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LARGE ENHANCEMENT OF DEUTERON POLARIZATION WITH FREQUENCY MODULATED MICROWAVES

The Spin Muon Collaboration (SMC)

Abstract

We report a large enhancement of 1.7 in deuteron polarization up to values of 0.6 due to frequency modulation of the polarizing microwaves in a two liters polarized target using the method of dynamic nuclear polarization. This target was used during a deep inelastic polarized muon-deuteron scattering experiment at CERN. Measurements of the electron paramagnetic resonance absorption spectra show that frequency modulation gives rise to additional microwave absorption in the spectral wings. Although these results are not understood theoretically, they may provide a useful testing ground for the deeper understanding of dynamic nuclear polarization.

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Measurements of deep inelastic scattering of polarized muons from polarized protons and deuterons determine the spin dependent structure functions of the nucleon which allow fundamental tests of quantum chromodynamics and of models of nucleon structure [1]. The precision of these experiments is strongly related to the polarization of the target nucleons. Therefore, the large enhancement of our deuteron target polarization which we discovered during the data-taking for deep inelastic muon scattering [2] had a significant impact on our experiment at CERN. The discovery was associated with a faulty regulator of the high voltage power supply of a microwave source. After controllable frequency modulation (FM) of the microwave tube was implemented, a gain by a factor of 1.7 in the maximum deuteron vector polarization and of 2.0 in the polarization growth rate were achieved. These increases have been of crucial importance because data-taking extends over many months and the statistical error is proportional to $1/PN^{1/2}$, in which $P =< I_z > /I$ is the target vector polarization and N is the number of scattered events. The magnitude of the enhancement has been reported earlier [3]. The purpose of this paper is to detail the full characteristics of this effect, to present new data on the electron paramagnetic resonance absorption (EPR) spectrum and to discuss briefly processes which may contribute to the FM phenomenon.

The polarized target [4, 5] consists of two cells each 40 cm long and 5 cm in diameter located in a large cylindrical multimode microwave cavity. The two target halves are polarized in opposite directions by dynamic nuclear polarization (DNP). The target material is glassy, perdeuterated 1-butanol, C_4D_9OD with 5% by weight of deuterium oxide, doped with the paramagnetic EDBA-Cr(V) complex [6] to a concentration of $7 \cdot 10^{19}$ cm^{-3} [7]. It is located in a magnetic field of 2.5 T with a uniformity of 10^{-4} over the volume and is cooled by a dilution refrigerator. The DNP is obtained by applying microwave power near the EPR frequency of the paramagnetic complex.

The deuteron vector polarization is measured with nuclear magnetic resonance (NMR) probes, each of which is part of a series tuned Q-meter circuit [8]. The material is sampled by five probes in each target cell. The polarization is determined from the integrated NMR signals, calibrated in thermal equilibrium at 1K. The relative accuracy of the measurement $\delta P/P$ is 5% [3].

The microwave power for DNP is produced by two extended interaction oscillators (EIO) with an emission bandwidth of about 0.1 MHz. The rate of polarization is optimized by controlling the microwave power and frequency. The frequency is controlled by the EIO cathode voltage with a sensitivity of about 0.4 MHz/V or by tuning the EIO cavity. The power is controlled by non-reflective attenuators.

For materials in which the solid effect [9, 10] dominates as a mechanism for DNP, it has been found that microwave FM can improve the rate of DNP. This appears to result from the fact that FM counteracts the effect of "hole burning" due to EPR absorption at a fixed frequency [11]. In the glassy alcohol materials with Cr(V) complexes, where the dynamic nuclear cooling [12] is the dominant mechanism for DNP, hole burning is not expected. However a polarization enhancement of 10% to 20% was observed in a fluorinated alcohol leading to polarizations of ≈ 0.80 for protons and ^{19}F [13]. References to enhancements of a few percent at polarizations around 0.70 or of about 15% for a material with only a few percent polarization can also be found in [13]. To our knowledge no studies have been reported for the effect of FM on deuteron polarization except in one

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case where FM was used to compensate for magnetic field inhomogeneity and improve the final polarization by 5% to ≈ 0.30 [14].

