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Ultrasonic Study of Screw Spin Antiferromagnetic Domains in $Gd_{0.7}Y_{0.2}Lu_{0.1}$

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Ultrasonic attenuation and sound velocity have been measured for a single crystal of the $Gd_{0.7}Y_{0.2}Lu_{0.1}$ alloy over the temperature range 160–230 K which includes the ferromagnetic and screw spin antiferromagnetic states. The anomalies observed in attenuation and velocity, which are present only when the specimen is heated up from the ferromagnetic state, are attributed to screw spin antiferromagnetic domains. This view is also confirmed by thermal cycling and magnetic field cycling studies.

§ 1. Introduction

The rare earth metals and alloys display a wide range of magnetic spin structures, one of the most fascinating being the antiferromagnetic screw spin structure with moments confined to the basal plane of the hexagonal structure. The moments in each basal plane are ferromagnetically alined but the direction of magnetization rotates from plane to plane by a constant turn angle. The Gadolinium-based alloys provide a useful way of studying the stability of this antiferromagnetic phase and various ferromagnetic arrangements of the 4f moments, since these alloys exhibits both screw spin and ferromagnetic phases.[1–4] The $Gd_{0.7}Y_{0.2}Lu_{0.1}$ alloy has also an antiferromagnetic screw spin phase between the high temperature paramagnetic and low temperature ferromagnetic phases.[5]

Ultrasonic measurement has proved to be a very sensitive technique for the study of a magnetic phase transition in rare earth metals and alloys.[6–10] In the previous paper[11] we have made ultrasonic measurements together with magnetization measurements successfully for the determination of the magnetic phase diagram for a $Gd_{0.7}Y_{0.2}Lu_{0.1}$ single crystal. In ref.11, anomalous peak in attenuation and anomalous drop in velocity, which are observed only when the specimen is heated up from the ferromagnetic state, are tentatively explained in terms of antiferromagnetic domains. In the present paper we report further experiments on the effects of thermal cycling and magnetic field cycling on the anomalous behaviour in ultrasonic propagation, which allow a more definite conclusion to be drawn on the possible screw spin antiferromagnetic domains.

§ 2. Experimental Details

The $Gd_{0.7}Y_{0.2}Lu_{0.1}$ alloy was prepared at the Ames Laboratory, Iowa State University, by arc-melting the appropriate weights of the elements and the single crystal was grown by a thermal annealing procedure. The surfaces of both sides of the specimen perpendicular to the hexagonal *c*-axis were polished in the parallel planes to the quality usually required for ultrasonic experiments. The final length of a rectangular specimen in the *c*-, *a*- and *b*-direction were 3.688, 5.709 and 4.165 mm, respectively.

Ultrasonic attenuation and sound velocity were measured by propagating longitudinal waves at a frequency of 10 MHz generated by *X*-cut quartz transducers along the hexagonal *c*-axis of the specimen. Nonaq stopcoch grease was used to maintain acoustic coupling between the transducer and specimen. Figure 1 shows the block diagram of our attenuation and velocity measurements. The pulsed oscillator (Matec Model 6600 with Model 760 V plug-in unit) is triggered by the cw output signal (frequency $f \simeq 400$ kHz) of the high resolution frequency source (Matec Model 110). The pulse repetition rate is varied by the frequency of the source and the divider. The width of the RF pulse is about 1μ sec and its amplitude is varied by means of the step attenuator by 0.5 dB step. The sound signals that have traversed the specimen are amplified and displayed on the oscilloscope. The amplitude of the signal is measured on the oscilloscope and converted to attenuation in dB. The sound velocity and its change are determined by the pulse-echo-overlap method in which the reference RF pulse of the pulsed oscillator and the sound signal are strobed by the

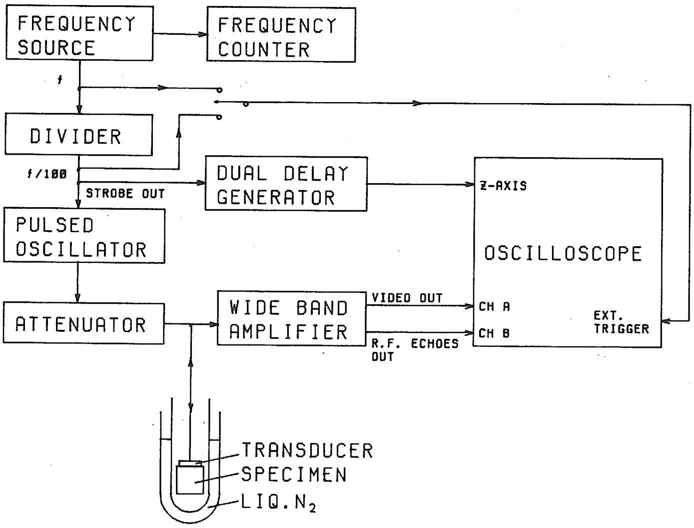

Fig. 1. Block diagram for the measurements of ultrasonic attenuation and sound velocity.

dual delay generator and overlaped on the oscilloscope by choosing the CW frequency f in such a way that f just coincides with τ^{-1}, where τ is the time of flight of the sound through the specimen. The resolution of the change in velocity in this system is 10^{-5}.

