the ternary $R^1_x R^2_{1-x} Fe_2$[10–12] cubic Laves compounds. The occurrence of several spin reorientations due to change in composition, or temperature, finds its explanations within this model in spite of the fact that the magnetoelastic effects were ignored. However, the giant magnetostrictions observed[1,2] indicate that magnetoelastic effects should play an important role.

Mössbauer-effect measurements in $SmFe_2$[13] have shown that the [110] direction is the easy axis of magnetization at low temperatures. Between 140 and 240 °K, a spin reorientation occurs in the course of which the easy axis of magnetization rotates continuously, with increasing temperature, within the $(1\bar{1}0)$ plane from the [110] towards the [111] direction. The Hamiltonian of the magnetic anisotropy of the heavy-rare-earth–iron Laves compounds has been expressed within a single J manifold.[13] For Sm^{3+} with $J=\frac{5}{2}$ as ground state, the use of a single J manifold precludes the existence of a [110] easy axis of magnetization. The mixing of the relatively low-lying excited J states in the ground state is of common occurrence in samarium compounds. Calculations involving the mixing of the $J=\frac{7}{2}$ and $\frac{9}{2}$ states into the ground state suggest the possibility of a low-temperature [110] easy axis of magnetization. In order to account for the presence of the [110] easy axis of magnetization in $SmFe_2$[13] at low temperatures and its rotation towards the [111] direction at higher temperature, a set of crystal-field parameters, inconsistent with those of the heavy-rare-earth–iron Laves compounds, is required. The existence of a wide transition region in $SmFe_2$,[13] in which the easy axis of magnetization points in directions other than the major axes of crystal symmetry, constitutes a further difficulty which cannot be resolved in the framework of the simple one-ion model.

The incompatibility of the one-ion model in its limited form with the magnetic behavior of $SmFe_2$ suggests that magnetoelastic effects should be considered. The objective of the present work was, therefore, to determine the behavior of the elastic properties and magnetostriction of $SmFe_2$ as a function of temperature and magnetic field, in order to evaluate the importance of magnetoelastic effects in this compound.

EXPERIMENTAL

$SmFe_2$ was prepared by arc-melting 99.9%-pure samarium and 99.99%-pure iron on a water-cooled copper hearth in an argon atmosphere. The arc-melted specimens contained several Sm-Fe intermetallic compounds.[14] An annealing treatment at 700 °C for three weeks produced an almost single-phased $SmFe_2$ sample, as confirmed by powder diffraction patterns taken with CoK_α

radiation. The lattice parameter was found to be 7.414 Å, in good agreement with the reported value.[15]

Specimens for ultrasonic measurements were flat and parallel disks 8 mm in diameter and about 4 mm thick. The parallelism of the sample faces was better than 2 parts in 10^4. The specimen thickness was determined to within $\pm 5\times10^{-4}$ mm. The longitudinal and transverse sound-wave velocities were measured by means of an ultrasonic pulse technique at a frequency of 10 MHz. The experimental details and method of data analysis were described elsewhere.[16] The room-temperature density was determined by means of a fluid-displacement technique using monobromobenzene. The measured density was within the experimental error (0.2%) of the calculated x-ray density. The temperature variation of the specimen density and acoustic path length due to thermal expansion was not taken into account in the calculation of the elastic moduli and were assumed to be negligible. Conventional cryogenic and magnetic (Varian 15-in. electromagnet) techniques were used. The maximum attainable magnetic field, in a gap of 5 cm, was 26 kOe. The sound velocities were determined during the slow warming, from liquid-helium temperature to the ambient, at a rate of 0.1 °K/min. The estimated experimental error in the determination of the elastic moduli is less than 0.4%, the relative values are better by a factor of 4. Magnetostriction measurements were performed by means of BLH (constantan foil) strain gauges.[17] A two-arm Wheatstone bridge was used in order to prevent erroneous effects on the measured values of magnetostriction due to temperature or magnetic fields. The specimen constituted one arm of the bridge and a dummy quartz specimen the other.

