Magnetostriction of 4f-electron compounds in high magnetic fields

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Abstract. Magnetostriction gives an insight into the interactions between the electronic and the lattice system of solids. Because only macroscopic methods can be used in fields above 20 T, miniaturized capacitive dilatometers were adapted to the strongest magnets. We performed experiments up to the highest available steady fields of 45 T and in 50 T pulsed field systems. The power of magnetoelastic investigations is illustrated by measurements at two 4f-intermetallics: SmCu₂ is an antiferromagnet below 23 K with a nearly compensated magnetic moment and, the monopnictid GdSb orders antiferromagnetically at 24 K. Both materials show magnetic transitions at applied fields of about 30 T when the ferromagnetic state is induced.

1. Introduction

The magnetostriction, i.e. the change of shape or volume caused by a magnetic field, is a general effect in solid state physics because any magnetization is directly connected to a lattice deformation. Magnetoelastic investigations gain an insight into the magnetization process, the domain dynamics and the magnetic and crystalline anisotropy. Current theories to model magnetostriction take into account the crystal electric field and the exchange striction. Moreover, magnetostriction can be calculated by ab initio electron structure theories. From the view of applications magnetostriction is important in magnetoacoustic devices, magnetomechanical actuators (e.g. magnetic shape memory materials) and others. Sometimes, a magnetostrictive mismatch is a problem in thin film technology.

In spite of all these points, measurements of magnetostriction are not widely established in high magnetic fields. In principle, they can be done by microscopic (x-rays or neutrons, sensitivity $10^{-5}$) or macroscopic (e.g. capacitive dilatometers: sensitivity $10^{-9}$, resistive extensometers: sensitivity $10^{-7}$) methods [1,2]. However, scattering techniques in high fields above 20 T are still underdeveloped. Recently, a lot of effort has been invested to adapt capacitive dilatometers to the strongest magnets ([3,4] for steady fields, [5-7] for pulsed fields for example). We focused our activities on developments of microdilatometers, for steady as well as for pulsed magnetic fields. With these devices the magnetoelasticity of f-electron antiferromagnets with a large variety of magnetic structures was studied.

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2. Experimental techniques
In steady fields we used a tilted plate capacitive dilatometer (\(\varnothing=20\) mm, \(h=20\) mm, material: silver) [8] (Fig.1). The field range was extended to the maximum available field of 45 T for the first time using the hybrid magnet at the NHMFL Tallahassee. The compact capacitor design allows one to turn the capacitor, so that the longitudinal as well as the transversal components of the strain tensor can be determined. The capacitance was measured with a self-balancing AH2500 bridge in tracking mode at 1 kHz. A \(\Delta l/l\) resolution in the order of \(10^{-7}\) was achieved by damping of the vibrations caused by the cooling water flow in the hybrid. Moreover, avoiding mechanical contacts between dilatometer and tube was necessary.

![Figure 1. Dilatometer for steady fields, tested up to 45 T (\(\varnothing=20\) mm, tilted plate silver capacitor [8] right).](image1)

![Figure 2. Dilatometer for pulsed fields, tested up to 50 T (\(\varnothing=10\) mm, brass plate capacitor right inside the plastic housing).](image2)

More complicated is the adaptation of capacitive dilatometers to pulsed high fields. Several precautions owing to mechanical and electrical noises generated by the pulse and a significant reduction of eddy current heating of the sample and the dilatometer are necessary. The last problem may result in thermal expansion effects often higher than the magnetostriction. A possible solution is the use of non-metallic dilatometers (for instance plastic [6] or quartz-glass). We used a hollow cylinder (\(\varnothing=10\) mm, \(h=15\) mm) made from GFR epoxy or PEEK as dilatometer body (Fig.2). In the mounting procedure two pistons are pressed into this body from both ends and glued with (non-magnetostrictive) GE varnish, one of which containing the fixed capacitor plate and the other one the sample and the movable plate. Both plates are thin brass disks (\(\varnothing=5\) mm, \(h=0.5\) mm) with radial slits. To make the plates parallel, a thin plastic foil is inserted between the plates while mounting which after removal gives the basic capacitance (1 pF - 5 pF). The equipment for capacitance measurements contains a GR bridge 1615A excited with 40 V AC voltage and a Yokogawa DSO DL750 (16 bit ADC, 1 MS/s, 100 kpoints) for fast data storage. The analysis of the bridge detuning voltage is done by a Lock-in computer program [9]. A C(V) calibration curve gives the capacitance. The frequency has to be optimized in the range 10 kHz - 50 kHz, above 200 kHz parasitic capacities prohibit experiments. All measurements were done in the 50 T, 10 ms pulsed magnet of the Hochfeldlabor Dresden (HLD) [10] located at IFW Dresden. A resolution of about \(10^{-5}\), nearly constant over the full field range, could be reached up to now. An improvement seems possible by optimized fixing and shielding of the wires.