§ 3. Experimental Results

The temperature dependence of the ultrasonic attenuation α is shown in Fig. 2. There is a marked difference between the results measured on warming and on cooling. In the case of the warming-up process, two obvious attenuation peaks appear. One is rather broad peak near 180 K with an accompanying shoulder on the high temperature side. The other is a small but sharp peak at 211 K. On the contrary in the case of cooling-down process, the anomaly near 180 K is strongly suppressed.

The temperature of 211 K at which a sharp peak in α appears is just in agreement with the temperature at which low field magnetization curves represent a sharp peak.[11] Therefore, this temperature is considered to be the Néel temperature T_N . This sharp peak in α may be due to the critical fluctuation of spins near the phase transition between paramagnetic and antiferromagnetic states.

The temperature of 180K where a broad peak appears in the α vs T curve measured on warming corresponds to the temperature where the magnetization as a function of temperature almost saturates.[11] Therefore, this temperature is reasonably chosen as the Curie temperature T_f, below which magnetic moments orders ferromagnetically. In the region between T_f and T_N, the specimen shows an

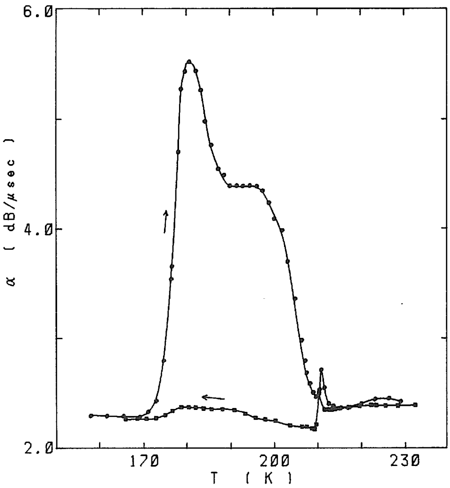

Fig. 2. Temperature dependenece of the ultrasonic attenuation α. The arrows indicate the direction of temperature change.

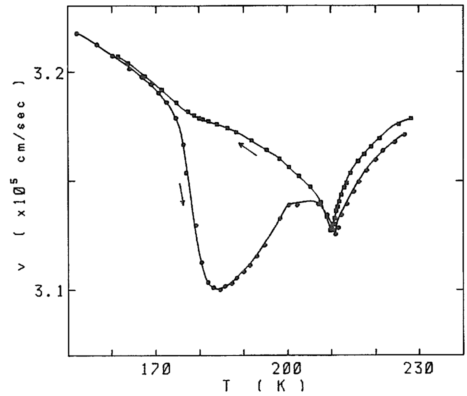

Fig. 3. Temperature dependence of the sound velocity v. The arrows indicate the direction of temperature change.

antiferromagnetic state with the screw spin structure.[5]

The temperature dependence of the sound velocity v is shown in Fig. 3. There is also a marked difference between the results measured on warming and cooling. In the case of the warming-up process, two anomalies appear as the drops in velocity at the corresponding temperatures of the attenuation peak. One is a broad drop near T_f and the other is a sharp drop at T_N. In the case of the cooling down process, the anomaly near T_f is very small just as the anomaly in α measured on cooling. But near this temperature a noticeable change in gradient is observed.

Further experiments are described on the effect of thermal cycling in order to clarify the nature of the anomalous attenuation near T_f observed in the warming-up process. The specimen is first cooled below T_f into the ferromagnetic phase and then warmed into the screw spin antiferromagnetic phase. The warming of the specimen is then halted at three different temperatures, 190 K, 200 K and 208 K, and the specimen is cooled below T_f while the behaviour of α is monitored. The results are shown in Fig.4. In this case the attenuation peak near T_f appears even in the cooling-down process. With raising the temperature from which the specimen is started cooling, the anomalous attenuation becomes smaller.