RESULTS AND DISCUSSION

The temperature dependence of the Young (E) and shear (G) polycrystalline elastic moduli of $SmFe_2$ at zero magnetic field and at 25 kOe is shown in Fig. 1. At zero field both E and G display prominent minima at 195 °K, the spin-rotation temperature T_{sr}. At temperatures above T_{sr} the easy direction of magnetization is [111], whereas below it the easy axis is directed along the [110] axis.[13] The change from the one easy direction of magnetization to the other extends over a wide temperature range suggesting a continuous, temperature-dependent process. The temperature variation of the elastic moduli at a field of 25 kOe (Fig. 1) illustrates this point. The character of the change in the elastic moduli at T_{sr} appears to be due to a first-order phase change, triggered by magnetoelastic effects. This implies that a distortion of the cubic symmetry below T_{sr} should be observed. The incompatibility of the one-ion model for $SmFe_2$ can, therefore, be explained by the presence of strong magnetoelastic effects which may lead to the occurrence of a first-order change at T_{sr}.

The elastic moduli E and G, at zero field, increase drastically by about 90% with decreasing temperature below T_{sr}. Such a behavior empha-

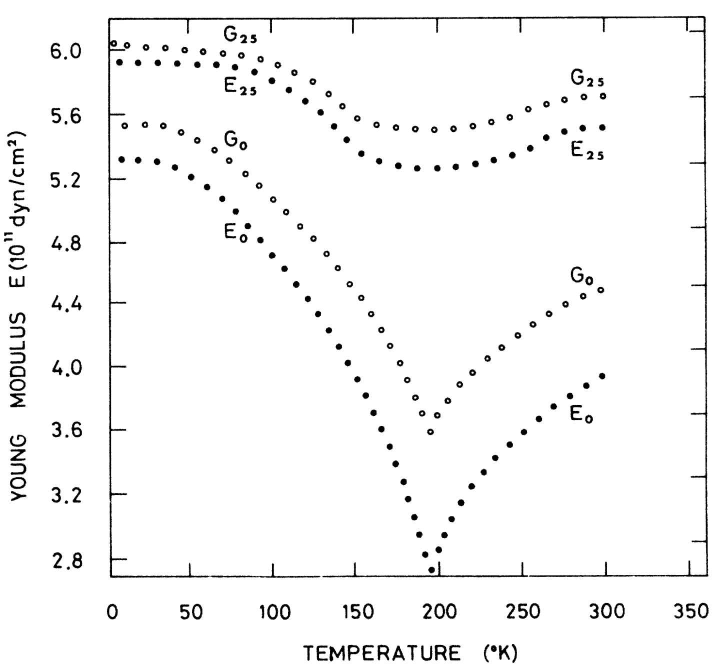

FIG. 1. Temperature dependence of the Young (E) and shear (G) moduli of $SmFe_2$, as a function of temperature at zero magnetic field and at 25 kOe.

sizes the large magnetoelastic contribution to the elastic stiffness of $SmFe_2$. Application of a magnetic field of 25 kOe, Fig. 1, does not produce saturation. Nevertheless, it diminishes and flattens out the minimum of the elastic moduli at T_{sr}. From T_{sr} down to liquid-helium temperature, the elastic moduli at a field of 25 kOe show a rather moderate increase, of about 10% only. Worthy of note is the magnetic field dependence of the absolute values of the elastic moduli over the whole temperature range investigated, as shown in Fig. 1. This *modulus change* will be discussed later.

The temperature dependence of the adiabatic compressibility K_s at zero field and at 25 kOe is displayed in Fig. 2. As expected, a sharp maximum of K_s at zero field indicates the spin-rotation temperature T_{sr}. The drastic decrease in the compressibility, at low temperatures, is in accord with the behavior of E and G (Fig. 1) in this temperature range. In contrast to the behavior of K_s at zero field, application of a magnetic field of 25 kOe causes a remarkable stiffening of the $SmFe_2$ lattice and yields an almost temperature-independent adiabatic compressibility, Fig. 2. This is an additional display of the magnetoelastic effects in $SmFe_2$.

