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Measuring magnetostriction with neutrons

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Abstract
Neutron diffraction has been used to measure magnetostriction and thermal expansion in an unusual way. The three-axis spectrometers IN12 and IN3 at the ILL, Grenoble, France, were used to determine the change of lattice constants with temperature and applied magnetic field by measuring the angular shift of selected Bragg reflections. The instrumental resolution of this method is hardly comparable with x-ray diffraction or dilatometric methods. Nevertheless, using neutrons allows us to overcome certain experimental difficulties encountered when using more conventional methods; capacitance dilatometry is sensitive to parasitic forces occurring when applying a magnetic field on highly anisotropic samples. Since the penetration depth of x-rays is very small, slight movements of the sample change the scattering geometry and introduce an error, which is difficult to estimate. Here we discuss two distinct cases for which neutrons helped to explain the apparent discrepancy in previously measured magnetostriction and thermal expansion data.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

If a magnetic field is applied to a magnetic material the shape of the material changes. This effect is known as magnetostriction and is used in technical applications such as generators of ultrasonic waves or high precision positioning devices. Most magnetostrictive materials for applications are based on rare earth compounds. They became available as pure specimens in the late 1960s and since then their magnetic properties have been investigated in detail by many experimental methods. Within the standard model [1], the deformation of the 4f charge density induced by the crystal electric field allows for magnetostrictive effects. A number of techniques are available to measure magnetostriction, which has become an important property for the characterization of magnetic materials. Table 1 represents the sensitivity of different experimental methods for thermal expansion and magnetostriction measurements.

Hereafter we will discuss three methods: dilatometry, x-ray diffraction and neutron diffraction.

Table 1. Different methods for measuring thermal expansion and magnetostriction.

Method	Sensitivity (dl/l)	External parameters
Neutron diffraction	10^{-4}	0.01–600 K, 0–15 T
EXAFS at ESRF	10^{-4}	300 K, 0–2 T
X-ray diffraction	10^{-5}	1.5–2000 K, 0–7 T
Strain gages	10^{-7}	1.5–700 K, 0–30 T
Interferometry	10^{-8}	4–2000 K, 0 T
Capacitive dilatometers	10^{-9}	0.01–1000 K, 0–40 T

1.1. Capacitance dilatometry

The capacitance method is one of the most sensitive methods for measuring small macroscopic changes in solids. Slight changes of sample size cause the capacitance plates to alter their orientation, e.g. the capacitance. Modern three-terminal capacitance measurement bridges have high resolution, providing relative length change measurements with precision up to 10^{-9} in static fields up to 45 T and in pulse fields up to 60 T. However, highly anisotropic samples

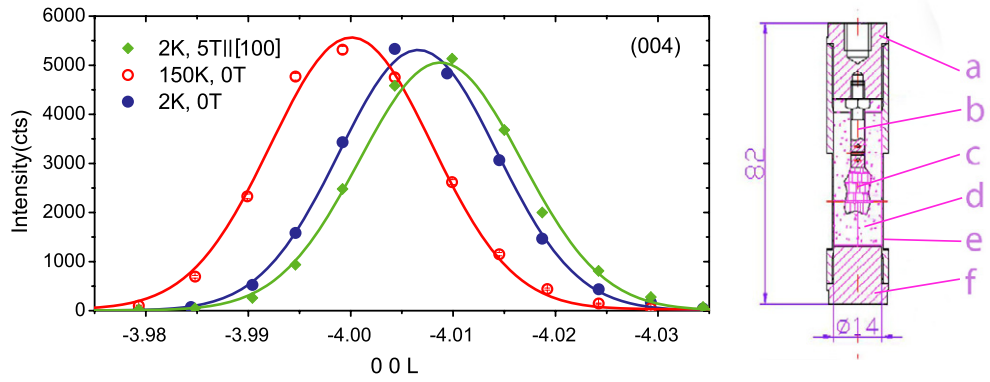

Figure 1. Relative shift of (004) reflection (on the left) as a function of temperature and magnetic field measured on the sub-thermal neutron three-axis spectrometer IN3 in a special sample holder (on the right): (a) upper cap; (b) inner sample rod; (c) sample; (d) aluminum powder; (e) aluminum tube; (f) bottom cap. Lines show results of a Gauss fit of the (004) reflection. Error bars are smaller than symbols.

can bend/rotate in applied magnetic field leading to a parasitic signal, which is difficult to quantify.