Next, the specimen is cooled from the paramagnetic phase into the screw spin antiferromagnetic phase. The cooling of the specimen is halted at two different temperatures, 207 K and 190 K, where a magnetic field is applied in the easy-basal-plane direction and subsequently reduced to zero. The applied field of 6.5 kOe is

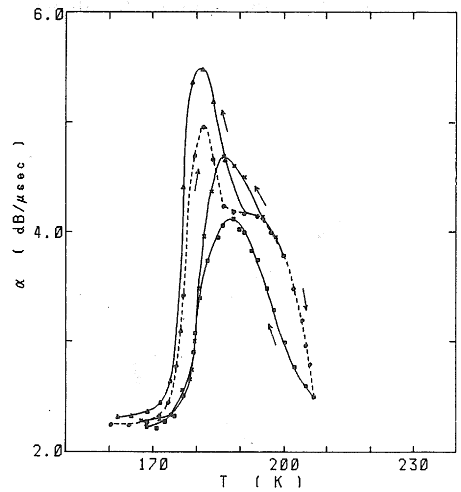

Fig. 4. The effect of thermal cycling on the ultrasonic attenuation α. The warming of the specimen is halted at three different temperatures, 190 K, 200 K and 208 K, and then the specimen is cooled down.

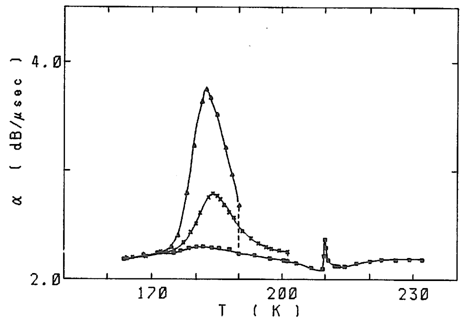

Fig. 5. The effect of field cycling on the ultrasonic attenuation α. The cooling of the specimen is halted at two different temperatures, 201 K and 190 K, where the magnetic field of 6.5 kOe is applied and subsequently reduced to zero. Then the specimen is cooled down.

adequate to produce ferromagnetism.[11] Then the specimen is cooled below T_f while the behaviour of α is monitored. The results are shown in Fig. 5. The anomalous attenuation peak near T_f appears in the cooling-down process for the specimen which experiences the field-induced ferromagnetic state.

§4. Discussion

The anomalous increase in α near T_f described above can be attributed to the magnetic domains in the screw spin structure. Domains which could interact with longitudinal waves propagating along the c-axis are the one where the domain wall is perpendicular to the c-axis and the two domains are distinguished by a change in sense of the spiral. Logitudinal waves propagating along the c-axis produce the periodical expansion and contraction of the spacing between the lattice planes, which varies periodically the exchange interaction between the spins lying in the different

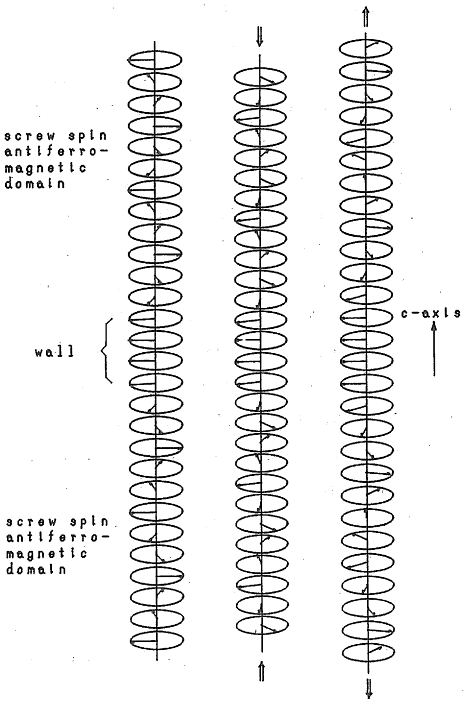

Fig. 6. Schematic model illustrating the reorientation of the spins due to the expansion and contraction of the spacing between basal planes produced by the longitudinal waves propagating along the c-axis.

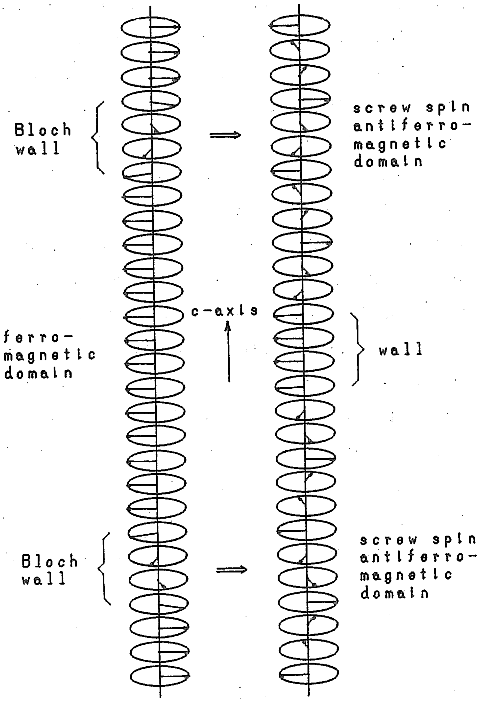

Fig. 7. Schematic model illustrating the formation of screw spin antiferromagnetic domains from the ferromagnetic domain walls (Bloch walls).