The magnetic field dependence of the elastic moduli E of $SmFe_2$ at 77 and 300 K, is shown in Fig. 3. The observed variation of the elastic moduli as a function of magnetization is due to the "ΔE" effect.[18] ΔE is the difference between the magnetized and unmagnetized Young moduli, E_H and E_0, respectively. This magnetomechanical loss is associated with the vibration of the magnetic domain walls in a ferromagnetic material under the influence of a propagating high-frequency stress wave. The applied stress alters the local magnetization through the magnetostrictive coupling. The change in the Young modulus ΔE as a function of magnetization is generally frequency dependent, increasing with decreasing frequency.[19] In the present study, the frequency of the ultrasonic waves was 10 MHz. Figure 3 shows that the modulus change for the 300 °K isotherm and a field of 25 kOe is 42%. For both isotherms 77 and 300 °K saturation was not achieved at a field of 25 kOe. The observed value of 42% for the ΔE effect of $SmFe_2$ at 300 °K, 25 kOe, and 10-MHz ultrasonic frequency, is considered to be remarkably high. However, it is smaller than in the rare-earth–iron Laves phase compound $TbFe_2$ for which a value of 56% was observed under iden-

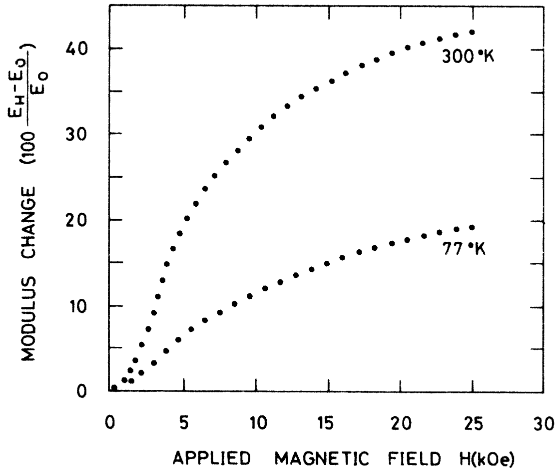

FIG. 3. Relative change in elastic modulus $(E_H-E_0)/E_0$ of $SmFe_2$ at 77 and 300 °K, as a function of applied magnetic field. E_H and E_0 are the magnetized and unmagnetized Young moduli, respectively.

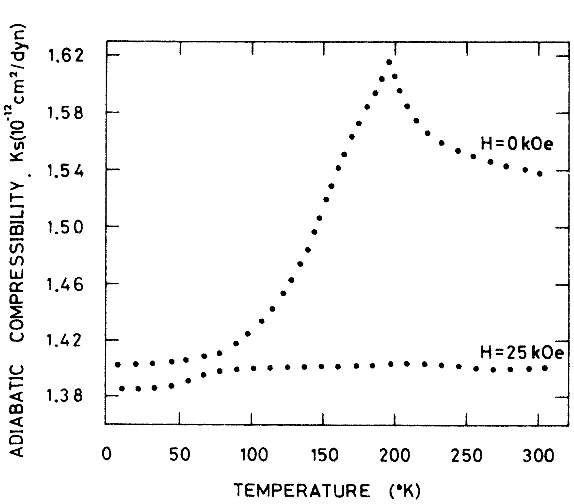

FIG. 2. Temperature dependence of the adiabatic compressibility (K_s) of $SmFe_2$, as a function of temperature at zero magnetic field and at 25 kOe.

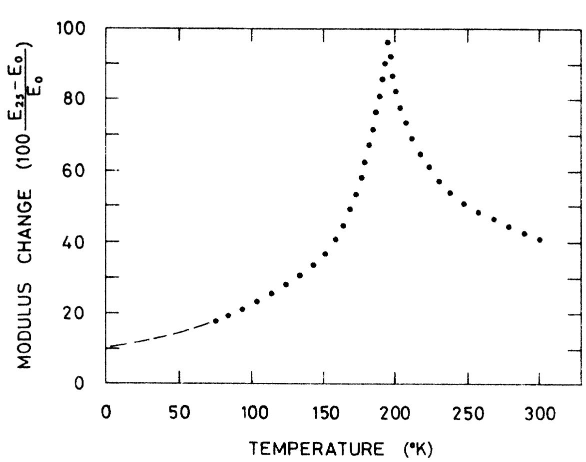

FIG. 4. Relative change in elastic modulus $(E_{25}-E_0)/E_0$ of $SmFe_2$, as a function of temperature. E_0 and E_{25} denote the Young moduli at zero magnetic field and at 25 kOe, respectively.