1.2. X-ray diffraction

X-ray diffraction is a commonly used method for measuring thermal expansion and performing structural investigations in solids. As with any other diffraction technique, x-rays provide microscopic information. The lattice parameters are determined by refining the diffraction pattern obtained in different temperature/field conditions. Modern x-ray diffractometers with a magnetic field option can perform measurements in fields up to 7 T. Within the x-ray penetration depth limitation, the scattering volume is small. Slight movements of the sample may change the scattering geometry and introduce an error, which is difficult to quantify.

1.3. Neutron diffraction

Neutron diffraction is also a microscopic method. The lattice constants are refined from the diffraction pattern. Due to positioning errors of the instrument as well as relatively big sample size the resulting resolution in dl/l is 10^{-4}. By filling a sample holder with high purity aluminum powder, the sample can be fixed inside the cryomagnet to insure that no field induced rotation occurs [2] (see figure 1). However, a small movement of the sample in the beam will not change the experimentally measured Bragg angle, as long as the sample remains in the beam. Therefore neutron diffraction provides a measurement free of the errors mentioned for the other methods.

Such neutron diffraction methods helped us to resolve certain discrepancies in the following two cases.

2. $PrCu_2$

$PrCu_2$ has a $CeCu_2$ type orthorhombic structure with space group *Imma* above 8.5 K ($a = 0.4408$ nm, $b = 0.7055$ nm, $c = 0.7444$ nm). Below 8.5 K the the lattice becomes monoclinic with space group $I2/m$ ($a = 0.4416$ nm, $b = 0.7405$ nm, $c = 0.7405$ nm, $\beta = 90.503°$) [3]. The reported crystallographic structure change is believed to be a result of a collective Jahn–Teller transition.

Some contradictory results in thermal expansion have been reported in the literature [4, 5]. In order to clarify the situation, multiple measurements of thermal expansion and magnetostriction have been performed at the sub-thermal neutron three-axis spectrometer IN3, ILL, Grenoble. The experiments were performed at a wavelength of $\lambda = 0.236$ nm selected by a Cu(111) monochromator using 30′ collimation before the sample. We used a PG-filter in the incident beam. A PG(002) analyzer was used with 40′ collimation between sample and analyzer and also between analyzer and ^{3}He detector. A vertical cryomagnet of 5 T was used to change the magnetic field and temperature. Thermal expansion and magnetostriction were deduced from the relative change of the fitted peak position of (040) and (00$\bar{4}$) reflections with temperature and field, respectively (see figure 1). The sample rotation has been monitored by realigning the sample to a corresponding reflection for each field step.

We measured the thermal expansion in the temperature region of 2–160 K and the magnetostriction up to 5 T. At high temperatures the thermal dependence follows the x-ray measurements [6]. Recent inelastic scattering investigations on IN14 [7] using the same sample indicate the existence of orbital excitations at low temperature, which were predicted within the model described in [8]. Any lattice parameter change related to quadrupolar order is hardly visible within the instrumental resolution in our single crystal (see figure 2). Lattice parameter measurements as a function of field (see figure 3) indicate a smooth negative slope at low temperature. Within the instrumental resolution no signature of any transition has been observed, contrary to previous reports [9] for field applied along the *b* direction.

3. Thulium

Thulium metal has a hexagonal structure with space group $P6_3/mcm$ ($a = 0.3534$ nm, $b = 0.3534$ nm, $c = 0.5573$ nm). It orders antiferromagnetically below 56 K. The magnetic structure is amplitude modulated along the *c*-axis. Near 42 K

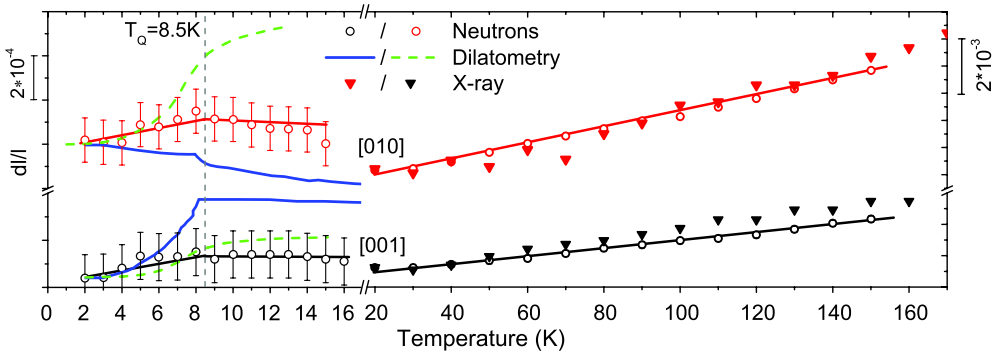

Figure 2. Thermal expansion of $PrCu_2$ measured with neutrons along [010] and [001] directions, plotted versus corresponding x-ray and dilatometric measurements. Dilatometric data are represented by a dashed line [4] or a solid line [5]. X-ray data are shown by triangles [6]. Open circles represent neutron measurements accompanied by linear fit. The vertical dashed line indicates the quadrupolar transition temperature. Errors bars are smaller than the symbols or line width.

