# $\mu^+$SR investigation of effect of diamagnetic doping in the stability of the frozen spin ice state. ESF report

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### A. Spin Ice systems - Brief summary

$Ho_2Sn_2O_7$ belongs to the general class of the rare-earth (RE) oxides $RE_2B_2O_7$. The crystalline structure of these compounds can be well described by a pyrochlore lattice (composed of corner-sharing tetrahedra) whose lattice points are occupied by the RE ions. The (2J+1)-fold degeneracy associated with the free $RE^{3+}$ ion ground state configuration (with electronic angular momentum J) can be partially removed by the crystal field (CF) depending on the peculiar features of the considered ion. When RE = (Ho, Dy), in particular, one finds that the degeneracy is lifted in such a way that the resulting ground state is the almost perfect $|m_J = \pm J\rangle$ doublet, namely the ionic magnetic moment is pointing inside or outward the considered tetrahedron with maximum value of its projection along the local $\langle 111\rangle$ direction. The ground state is separated from the first excited state by a thermal energy of about 250K [1]. At low temperatures, then, the strong CF-induced axial magnetic anisotropy together with the ferromagnetic interactions between the moments make it possible to map the system onto another one in which fictitious Ising variables are antiferromagnetically interacting [2]. This leads to a geometrically frustrated ground state for the real system in which, for each tetrahedron, two moments point inside and the other two outward [3], [4]. The great similarity of this magnetic configuration with the microscopic ground state of protons around the oxygen ions in water $I_h$ ice leads to the name *spin ice* for the magnetic compounds. The ground state's high degeneracy induced at low temperatures is at the root of the peculiar thermodynamic property of the zero-point entropy, theorized by Pauling on water ice [5] and measured for the first time on the magnetic compound $Dy_2Ti_2O_7$ [6].

### B. Diamagnetic dilution effects in $Ho_{2-x}Y_xSn_2O_7$

The described system can be considerably modified by the gradual replacement of the RE ions with nonmagnetic ions. In particular, we studied the effects of the magnetic dilution in $Ho_{2-x}Y_xSn_2O_7$ and the results are presented in a recent work to be published on the Journal of Physics: Conference Series [8]. Our studies were basically focussed on three aspects.

- The properties of the ground state. These were examined by specific heat (and, then, entropy) measurements at low temperatures.
- The properties of the low-energy excitations of the system by means of $\mu^+$SR measurements.
- The modification of the CF energy levels. Starting from some $^{119}$Sn-NMR $\frac{1}{T_1}$ measurements one can derive the theoretical behaviour due to transitions between these levels as induced by the spin-phonon interaction.

$\mu^+$SR measurements, in particular, were carried out at ISIS in the Rutherford-Appleton Laboratories (RAL) near Oxford, UK. A fully spin-polarized pulsed beam of muons is implanted inside the sample, where each muon interacts with the local magnetic field through its magnetic moment giving rise to a Larmor precession. The weak decay into a positron emitted in the direction of the muon's moment after a mean time of $\sim 2.2\mu$s allows one to follow the statistical time evolution of the muons' spin polarization due to magnetic effects inside the sample inferring important information about the nature of the local magnetic fields. $Ho_{2-x}Y_xSn_2O_7$ samples characterized by x = 0, 0.1 and 0.3 were studied in the temperature range of 90mK $\div$ 200K in zero-field (ZF) condition and upon the application of external longitudinal static magnetic fields up to 0.2T. In spin ice systems the expected trend for the longitudinal depolarization function $p_z(t)$ is given by the Kubo-Toyabe (KT) function [7]. The static KT function is characterized by a fast full loss of the initial asymmetry $a_0 \sim 25\%$ followed by a recovery of a constant value of about $\frac{a_0}{3}$ (the so-called $\frac{1}{3}$-tail). This is obtained in a dense magnetic system characterized by static local magnetic fields. Slowly fluctuating magnetic fields with frequency $\nu_c$ lead to the slow exponential decay of the $\frac{1}{3}$-tail with a reaxation rate $\lambda \propto \nu_c$ while fast dynamical processes make the whole function to closely resemble an exponential-like decay characterized by $\lambda \propto \nu_c^{-1}$. In $Ho_2Sn_2O_7$ the high magnetic field associated with the $Ho^{3+}$ magnetic moment causes each muon to start a very fast precession. This fact, together with the instrumental dead time and the low temporal resolution due to the finite width of each beam's pulse, makes it impossible to follow the precise evolution of the fast depolarization at short times. As a result, $\frac{2}{3}$ of the initial asymmetry is completely lost and only a slow exponential decay can be observed. This behaviour was interpreted as the observation of the slow decaying $\frac{1}{3}$-tail (the short-time dip being prevented from a direct