The large enhancement of deuteron polarization in our target due to FM came therefore as a surprise. Figure 1 shows the typical time evolution of the deuteron polarization P_D without and with FM. For this figure the cathode voltages were modulated at 1 kHz with a ≈ 50 volt peak-to-peak amplitude leading to a FM amplitude $\Delta f \approx 20$ MHz for the 69 GHz microwave source. The maximum deuteron vector polarizations under these conditions were 0.43 and -0.49.

The EPR spectrum was measured in our target at a constant frequency by scanning the magnetic field. Such a spectrum, shown in Figure 2 without FM, was obtained using a 220 Ω Speer composite carbon resistor as a bolometer, located in the dilute phase of the mixing chamber outside the target material [15]. The input power to the microwave cavity $\dot{Q}_{IN}$ is the sum of $\dot{Q}_{MAT}$, the power absorbed by the material in the EPR process, and $\dot{Q}_{NR}$ the non-resonant power absorbed into the cavity. The power absorbed by the bolometer $\dot{Q}_{SP}$ is a constant fraction r of $\dot{Q}_{NR}$. It can be expressed as $\dot{Q}_{SP} = c(T_{SP}^4 - T_{HE}^4)$ where T_{SP} is the temperature of the bolometer, T_{HE} is the temperature of the dilute phase and c is a constant [16]. During the EPR measurement the input power $\dot{Q}_{IN}$ remains constant and we can neglect the variations of T_{HE}^4. Consequently the relation $\dot{Q}_{IN} = \dot{Q}_{MAT} + \dot{Q}_{NR} = \dot{Q}_{MAT} + (c/r) \times (T_{SP}^4 - T_{HE}^4)$ shows that $\dot{Q}_{MAT}$ is a linear function of T_{SP}^4. The broad absorption band seen in Figure 2 is due to the anisotropy of the $g-$factor of the EDBA-Cr(V) electron spin. The highest positive and negative polarizations without FM were obtained at frequencies $f_0^+ = 69.090$ GHz and $f_0^- = 69.520$ GHz, respectively.

The EPR spectra with better resolution at the edges of the absorption band are shown in Figure 3a and 3b, both with and without FM. The data points with FM were obtained using a modulation amplitude $\Delta f = 4$ MHz to keep a good resolution in our spectra. In order to measure the small change in EPR aborption due to FM a novel technique of making consecutive measurements of the bolometer resistance with and without FM at each field step was employed. In Figures 3c and 3d we display the difference $\Delta T_{SP}^4 = (T_{SP}^{\text{off}})^4 - (T_{SP}^{\text{on}})^4 = (\dot{Q}_{MAT}^{\text{on}} - \dot{Q}_{MAT}^{\text{off}})/c$. These data demonstrate that FM increases $\dot{Q}_{MAT}$ in the edges of the EPR spectrum. Note that the structures in Figure 3a and 3b which extend down to 69.00 GHz and up to 69.60 GHz are almost entirely eliminated in the presence of FM even though the amplitude of FM is small compared to their width.

In Figure 4 we show the difference ΔT_{SP}^4 as a function of the frequency of FM for different input power levels $\dot{Q}_{IN}$ with an FM amplitude $\Delta f = 30$ MHz at 69.090 GHz where ΔT_{SP}^4 reaches a maximum. This difference grows with the modulation frequency up to a maximum value (indicated by the arrows) and then remains constant. The frequencies at which the additional EPR absorption reaches its maximum value increase roughly linearly with $\dot{Q}_{IN}$. A study of the polarization growth rate was performed at high negative P_D values for a setting of $\dot{Q}_{IN}$ close to the one which was used for curve 2 of Fig.4. The rate increased with modulation frequency and reached a maximum value of -0.8% per hour when modulating at 10 Hz. At this $\dot{Q}_{IN}$ value, ΔT_{SP}^4 reaches a maximum at this frequency which suggests strongly that the additional EPR absorption due to FM is what leads to the enhanced DNP.