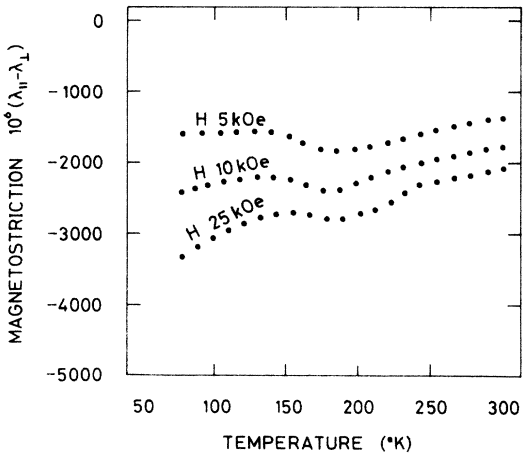

FIG. 5. Temperature and magnetic field dependence of magnetostriction in $SmFe_2$. $\lambda_\parallel$ and $\lambda_\perp$ are the magnetostrains parallel and perpendicular to the applied magnetic field direction, respectively.

tical conditions.[12] For both, $SmFe_2$ and $TbFe_2$, it is expected that the presence of high magnetoelastic effects lead to a loss of the perfect cubic symmetry.

Figure 4 shows the temperature dependence of the modulus change $(E_{25} - E_0)/E_0$, where E_{25} and E_0 are the Young moduli at a magnetic field of 25 kOe and zero, respectively. A modulus change of 97% is observed at the spin-rotation temperature T_{sr} of 195 K. This modulus change decreases at temperatures remote from T_{sr}. Such a behavior again emphasizes the presence of magnetoelastic effects in $SmFe_2$.

The rare-earth–iron Laves compounds possess extremely high room- temperature magnetostrictions. $TbFe_2$ was reported[20] to have a magnetostriction of 2630×10^{-6} at room temperature and at a magnetic field of 25 kOe. The temperature and field dependence of the magnetostriction in $SmFe_2$ is shown in Fig. 5. It is negative and has a value of -2100×10^{-6} at room temperature and 25 kOe. A flat minimum is displayed in the temperature dependence of the magnetostriction in the vicinity of T_{sr}. With decreasing temperature, for any magnetic field, the negative value of the magnetostriction increases. At 77 °K, and at a field of 25 kOe, the magnetostriction for $SmFe_2$ is -3340×10^{-6}. The magnetostrictive strains are generally related to spin correlation functions similar to those causing magnetic anisotropy.[21] It may therefore be possible that the origin of such large magnetostrictions in $SmFe_2$ is due to the large strain-dependent anisotropy of the rare-earth ions.

The Debye temperatures of $SmFe_2$ at the absolute zero (Θ_D^0) and at room temperature (Θ_D^{300}) were calculated from the measured longitudinal and transverse sound velocities near the respective temperatures, using well-known relations.[16,22] The following values were obtained:

$$\Theta_D^0(H=0) = 200\ °\mathrm{K}, \qquad \Theta_D^{300}(H=0) = 171\ °\mathrm{K},$$

$$\Theta_D^0(H=25\ \mathrm{kOe}) = 212\ °\mathrm{K}, \quad \Theta_D^{300}(H=25\ \mathrm{kOe}) = 207\ °\mathrm{K}.$$

CONCLUSIONS

(1) The rare-earth–iron Laves phase compound $SmFe_2$ displays a spin reorientation at T_{sr} (195 °K) from the high-temperature easy direction of magnetization [111] to the low-temperature easy axis directed along [110]. (2) The behavior of the elastic moduli in the vacinity of T_{sr} suggests a first-order phase change. (3) The elastic moduli of $SmFe_2$ increase significantly with applied magnetic fields, yielding a rather low and temperature-independent adiabatic compressibility. (4) A remarkably high value (42%) of the ΔE effect was observed in $SmFe_2$ at room temperature, 10-MHz ultrasonic frequency, and an applied field of 25 kOe. (5) $SmFe_2$ has a large negative magnetostriction. At a field of 25 kOe the magnetostriction values are -2100×10^{-6} and -3340×10^{-6}, for room temperature, and 77 °K, respectively. (6) The absolute zero Debye temperatures were found to be 200 and 212 °K, for the unmagnetized state and at 25 kOe, respectively.

ACKNOWLEDGMENTS

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