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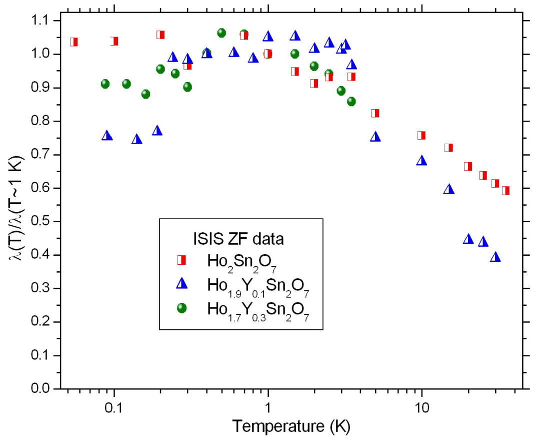


FIG. 1: Fit results of a single-exponential decay (with rate $\lambda$) for the longitudinal depolarization function $p_z(t)$ as a function of temperature T in ZF condition (as obtained in ISIS - see [8]).

observation by instrumental limitations). The examined samples possibly show the persistence of slow dynamical processes (down to the lowest accessible temperatures) superposed to mainly static local magnetic fields - as confirmed by the high sensitivity to the application of an external longitudinal magnetic field. This can be deduced from an increase in $\lambda$ with decreasing T till a flattening about a constant value of $\sim 0.25\mu s^{-1}$ for T $\lesssim$ 2K (see Fig. 1). This behaviour is common to other geometrically frustrated pyrochlore materials such as $Tb_2Ti_2O_7$ or $Tb_2Sn_2O_7$ [9], [10].

## C. PSI measurements. $4^{th}$ - $5^{th}$ November 2008

### 1. *Aim of the experiment*

Other measurements have been performed at the S$\mu$S laboratory (LTF facility) in Paul Scherrer Institute (PSI) near Zurich, Switzerland. Our main purpose was the investigation of the short-time behaviour of the longitudinal depolarization function at different temperatures and longitudinal magnetic fields. This study is possible because of the better temporal resolution associated with the continuous muons' beam produced at PSI. We decided to first examine the pure sample (x = 0) in this two-days measurement leaving for future studies the diluted samples. Two different kinds of measurements were performed.

- The longitudinal depolarization function $p_z(t)$ was studied at very low temperatures (100mK $\div$ 1K) in ZF condition. The selected range of temperatures is safely deep into the so-called "frozen spin ice region" [7].

TABLE I: Fit results for ZF longitudinal depolarization function at different values of temperature T. The estimated parameters refer to Eq. 1.

| Temperature | $\lambda_F$ ($\mu s^{-1}$) | $a_0$ | $\lambda_S$ ($\mu s^{-1}$) | $a_1$ |
|---|---|---|---|---|
| 100mK | 43.9±3.0 | 18.7±1.0 | 0.375±0.015 | 5.8±0.1 |
| 200mK | 41.5±2.9 | 17.6±0.9 | 0.369±0.014 | 6.0±0.1 |
| 300mK | 41.5±2.9 | 18.0±0.9 | 0.368±0.015 | 5.7±0.1 |
| 600mK | 41.5±2.6 | 19.5±0.9 | 0.300±0.014 | 5.1±0.1 |
| 800mK | 46.0±3.0 | 20.5±1.0 | 0.297±0.014 | 5.0±0.1 |
| 1K | 44.9±3.8 | 19.2±1.2 | 0.325±0.018 | 5.2±0.1 |