In further measurements, we have established that the highest positive and negative polarizations with FM were obtained using $\Delta f \approx 30$ MHz at $f_0^+ = 69.070$ GHz and $f_0^- = 69.540$ GHz, respectively. The gain in maximum polarization due to FM is 1.7 and the increase in polarizing speed is about two. The homogeneity of the deuteron polarization throughout the target volume was investigated . Two radially superimposed coils measuring polarization at radii of 1.5 cm and 0.5 cm showed a deuteron polarization ratio from the small to the large coil of 1.20 before and 1.06 after applying FM [3]. A study of the deuteron NMR line asymmetry [3] provided us with an upper limit ΔP_D for the spatial variation of P_D. Typical values for ΔP_D were 0.30 without FM and 0.15 with FM which confirmed that FM improves the uniformity of polarization.

The large enhancement has been confirmed recently in our new 2.5 l target [17] where maximum deuteron polarizations of 0.46 and -0.60 were observed using FM. In spite of the improved magnetic field uniformity of 3.10^{-5} of the new target, the optimum $\Delta f/f$ for FM was about $0.5.10^{-3}$ as in the previous target with 10^{-4} field uniformity. Also we find that $\Delta f/f$ is large compared to the target magnetic field inhomogeneity in both targets. We conclude that the mechanism by which FM improves polarization has little to do with the field nonuniformity. For protons FM increased the polarization, typically from 0.75 to 0.85, and to maximum values as high as 0.95 [18].

The existing theory [19, 20, 21] provides a qualitative understanding of the DNP for our target material; however, the large polarization enhancement due to microwave FM may require additional mechanisms. An example is the cross-relaxation within the system of electron spins which has been assumed to be fast. It has been suggested [22] that a slow cross-relaxation may lead to a lack of thermal equilibrium among electron spins and hence to unequal spin temperatures for different nuclei which results in lower nuclear polarization. FM may counteract this effect by increasing the number of electron spins which are saturated.

A possibly related effect is the local depletion of the electron spin packets which has been observed for materials whose EPR lines are broadened by hyperfine interactions when irradiated at fixed frequency. With FM, this local depletion can be avoided and a migration of spin packets occurs towards the wings of the EPR band [23]. This may result in a stronger EPR absorption in the wings.

Since the aim of our experiment was to measure spin-dependent asymmetries in polarized deep inelastic scattering, we did not attempt a more detailed study of the effect of FM on the target polarization. Our observations of the EPR absorption were used as a guide to optimize the parameters of the FM.

In conclusion, we discovered a large increase in deuteron polarization due to frequency modulation which is of great value for our high energy physics experiment. We found that an amplitude of FM of $\approx$ 30 MHz and a frequency of 1 kHz improved the deuteron polarization growth rate by a factor of 2 and resulted in deuteron polarizations as high as 0.60 with improved spatial uniformity over the target volume. Relations of this large FM effect to features of the EPR absorption mechanism were found and may provide useful information to a deeper understanding of dynamic nuclear polarization.

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Figure Caption

Figure 1

Deuteron polarization as a function of time without FM (dark circles) and with FM (open circles). Positive and negative polarizations are shown.

Figure 2

Electron paramagnetic resonance absorption band for the glassy perdeuterated butanol of the SMC polarized target (the dotted line guides the eye). The temperature T_{SP} is derived from the value of a Speer carbon composite resistor located near the material. The measurements were performed at a constant frequency $f_0 = 69.520$ GHz by stepping the magnetic field. The field values H are converted to the equivalent frequencies $f = f_0 H/H_0$ at $H_0 = 2.5\ T$.

Figure 3

Enhancement in the wings of the EPR absorption spectrum observed when the microwave frequency was modulated with an amplitude of 4 MHz at 1 kHz frequency. Figures a and b show the EPR spectra obtained without (dark circles) and with FM (open circles) for the domain of frequency leading to positive (a) and negative (b) polarizations. Figures c and d show the differential effect.

