The magnetic properties of the multi-functional intermetallic compound Pr$_{1-x-y}$La$_x$Pb$_y$Te in high magnetic fields

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Abstract. The intermetallic compound Pr$_{1-x-y}$La$_x$Pb$_y$Te shows a wide spectrum of physical phenomena. Depending on the metallurgical composition as function of $x$ and $y$, the compound changes its behavior from van Vleck paramagnetism and hyperfine-enhanced nuclear-magnetic order to super- or semiconductivity. In addition, there are remarkable interplay effects between these ground states. In consequence, Pr$_{1-x-y}$La$_x$Pb$_y$Te may serve as an interesting material for nuclear-spin quantum-computing experiments. In this contribution, we focus on measurements of the magnetic properties performed in high magnetic fields. We present first data of the magnetization of Pr$_{1-y}$Pb$_y$Te taken in pulsed magnetic fields up to 47 T for the compositions $y = 0$, 50, and 90 %.

1. Introduction

Multi-functional materials, such as magnetic semiconductors are in the focus of interest of fundamental and applied solid state physics. Their variety of physical phenomena on one hand and their technical applicability on the other motivate an ongoing trend to classify known and to search for new multi-functional compounds. Their potential to influence strongly the design of next generation micro and nano circuits give them a key role in the future development of information technology. As a new technological approach, materials which make use of the localized nuclear magnetic moments could have an impact in quantum-computing concepts. In one of these concepts, firstly proposed by Lloyd et al. [1], structures of miniaturized NMR setups could replace integrated hardware circuits which are used in computer architecture nowadays. Compared to the classical hardware circuits which are predefined and limited in their functionality, a wide variety of logic operations can be performed by means of NMR pulse sequences as well as subsequent free-induction decays and spin echoes of the interacting nuclear spins. Moreover, the response of the nuclear magnetic spin systems would reflect their quantum-mechanical nature. In consequence, the interference of quantum-mechanical states could be used to solve computational challenges like factorization of large numbers (see, e.g. in [2, 3]), not possible in classical computer architecture, so far. A recent work of G. Yusa et al. [4] accelerated the progress in this field considerably and brought a technical application of NMR qubits much closer to reality.
Here we report studies on the intermetallic compound Pr\(_{1-x-y}\)La\(_x\)Pb\(_y\)Te which is a possible material for quantum-computing applications due to its wide spectrum of physical phenomena which varies from nuclear magnetic order to super- or semiconductivity. This variety depends on the metallurgical composition as function of \(x\) and \(y\). In more detail, in PrTe (\(x = y = 0\)) the crystal-electrical-field splitting with a singlet-ground state leads to van Vleck paramagnetism only, whereas several other rare-earth tellurides possess a magnetically ordered ground state. However, there is a special type of magnetic moments, so-called hyperfine-enhanced nuclear magnetic moments of the Pr atoms which make PrTe attractive. Compared to ordinary nuclear magnets the effective nuclear magnetic moment in PrTe are enhanced by a factor of 10 and have, in consequence, already a size of a small electronic magnetic moment of 0.022 \(\mu_B\) [5, 6]. Using materials with hyperfine-enhanced nuclear magnetic moments instead of ordinary nuclear magnets, such as Ga\(_{1-x}\)Al\(_x\)As, nuclear spin quantum computing could be more easily demonstrated, operate with greater stability and could be driven under less critical conditions. Further, LaTe (\(x = 1\), \(y = 0\)) is a superconductor with \(T_c = 5.7\) K. The singlet ground state of the Pr ions in Pr\(_{1-x}\)La\(_x\)Te leads to a large critical Pr concentration, \(x = 50\%\), for Cooper-pair breaking [6]. Finally, PbTe (\(x = 0\), \(y = 1\)) is a narrow-band semiconductor and Pr\(_{1-y}\)Pb\(_y\)Te appears to be a nuclear magnetic semiconductor at high Pb concentrations.

2. Hyperfine-enhanced nuclear magnetism and its interplay with superconductivity
So far, we have presented data on the hyperfine enhanced nuclear magnetism of Pr\(_{1-x-y}\)La\(_x\)Pb\(_y\)Te, in particular on the nuclear antiferromagnetic ordering behavior of PrTe at \(T_N = 0.6\) mK, as well as on the competition of hyperfine-enhanced nuclear magnetism with superconductivity in Pr\(_{1-x}\)La\(_x\)Te [5, 6]. We have observed a strong competence between the hyperfine enhanced nuclear magnetism and superconductivity, in particular at high Pr concentrations around \(x = 50\%\), where the van Vleck paramagnetic contribution almost leads to a suppression of superconductivity. In Pr\(_{0.50}\)La\(_{0.50}\)Te reentrant superconductivity has been observed where superconductivity is affected below about 20 mK in zero field and finally destroyed at sub-mK temperatures [5, 6].