- The behaviour of the longitudinal depolarization function at T = 100mK was studied upon the application of a longitudinal magnetic field H in a wide range of values (0.02T $\div$ 2T).

### 2. *$\lambda$ decay rate vs T in ZF condition*

All the data to be presented seem to be of better quality than the ones obtained at ISIS due to a much lower level of constant background signal, estimated to be b $\sim$ 3% (against a value of b $\sim$ 7% characteristic of ISIS measurements). b is due to the fraction of those muons that miss the sample and are implanted, for instance, in the silver sample holder giving a non-relaxing component. The fit have been obtained through WIMDA software.

At a first sight, $p_z(t)$ can always be fitted by a single exponential function (characterized by a decay rate $\lambda$) starting from low values of the initial asymmetry $a_0 \simeq 10\%$ (see Fig. 2). These features are in perfect agreement with our data from ISIS measurements. A more precise inspection of the short-time decay shape, anyway, shows a very interesting feature, namely an initial fast exponential relaxation (decaying in $\sim 0.05\mu s$) characterized by a high $\lambda_F$ rate suddenly followed by the much slower exponential decay with $\lambda_S$ (see Fig. 2 - inset). The general phenomenological function used to fit the data, then, was chosen to be

$$p_z(t) = a_0 \exp(-\lambda_F t) + a_1 \exp(-\lambda_S t) + b \qquad (1)$$

where b was kept fixed to the value 3%.

The temperature dependence of $\lambda_F(T)$ and $\lambda_S(T)$ is reported in Figs. 3 and 4 while all the data relative to the fits have been reported in Tab. I. The analysis of the ZF data (and, in particular, of the slow-decaying component) shows an overall agreement with our first measurements performed at ISIS. $\lambda_S(T)$, anyway, assumes values in the order of magnitude of $0.3\mu s^{-1}$ systematically higher than the values of about $0.2\mu s^{-1}$ derived at ISIS. With decreasing temperature under 500mK, moreover, one notes an abrupt increase of $\lambda_S$ from $\sim 0.3\mu s^{-1}$ to $\sim 0.36\mu s^{-1}$. A similar but less marked effect was already observed in ISIS data. It is interesting to underline that the opposite effect was observed in the diluted samples, namely a sudden decrease in $\lambda_S$ for T $\lesssim$ 300mK. $\lambda_F(T)$, on the

