Methods of dilatometric investigations under extreme conditions and the case of spin-ice compounds

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Abstract. We give an overview on how dilatometric methods have been developed in the last decade. The concept of capacitive dilatometry was successfully adapted to dilution refrigerators with a resolution of 10⁻⁹. Miniaturized dilatometers with an overall diameter of 18 mm or less are optimally suited for measuring longitudinal and transversal components of the striction tensor. Going to another extreme, to the highest (pulsed) fields, optical methods, such as the FBG technology, were developed for investigations up to 100 T.

As examples for utilizing dilatometry at low temperatures we show results for the spin-ice materials Dy₂Ti₂O₇ and Ho₂Ti₂O₇. To characterise the magneto-elastic coupling in these materials, we investigated the thermal expansion and magnetostriction between 80 mK and 15 K and in magnetic fields aligned along the [111] direction and found field-induced phases and strong correlations below 500 mK. Our data demonstrate, that the formation of the field-induced phase is strongly influenced by lattice distortions: any change in interatomic distances will result in a variation of the exchange couplings.

1. Introduction

Dilatometric investigations, i.e. measurements of the changes in sample dimensions or crystal symmetries, can shed light on how the lattice couples to electronic degrees of freedom. Another aspect is the connection to the elastic properties such as elastic moduli etc. Besides the common thermal expansion, the effects in magnetic fields (molecular fields or external ones) shifted into the scope of research and application. Basics, physical models and experimental results of magnetostriction were published in review articles, e.g. [1].

Magnetic materials change their lattice parameters when exposed to a magnetic field, but the effects can even beyond the experimental resolution. This requires to improve the sensitivity of existing methods. Nowadays, length variations in the sub-Å range can be resolved. Additionally, the recent interest in quantum criticality and magnetic frustration has made it necessary to extend the temperature range to the millikelvin region. On the other hand, measurements in fields up to 100 T can be utilized now to overcome exchange forces and to study magnetoelastic interactions.

A focus of current research in solid-state physics is on highly frustrated magnetism, mainly driven by competing interactions in a geometrically frustrated lattice. Long-range magnetic
order is suppressed and new ground states and exotic excitations, as for example magnetic monopoles [2, 3], are studied. Prominent examples are pyrochlores $R_2M_2O_7$ ($R =$ rare earth; $M =$ Ti,Zr,Hf,Sn). In these materials, highly degenerated spin-ice or quantum spin-ice ground states exist and new kinds of excitations emerge in magnetic fields and in the millikelvin range [4]. In this context, magnetostriction is an important analytical method because it detects lattice expansions or distortions which are strongly related to the magnetic exchange couplings. Simulation of the magnetostrictive behaviour, e.g. with the program package McPhase, may help to elucidate if the lattice is affected either by crystal-field or exchange striction.

2. Measurement techniques

Magnetostriction of three-dimensional solids (bulk) can be measured, in principle, either by A) electromechanical or B) optical methods. Method A) comprises the commonly used capacitive dilatometers and the resistive strain-gauges whereas in method B) interferometry, lately also using fibre-bragg gratings (FBG, see below) and optical transmission are included.

To extend the measurements to millikelvin temperatures, the existing capacitive dilatometers, where the sample is connected to a flexible plate of a capacitor, are mounted into dilution refrigerators. The first report of millikelvin-range experiments was given in [5], followed by a number of other sophisticated designs ([6] and others). The developments boomed especially in the last decade. Some designs are published in [7, 8, 9]. To realize a sensitivity of $10^{-9}$, the temperature stability and homogeneity over the dilatometer cell is an important issue. An appropriate material with high thermal conductivity has to be used. Additionally, in the case of magnetostriction eddy currents should be reduced. Geometrically, conducting loops perpendicular to the field have to be as small as possible. Again, such effects can be reduced by the right material choice, i.e. copper beryllium alloys or brass. Mechanical stability is another important ingredient.

In our case, a tilted-plate miniature dilatometer [10] (material: brass) was mounted inside a top-loading dilution refrigerator equipped with a superconducting 15 T magnet, see fig. 1. The sample is mounted inside the capacitor and the overall outer diameter is 18 mm. All components of the striction tensor can be measured. In contrast to other devices, the dilatometer is located in the mixing chamber, i.e. immersed directly into the $^3$He-$^4$He mixture. Although a dielectric fluid is sometimes discussed to cause problems due to evaporation and generation of gas bubbles inside the capacitor, we never encountered such problems. The capacitance indeed is stable and the sensitivity is in the range of $10^{-9}$ over the full temperature range 0.015 K to 2 K.