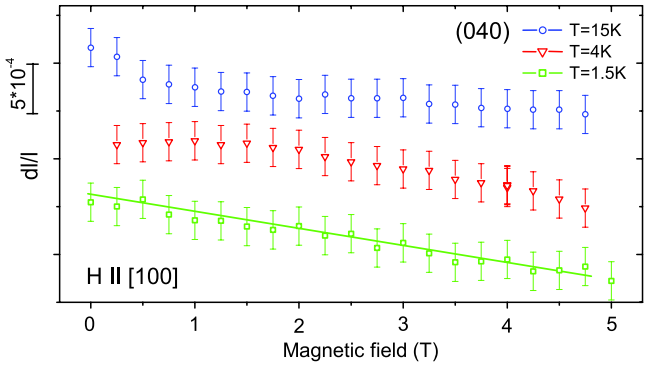

Figure 3. $PrCu_2$ magnetostriction measured with neutrons for different temperatures with field parallel to [100]. Squares, triangles, and circles represent magnetostrictive responses for temperatures of 1.5, 4, and 15 K, respectively. The line represents a linear interpolation.

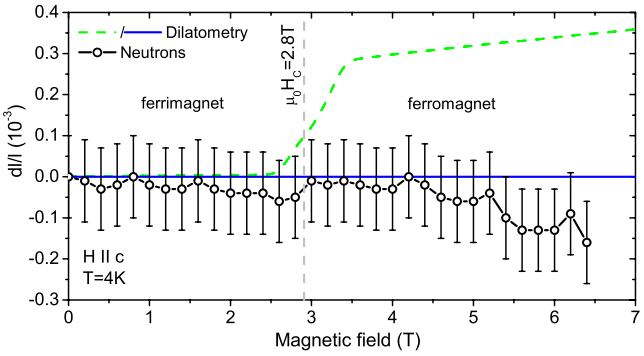

Figure 4. Thulium magnetostriction (black circles) measured with neutrons plotted versus dilatometric data represented by a dashed line [14] and solid line. The vertical dashed line represents the transition field between ferrimagnetic and ferromagnetic states. Error bars are smaller than the line width.

the structure starts to square up and shows a lock-in transition at 32 K to ferrimagnetic order with seven layers of periodicity (four layers up, three layers down) [10, 11]. Thulium is known to have high magnetic anisotropy [12].

Magnetostriction measurements with the capacitance method published in 1992 by Zhochowski *et al* [13] showed giant magnetostriction of about 10^{-2} in dl/l at a field of about 3 T. Barcza *et al* reported [14] nearly an order of magnitude less magnetostriction signal. Numerous attempts to reproduce this giant lattice change led to a large amount of inconsistent data. The relative length change varied in the range of 10^{-3}–10^{-7} each time the sample was remounted into the dilatometer.

In order to resolve this large discrepancy an attempt to measure magnetostriction has been made at the cold neutron three-axis spectrometer IN12, ILL, Grenoble. Vertical (12 T) and horizontal (4 T) magnets were used to change the magnetic field. Magnetostriction data were deduced from the relative change of the fitted peak position of $(\bar{1}00)$ and (010) reflections. The sample rotation has been monitored by realigning the instrument to a corresponding reflection for each field step. The experiments were performed at a wavelength of $\lambda = 0.418$ nm selected by a Cu(111) monochromator using 30′ collimation before the sample. We used a Be-filter in the incident beam. A PG(002) analyzer was used with 30′ collimation between sample and analyzer.

As expected, above a field of 2.8 T the peak amplitude increased significantly due to induced ferromagnetic order. Within the instrumental resolution the fitted peak position shows a smooth tendency to actual decrease, but no discontinuity at the critical field of 2.8 T is observed (see figure 4).

4. Conclusions

For two examples, we showed how to resolve apparent discrepancies of magnetostriction data with neutron scattering.

The thermal expansion of $PrCu_2$ is positive over a wide temperature range, in agreement with x-ray data. In the low temperature region a smooth variation has been observed. Quadrupolar features are hardly visible at low temperatures. This indicates that the quadrupolar order is not as strongly coupled to the lattice as assumed in the literature.

The magnetostriction of thulium shows a meta-magnetic transition at 2.8 T. It is visible from a peak intensity increase

for fields above 2.8 T due to the magnetic scattering. The large magnetostriction features measured with dilatometry cannot be reproduced by neutrons.

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