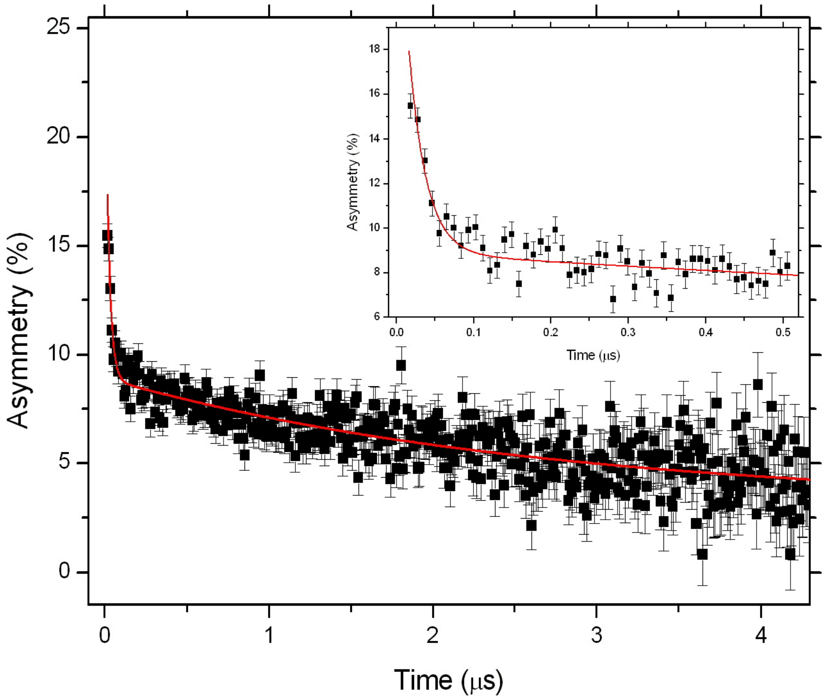


FIG. 2: Longitudinal depolarization function at T = 100mK in ZF condition. Inset: enlargement at short times. Red lines represent the fit composed of two exponential decays (see text).

TABLE II: Fit results for longitudinal depolarization function at different values of applied external longitudinal field at T = 100mK. The estimated parameters refer to Eq. 1.

| Field (T) | $\lambda_F$ ($\mu s^{-1}$) | $a_0$ | $\lambda_S$ ($\mu s^{-1}$) | $a_1$ |
|---|---|---|---|---|
| 0 | 43.9±3.0 | 18.7±1.0 | 0.375±0.015 | 5.8±0.1 |
| 0.02 | 38.0±3.7 | 17.1±1.8 | 0.035±0.009 | 4.2±0.1 |
| 0.05 | 34.8±4.2 | 12.7±1.6 | 0.026±0.008 | 4.8±0.1 |
| 0.1 | 33.1±4.3 | 11.3±1.5 | 0.034±0.007 | 5.2±0.1 |
| 0.2 | 30.3±3.5 | 11.6±1.3 | 0.036±0.007 | 6.0±0.1 |
| 0.5 | 22.4±2.3 | 10.2±0.9 | 0.030±0.004 | 8.6±0.1 |
| 1 | 12.8±1.2 | 7.2±0.5 | 0.058±0.003 | 12.1±0.1 |
| 1.5 | 4.1±0.4 | 4.3±0.2 | 0.034±0.003 | 14.9±0.1 |
| 2 | 4.6±0.5 | 3.8±0.2 | 0.029±0.002 | 18.8±0.1 |

other hand,is nearly T-independent reaching a value of $\sim 43\mu s^{-1}$. At each temperature, then, one can note that $\lambda_F \simeq 10^2 \lambda_S$.

### *3. λ decay rate vs H at T = 100mK*

The magnetic field dependence of $\lambda$ for x = 0 sample (at T = 100mK) was studied over a wide range of H values (0.02T ÷ 2T). The application of such fields, as already cited, is very useful in order to obtain information on the dynamical behaviour of the local magnetic fields inside the sample. The sensitivity of the depolarization function on the applied external field, in particular, is consistent with a mainly static distribution of local magnetic fields. The obtained field dependence for some fields is reported in Fig. 5. The analysis of the data was performed by means of the fit function reported in Eq. 1. Fit parameters are presented in Tab. II and in Figs. 6 and 7. The field dependence of the depolarization function as measured for the x = 0.3 sample at ISIS seems to be somewhat different with respect to the trend observed at PSI for the x = 0 sample. In both cases, the application of an external static magnetic field is useful in order to obtain the constant full initial asymmetry of ∼0.25% at sufficiently high values of the field's intensity. In the pure sample, anyway, the recovery of the full initial asymmetry seems to be very similar to the one of $Dy_2Ti_2O_7$ compound [7], namely much slower than in x = 0.3, where a field of about 0.02T could lead to the recovery of half-asymmetry. The application of fields as low as 0.02T does not lead to any sizeable modification of the asymmetry value of the tail and fields greater than 0.5T are required in order to substantially modify it. The two-component decay shape is not affected by the application of magnetic fields but the initial fast decay seems to be more and more quenched by increasing the magnetic field's intensity (as can be seen in Fig. 7