In general, all existing capacitive dilatometers are applicable in steady-field magnets, but the available field is limited to 45 T. To extend the field range to 100 T the pulsed-magnet technique with pulse durations between 10 ms and 1 s is state of the art. The fast field-sweep rates make eddy-current effects a critical issue. Thereby, not only the thermalisation is a crucial challenge, but also the induced magnetic moments. As a first try [11] a push-rod design was used with the capacitor outside the magnet. However, the not temperature stabilized push-rod reduced the resolution. Next, an attempt was made with nonmetallic miniaturised capacitors using electrically insulating glass plates coated with slitted metallic films [12, 13, 14]. However, temperature instabilities

Figure 1. Millikelvin dilatometer ready for installation into a dilution refrigerator.
Figure 2. Details of the FBG setup. Left: Longitudinal geometry, fibre coming from left. Right: Transversal geometry, fibre bent, coming from right. Sample is small part in black.

and, more severely, mechanical noise limited the sensitivity to $10^{-5}$. A real breakthrough was achieved using the optical fibre-bragg gratings. It is based on the modulation of the refractive index in an optical fibre. A length change of the fibre is detected by a change of the wavelength of reflected light. The fibre can be glued directly to the sample surface ([15] and fig. 2). This technique improved the sensitivity to $10^{-7}$ and beyond [16, 17].

The next step to measure dilatometry in even more extreme experimental conditions is to apply high pressure. The FBG technique is perfectly suited to be coupled to pressure cells [18]. Compact size piston-cylinder cells give the ability to perform dilatometry studies at pressures up to 3 GPa and at temperatures down to 25 mK.

3. Results
The magnetostriction of the spin-ice compounds Dy$_2$Ti$_2$O$_7$ and Ho$_2$Ti$_2$O$_7$ was measured longitudinally with the miniature dilatometer shown in fig. 1 in order to study the coupling of the lattice to the magnetic properties. The external field was applied along the [111] direction. To separate magnetodielectric effects [19] the sample was completely shielded by a metallic foil and/or a metallic shield installed into the dilatometer.

The magnetostriction of Dy$_2$Ti$_2$O$_7$ at low temperatures (fig. 3) shows a steady increase of the sample length up to about 1.2 T followed by a sudden drop and saturation above 2 T. The anomalies can be understood by a change from the 2-in/2-out ground state to the Kagomé-ice state. Above 1 T, this configuration switches to the 3-in/1-out (or vice-versa) configuration visible by the negative expansion. In zero field, the spin-ice state breaks down at temperatures above 1.5 K into a thermally excited non-ice-rule configuration [20, 21]. Our magnetostrictive investigations agree with the phase diagram reported in [22].

The situation for Ho$_2$Ti$_2$O$_7$ (fig. 4) is similar. The relative lattice changes are again of the order of $10^{-4}$. A clear maximum in the magnetostriction is visible above 2.0 T that separates the Kagomé-ice state from the 3-in/1-out configuration. The data at 0.1 K and 0.3 K are highly hysteretic in field, consistent with [23]. This behavior is related to the slow spin dynamics [24]. Again, at higher temperatures the maximum of the magnetostriction anomaly shifts to higher fields characteristic for predominantly ferromagnetic correlations.

In a first simulation using a mean-field theory (program McPhase, www.mcphase.de) the behaviour can be explained by a dominant crystal-field striction process where two-ion striction effects seem to be negligible.

In conclusion, we may state that capacitive dilatometry is a versatile technique that allows magnetoelastic investigations even in the millikelvin range. As examples, we showed some results of such measurements for the spin-ice materials Dy$_2$Ti$_2$O$_7$ and Ho$_2$Ti$_2$O$_7$. These compounds have comparable features in the magnetostriction. At the transition from the Kagomé-ice state to the 3-in/1-out configuration clear anomalies in the magnetostriction signal are observed. The strong lattice effects indicate the influence of lattice distortions and interatomic distances.
Figure 3. Magnetostriction of Dy$_2$Ti$_2$O$_7$. (The data are shifted for a better visibility).

Figure 4. Magnetostriction of Ho$_2$Ti$_2$O$_7$. (The data are shifted for a better visibility).

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