The Barocaloric Effect in $\mathrm{Ce_xLa_{1-x}Sb}$

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Recently a new adiabatic cooling technique based on a pressure-induced structural phase transition was described for $\mathrm{Pr_{1-x}La_xNiO_3}$. *Here we report on the pressure-induced magnetic phase transition of* CeSb *used for cooling.*

In the barocaloric effect [BCE] a change of the isothermal total entropy of the system is used to establish adiabatic cooling in a similar manner as in the well-known magneto-caloric effect [1]. However in the BCE we rather use external pressure in order to change the magnetic entropy than huge external magnetic fields which are still needed for magneto-caloric cooling. The magnetic entropy S_m of a rare-earth compound can be written as

$$S_m = -R\sum_i p_i \ln p_i, \quad \text{with} \quad p_i = e^{-E_i/k_BT} \qquad (1)$$

with E_i the crystal-field energy levels of the rare-earth ion at temperature T and R the molar gas constant. Any change of the E_i's immediately affects S_m and adiabatic cooling from state 1 to state 2 gets possible, if

$$S(T,1) < S(T,2), \quad \text{with} \quad S = S_m + S_l + S_{e^-} \qquad (2)$$

the total entropy of the system and S_l and S_{e^-} its lattice and electronic contributions, respectively. The change in the energy levels may arise by a pressure-induced structural phase transition as in the case of $\mathrm{Pr_{1-x}La_xNiO_3}$ or due to Zeeman splitting in a pressure-induced magnetic transition as in the case of CeSb [2].
CeSb crystallizes in the NaCl structure. Below $T_N \approx 16\,\mathrm{K}$ antiferromagnetic ordering occurs in various phases. From neutron diffraction T_N is known to increase linearly upon uniaxial pressure as $dT_N/dp \approx 8\mathrm{K}/\,\mathrm{GPa}$ [3]. Thus the application of pressure at temperatures just above $16\,\mathrm{K}$ results in a magnetic phase transition from the paramagnetic into the ordered state and vice versa upon the removal of pressure. Since in the ordered state the isothermal entropy is smaller than in the paramagnetic state, cooling can be realized by adiabatic release of pressure (see Eq. 2).
For the direct measurement of the barocaloric effect, a cubic single crystal of 3 mm side length was mounted on an uniaxial pressure device allowing the *in situ* change of p at cryogenic temperatures. The temperature of the crystal was tracked by a thermocouple glued on one side of the crystal. The socket and the anvil of the pressure device were made of ZrO ensuring good insulation and thus almost adiabatic conditions for small time scales.

h	k	l		F^{exp}	F^{HF}	F^{DF}	F^{sph}
1	1	1	c	154.91	153.52	152.90	152.82
2	2	0	c	180.70	182.11	182.08	182.91

Table 1: This table is an example, it does not belong to the rest of the report.

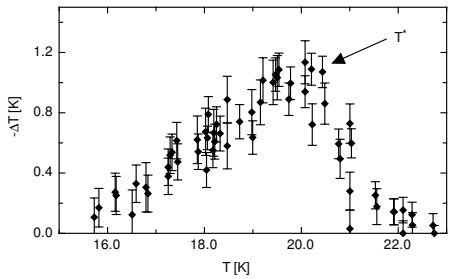

Figure 1: Measured BCE $-\Delta T$ versus initial temperature upon the release of $p = 0.26\,\mathrm{GPa}$.

Fig. 1 shows the cooling effect $-\Delta T$ in function of the initial temperature T prior to the release of pressure.
The cooling effect can be calculated, if the entropies at ambient and elevated pressure are known. Based on a molecular field calculation we expect bigger cooling rates $-\Delta T$ than actually observed (Fig. 2). This largely results due to the inertia of the thermocouple as well as insufficient insulation of the sample. Thus we may not have realized fully adiabatic transitions. So far we could observe a maximal cooling of $-2\,\mathrm{K}$ at a pressure release of $0.53\,\mathrm{GPa}$.

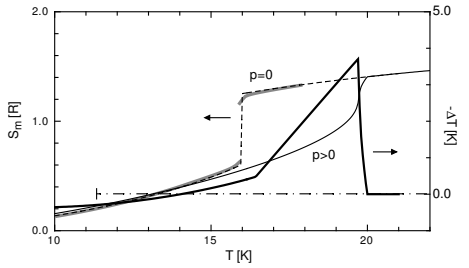

Figure 2: BCE based on a molecular field calculation (*left*: experimental entropy for $p = 0$ (from c_p) [gray] and calculated entropies for $p = 0$ [dashed black] and $p > 0$ [solid black], *right*: expected cooling $-\Delta T$).

[1] K. A. Müller, M. Koch, S. Fischer, F. Fauth, A. Furrer, Appl. Phys. Lett. **73**, 1056 (1998)

[2] Th. Strässle, A. Furrer, K. A. Müller, J. Alloys and Compounds **303-304**, 288 (2000)

[3] B. Hälg, A. Furrer and O. Vogt, Phys. Rev. Lett. **57**, 2745 (1986)

Work partially/fully performed at SINQ
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