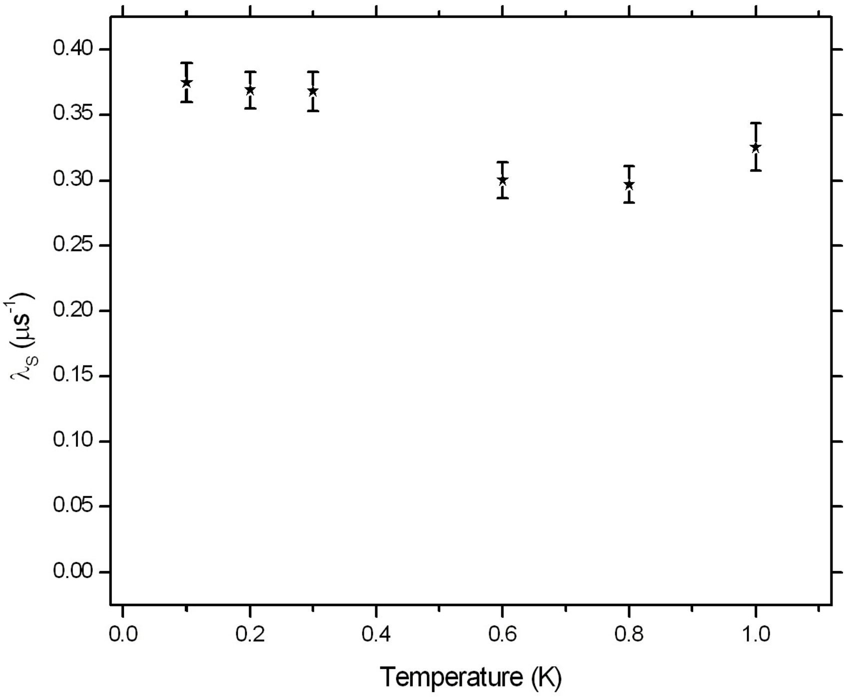


FIG. 3: Temperature dependence of $\lambda_S(T)$ namely of the slow exponentially-decaying component of the longitudinal depolarization function in ZF condition (see text).

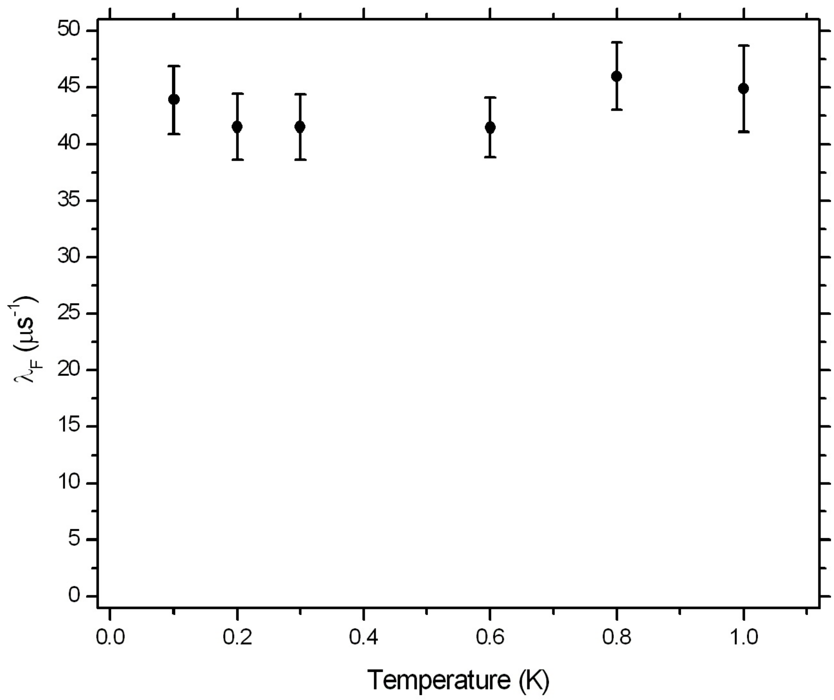


FIG. 4: Temperature dependence of $\lambda_F(T)$ namely of the fast exponentially-decaying component of the longitudinal depolarization function in ZF condition (see text).