3. Hyperfine enhanced nuclear magnetism in the metallic and semiconducting phase
In a subsequent investigation of Pr\(_{1-y}\)Pb\(_y\)Te we have studied the influence of the Pb dilution, \(y\), on the van Vleck paramagnetism per Pr content, \(1-y\) [7]. We have performed measurements of the magnetization of Pr\(_{1-y}\)Pb\(_y\)Te samples in a commercial SQUID magnetometer at 1.8 K \(\leq T \leq 350\) K in a magnetic field \(B = 0.5\) T. It has been demonstrated that nonmagnetic Pb dilution decreases the size, but does not eliminate the hyperfine enhancement, of the nuclear magnetic moments of \(^{141}\)Pr in Pr\(_{1-y}\)Pb\(_y\)Te. In particular, at high Pb concentrations, Pr\(_{1-y}\)Pb\(_y\)Te could be used as a hyperfine-enhanced nuclear magnetic semiconductor for quantum computing. We have observed a clear influence of the Pb content on the crystal-electrical-field parameters obtained from fitting the singlet-triplet expression [8] of the van Vleck equation to the magnetization data [7]. As an example, we extracted the energy splitting between the singlet ground and first excited triplet state, \(\Delta = (70 \pm 2)\) K, for PrTe (\(y = 0\)) which is in reasonable agreement with the literature value \(\Delta_n = 76\) K obtained through inelastic neutron diffraction [9]. We observed unexpected values of the ground state splitting for \(y = 90\%\) and \(y = 99\%\) [7]. In addition, we have found a surprising change from van Vleck type to single-impurity Langevin-type electronic magnetism at a high Pb concentration, \(y \geq 99.9\%\), where the system is already transformed from a diluted intermetallic compound into a magnetically doped semiconductor.

4. Magnetic Properties of Pr\(_{1-y}\)Pb\(_y\)Te in high magnetic fields
For a better understanding of the influence of the Pb content on the crystal-electrical-field parameters of the Pr-4f electrons in Pr\(_{1-y}\)Pb\(_y\)Te, we have started now to perform investigations in high magnetic fields. The application of high magnetic fields leads to a shift of the crystal-electrical-field levels. Vice versa, crystal-electrical-field properties can be analyzed from and compared to susceptibility and magnetization data for the case that the fields are large enough to influence the size of the van Vleck susceptibility [10]. Here, we report data of the magnetic properties of PrTe, Pr\(_{0.90}\)Pb\(_{0.10}\)Te, and Pr\(_{0.10}\)Pb\(_{0.90}\)Te taken in fields up to 47 T. The measurements have been performed at the high magnetic
field laboratory of the IFW Dresden [11]. As for our previous investigations of this material class, data have been taken on samples produced by the commercial supplier MaTeck (Juelich, Germany). Throughout the measurements, powdered samples in order to minimize eddy-current effects have been immersed in liquid helium at 4.2 K. A pulsed-field coil energized by a 10 kV, 1.0 MJ capacitive pulsed-power supply was used to generate magnetic-field pulses up to 47 T. A crowbar circuit of the power supply was used in order to generate a field reversal to -5 T (a pulse profile taken during the measurements is given in Fig. 2).

In Fig. 1 data of the magnetic moment as function of the applied magnetic field are shown. The magnetic moment per mass PrTe, Pr$_{0.50}$Pb$_{0.50}$Te, as well as of Pr$_{0.10}$Pb$_{0.90}$Te is clearly affected by the field. The observed curvatures of these data give the chance to derive the crystal-electrical-field parameters in more detail now. A numerical method for this analysis is given by the “McPhase” software created by M. Rotter [12].

![Figure 1](image1.png)

**Figure 1.** Magnetic moment scaled by sample mass of PrTe, Pr$_{0.10}$Pb$_{0.90}$Te, and Pr$_{0.50}$Pb$_{0.50}$Te as function of the applied magnetic field. The temperature has been fixed at 4.2 K.

In Fig. 2 the data of the magnetic moment are taken to calculate the static magnetic susceptibility scaled by the molar content of Pr as function of the applied magnetic field. For a better visibility, we have displayed the results for PrTe and Pr$_{0.10}$Pb$_{0.90}$Te only. The susceptibility of Pr$_{0.50}$Pb$_{0.50}$Te is about the same as the one of Pr$_{0.10}$Pb$_{0.90}$Te, i.e., the susceptibility scales with the Pr content for these concentrations. In Fig. 2 also

![Figure 2](image2.png)

**Figure 2.** Static magnetic susceptibility of PrTe and Pr$_{0.10}$Pb$_{0.90}$Te at T = 4.2 K as function of the applied magnetic field measured in a pulsed field coil. The arrows indicate the field-sweep directions. The pulsed-field profile is shown in the inset. For further details, see text.
the possibility for the occurrence of hyperfine-enhanced nuclear magneto-caloric effects is visible: at the start of the field pulse the susceptibility of the samples is lower than at the end. This may be explained by a polarization (of up to a few percent using $T = 4.2$ K, $B_{\text{max}} = 47$ T, and moments of a size of 0.022 $\mu_B$) and pre-cooling during the pulse, and subsequently demagnetization and local cooling of the hyperfine-enhanced Pr nuclear moments during the decay of the pulse.

5. Conclusion
We have measured the van Vleck susceptibility of Pr$_{1-y}$Pb$_y$Te for $y = 0, 0.50$, and 0.10 and observed a clear depression of its size with the magnetic field. The data may serve as a basis to numerically determine the crystal-electrical-field parameters of the investigated Pr$_{1-y}$Pb$_y$Te compounds, e.g. by using the “McPhase” software which has been established by M. Rotter [12]. As next steps we will also investigate samples with a smaller content of Pr, in particular samples with Pr concentrations close to 0.1 %, where a change of van Vleck paramagnetic to Langevin-type single-impurity magnetic behavior has been observed. In addition, we will perform magnetization measurements to higher magnetic fields.

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