3. Results
The 1st example is intermetallic \(\text{SmCu}_2\), that is a good candidate to detect metamagnetic transitions in high fields [11]. It crystallizes in an orthorhombic structure with antiferromagnetic
order below 23 K. Below $T_N$ commensurate or incommensurate magnetic phases (AF1, AF2, AF3, LT) exist. The saturation moment is only $0.5 \mu_B$/Sm-ion because of partial compensation of spin and orbital momentum [12]. Because of the small moment the critical transition field to induce the ferromagnetic state is rather high (25 T) [13]. The steady field magnetostriction at 4 K [11] shows significant changes of the sample length, e.g. $\varepsilon^b = -5 \times 10^{-4}$ (inset Fig. 3). The volume effect is small. Nevertheless, a distortion in the $ac$ plane can be seen.

![Figure 3](image1.png)  
**Figure 3.** Longitudinal magnetostriction $\varepsilon^b$ of SmCu$_2$ measured in 50 T, 10 ms pulsed field at different temperatures. The inset shows the steady field data measured at 4 K for comparison.

![Figure 4](image2.png)  
**Figure 4.** Phase diagram of SmCu$_2$. The transitions are detected by pulsed field magnetostriction (fullcircles = increasing, open-circles = decreasing field) and by magnetization, resistivity, specific heat (triangles).

The pulsed field results along the easy $b$-direction at different temperatures (Fig. 3) agree with these findings. The jump in length is $\varepsilon^b = -4 \times 10^{-4}$ at low temperatures and 27 T. This field is slightly higher than in steady fields. The magnetostriction has a small hysteresis of about 1 T. It is not temperature dependent and not visible in steady fields. Therefore, the hysteresis is caused more by the mechanical inertness of the dilatometer than by intrinsic sample properties. Moreover, the sharp transitions allows us to verify the magnetic phase diagram of SmCu$_2$ [13] constructed from magnetization. The high field transition line is confirmed completely (Fig. 4).

The 2nd investigated compound is the monopnictid GdSb that orders antiferromagnetically at 24 K. Because Gd$^{3+}$ is a pure spin system with $L=0$ there is no crystal field magnetostriction. This and the (cubic) high lattice symmetry makes it a good candidate to calculate the change of volume and shape at the afm/fm transition by first-principle band structure calculations. GdSb shows the "magnetoelastic paradox" (MEP [14]). This is the absence of a lattice distortion at $T_N$ in contrast to the expectations of the exchange striction model. Applying an external field parallel to the $[100]$ (which is away from the [111] propagation direction) the magnetization increases monotonously up to the saturation at 33 T [15]. Our longitudinal and transversal measurements in steady fields (Fig. 5) show a large change in length over a wide field range between 20 T and 35 T, approximately. The absolute strain at 1.5 K is about $\varepsilon^a = -3.2 \times 10^{-4}$, $\varepsilon^b = 1.8 \times 10^{-4}$ resulting in a negligible volume effect of $(\varepsilon^a + 2\varepsilon^b)/3 = 1.3 \times 10^{-5}$. The behaviour can be explained by the continuous decrease of the rhombohedral distortion in the $ab$ plane which should vanish in the ferromagnetic state. This point is visible by a kink at $(33\pm2)$ T.
Related to the MEP, from magnetoelastic effects of about $10^{-4}$ a peak broadening of $2 \cdot 10^{-2}$ in the x-ray scattering angle can be estimated. X-ray diffractometers are able to resolve this and to confirm or confute the MEP in the example of GdSb.

![Figure 5. Longitudinal $\varepsilon^a$ and transversal $\varepsilon^b$ magnetostriction of GdSb measured in the 45 T hybrid at 1.5 K and 4 K.](image)

### 4. Summary
The data confirms the high potential of magnetoelastic investigations by capacitive dilatometry also at the highest available magnetic fields. The method is complementary to other experimental methods as magnetization and ultrasound. In pulsed fields a resolution less than $10^{-5}$ is reached. For example, by magnetostriction measurements fundamental interactions between electronic and crystallographic properties can be seen as represented in the stability range of magnetic phases in SmCu$_2$ and the lattice distortion effects in GdSb.

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