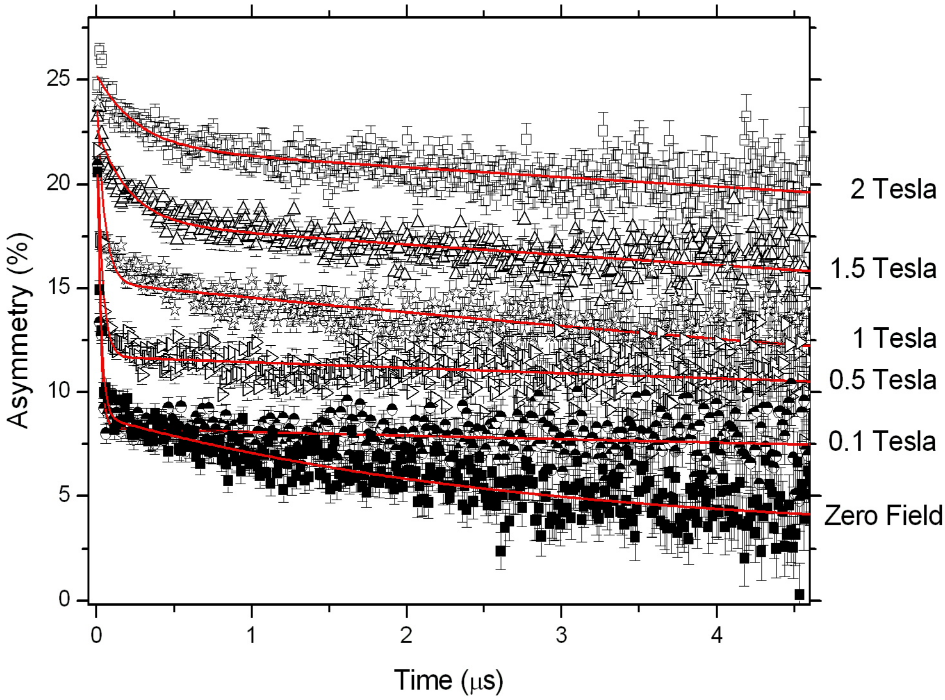


FIG. 5: Longitudinal depolarization function at T = 100mK with different applied external longitudinal fields. Red lines refer to the two-component exponential fit (see text).

both $\lambda_F$(H) and $a_0$(H) show a monotonic decrease). The origin of the initial fast decay, then, is possibly explained by the effect of mainly static local high magnetic fields - associated with the large electronic magnetic moment of the $Ho^{3+}$ ion - causing a very fast loss of the initial beam polarization (that, anyway, cannot be entirely followed). The decoupling of such a contribution by the application of an external magnetic field is consistent with the static origin of the local magnetic field. At the same time, one can see that - after an abrupt drop of one order-of-magnitude due to the application of the lowest external magnetic field - $\lambda_S$(H) takes an almost H-independent value of $\sim 0.03\mu s^{-1}$, displaying in such way its dynamical origin.