basal planes. Owing to this change of the interaction, the spins rotate around the c-axis to reach a new stable state with a different screw spin turn angle in the distorted configulation. For spins lying far from the domain wall, the energy required to rotate the spins is thought to be very small because the anisotropy within the basal plane is very small. However inside and just outside of the wall, there exists a large friction for the spin rotation because of the presence of pinned spins in the wall, which probably leads to large energy absorption and hence large attenuation of the acoustic wave. This behavior is illustrated in Fig. 6.

It is natural to consider that an anomalous drop in v measured on warming is also related to the relaxation of the spins inside and just outside of the domain wall. This spin rotation changes locally the magnetization near the domain wall, which may produce a magnetostrictive strain and may decrease the elastic constant.

The large difference between the results measured on warming and on cooling can be explained by considering that there are always more screw spin antiferromagnetic domains present in the crystal warmed from below T_f than those present cooled from above T_N. Domains will be present when the specimen is cooled down into the antiferromagnetic state, and the walls are expected to be pinned to pre-existing impurities and imperfections in the crystal. However they will be far less numerous than ferromagnetic domains present in the ferromagnetic state because the energy gain by forming the domains is small in the screw spin structure. When the specimen is warmed from below T_f further domains could be nucleated from the domains already present in the ferromagnetic phase. The model of the formation of screw spin antiferromagnetic domains is illustrated in Fig. 7. Assuming that screw spin structures grow from Bloch walls with opposite sense of spiral in the ferromagnetic state, the antiferromagnetic domain walls are inevitably produced.

The screw spin antiferromagnetic domain structure, which is formed under the influence of the ferromagnetic domains, will be most stable just above T_f and hence the friction arising from the domain wall will be most strong at this temperature. As the temperature is increased from T_f, the wall begins to be unstable and the spins within the wall become easy to rotate. As a result, the wall breaks with increasing temperature and the number of domains decreases. This is in keeping with the experimental evidence that the maximum of attenuation peak appears near T_f and the anomaly becomes smaller with raising the temepratu re from which the specimen is started cooling in Fig. 4. The appearance of attenuation peak on the cooling down process in Fig. 4 results from the stabilization of the screw spin antiferromagnetic domains with decreasing temperature in the antiferromagnetic region. In this case the number of the domains will remain essentially constant.

The result in Fig. 5 also supports that screw spin antiferromagnetic domains are produced by nucleation from the ferromagnetic domains. In this case the ferromagnetism is field induced in the antiferromagnetic region. The rise in α, which is induced by the application of the easy direction magnetic field and subsequent reduction in the field, is expected to be proportional to the number of domains produced by this field cycling.

References

1) W. C. Thoburn, S. Legvold and F. H. Spedding : Phys. Rev. **110** (1958) 1298.
2) T. Ito : J. Sci. Hiroshima Univ., Ser.A **37** (1973) 107.
3) D. M. S. Bagguley, J. Leisegang and K. Robinson : J. Phys. F : Metal Phys. **4** (1974) 594.
4) S. Legvold, T. Ito and B. J. Beaudry : Phys. Rev. Lett. **45** (1980) 1275 ; T. Ito, S. Legvold and B. J. Beaudry : Phys. Rev. B**23** (1981) 3409.
5) T. Ito, M. Oka, S. Legvold and B. J. Beaudry : Phys. Rev. B**29** (1984) 6276.
6) S. Maekawa, R. A. Treder, M. Tachiki, M. C. Lee and M. Levy : Phys. Rev. B **13**(1976) 1284.
7) C. Isci and S. B. Palmer : J. Phys. F: Metal Phys. **8** (1978) 247.
8) M. C. Lee and M. Levy : J. Phys. Chem. Solids **34** (1973) 995.
9) S. B. Palmer, D. Hukin and C. Isci : J. Phys. F : Metal Phys. **7** (1977) 2381.
10) J. B. Sousa, R. P. Pinto, M. F. Pinheiro, M. E. Braga, J. M. Moreira, G. N. Blackie, S. B. Palmer and D. Hukin : J. Magn. & Magn. Mater.**28** (1982) 175.
11) J. Takeuchi, T. Ito and M. Oka : J. Phys. Soc. Jpn.**54** (1985) 3076.