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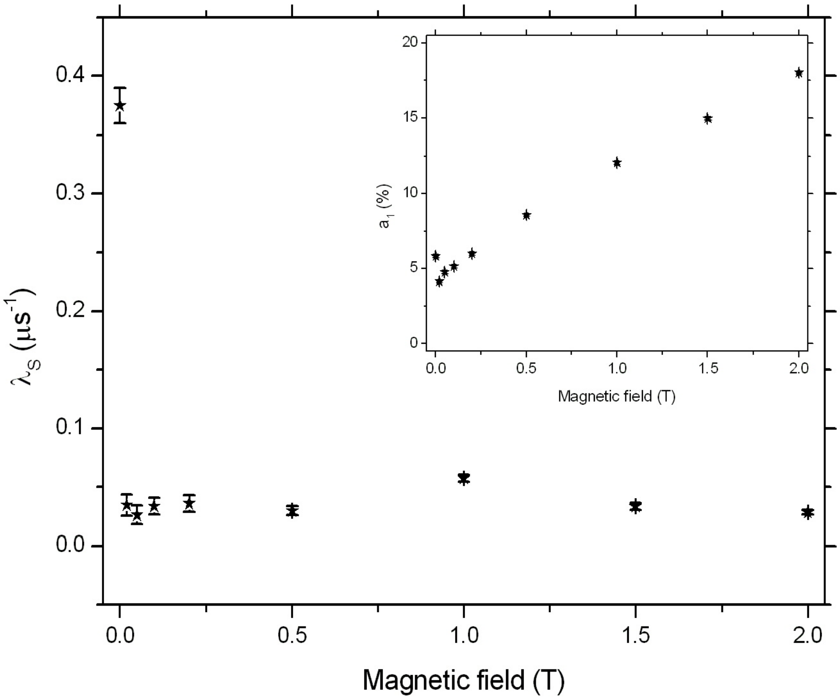


FIG. 6: Field dependence of $\lambda_S$(H) namely of the slow exponentially-decaying component of the longitudinal depolarization function at T = 100mK (see text). Inset: evolution of $a_1$(H).

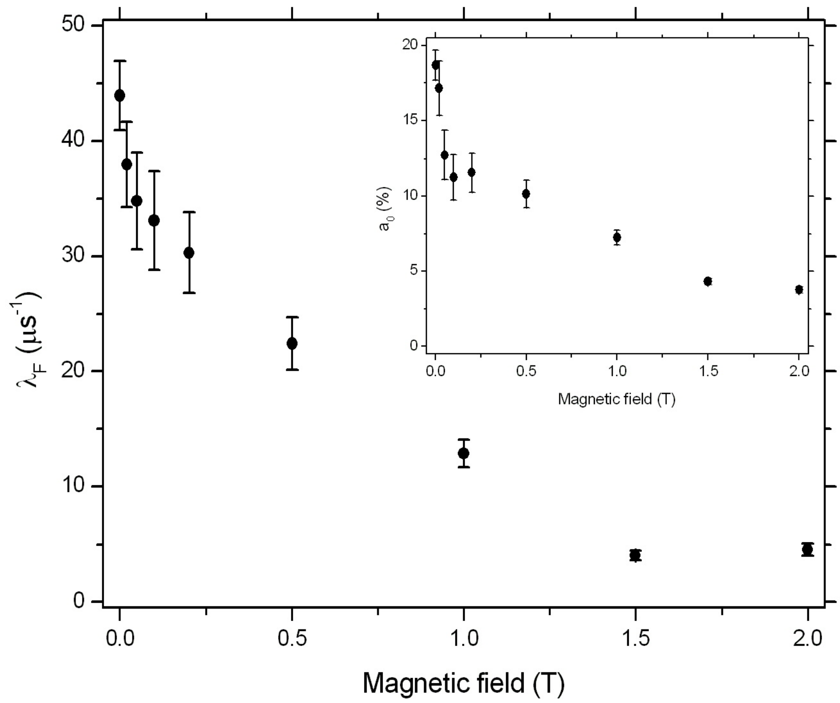


FIG. 7: Field dependence of $\lambda_F$(H) namely of the fast exponentially-decaying component of the longitudinal depolarization function at T = 100mK (see text). Inset: evolution of $a_0$(H).