Figure 4

Enhancement of the EPR absorption as a function of the FM frequency for different values of the input microwave power $\dot{Q}_{IN}$. The arrows show the frequencies at which the maximum enhancement is reached. The four curves labelled 1, 2, 3 and 4 were obtained at levels of input power $\dot{Q}_{IN}$ increased successively by a factor 4.

FIGURE 1

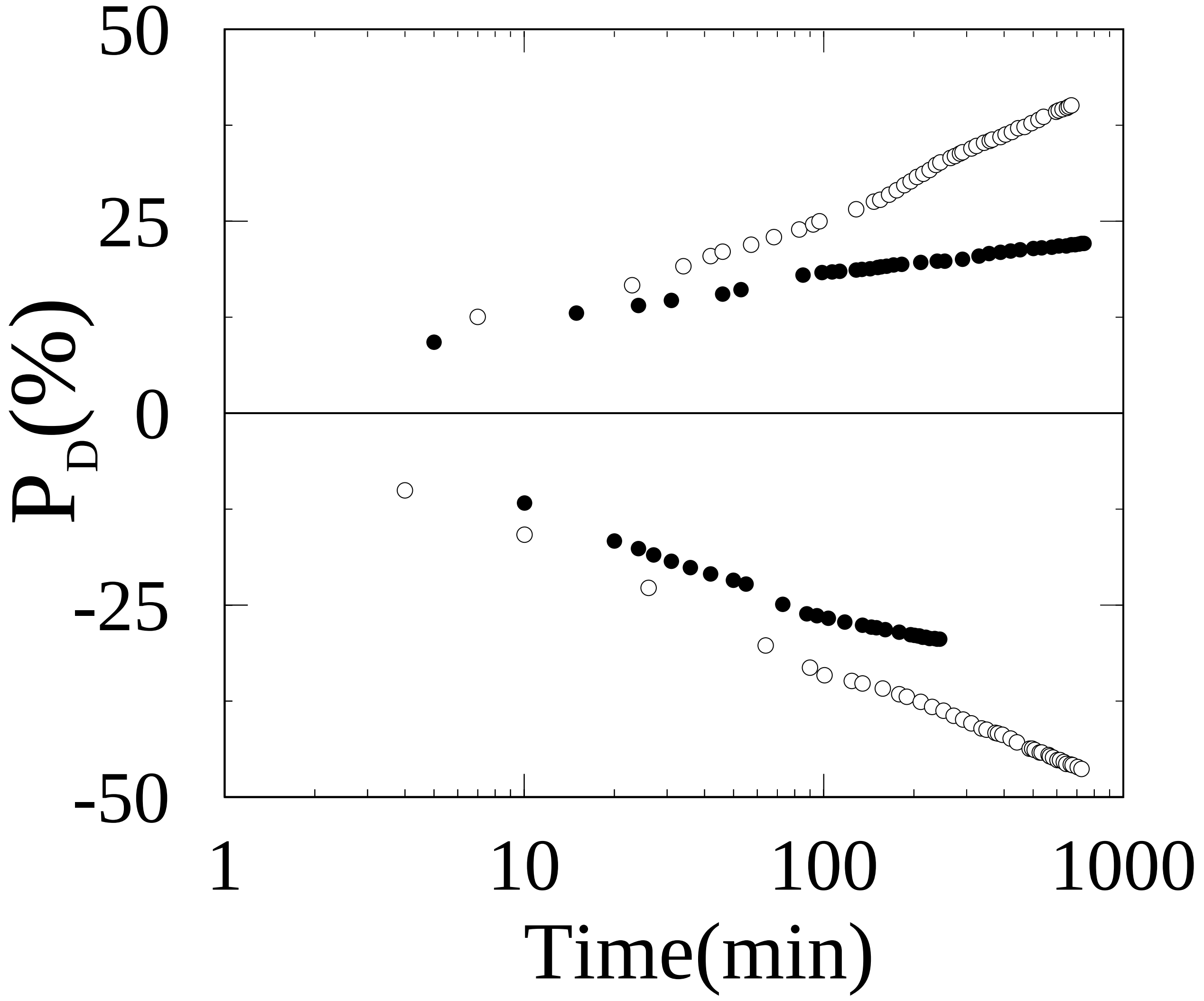


FIGURE 2

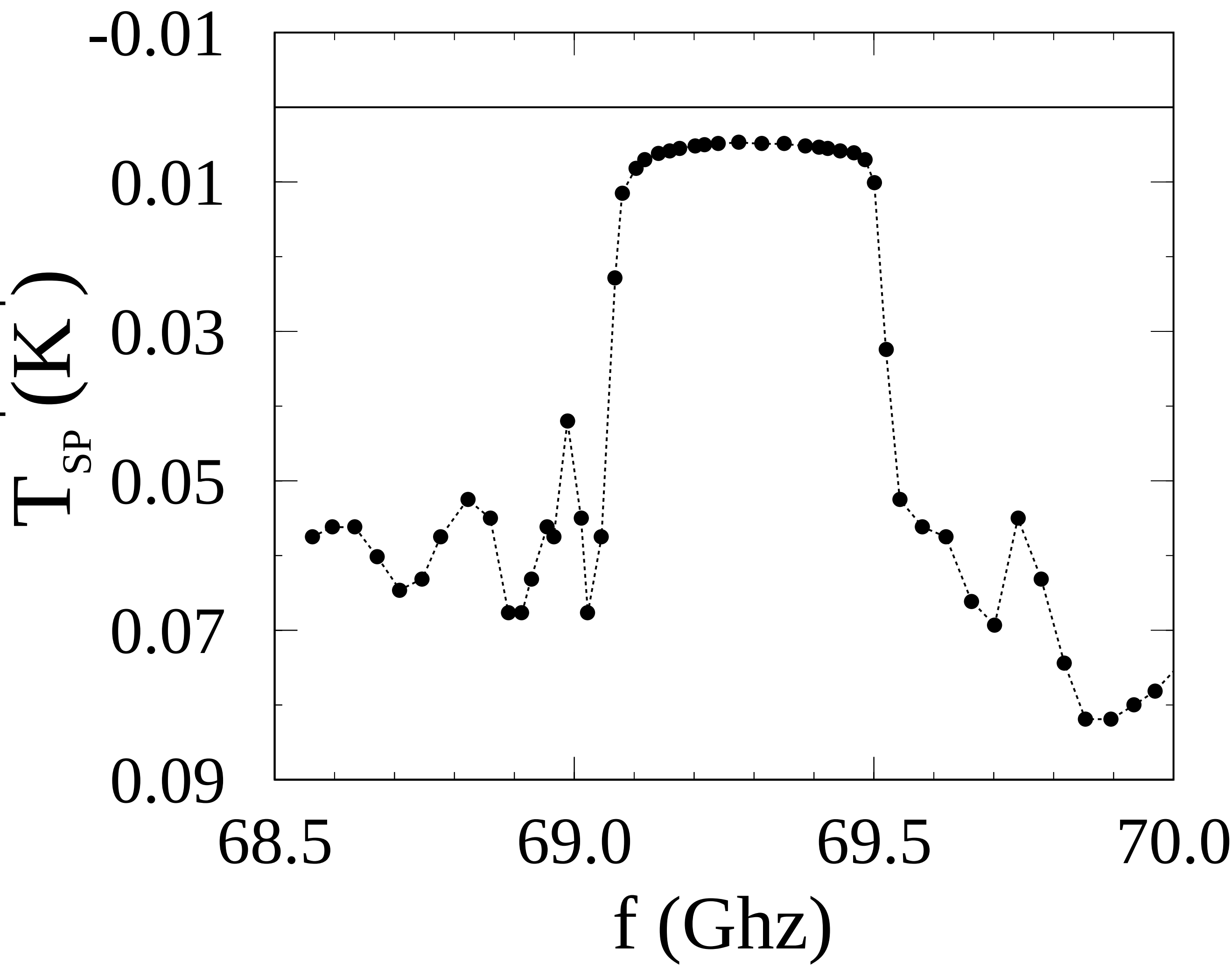
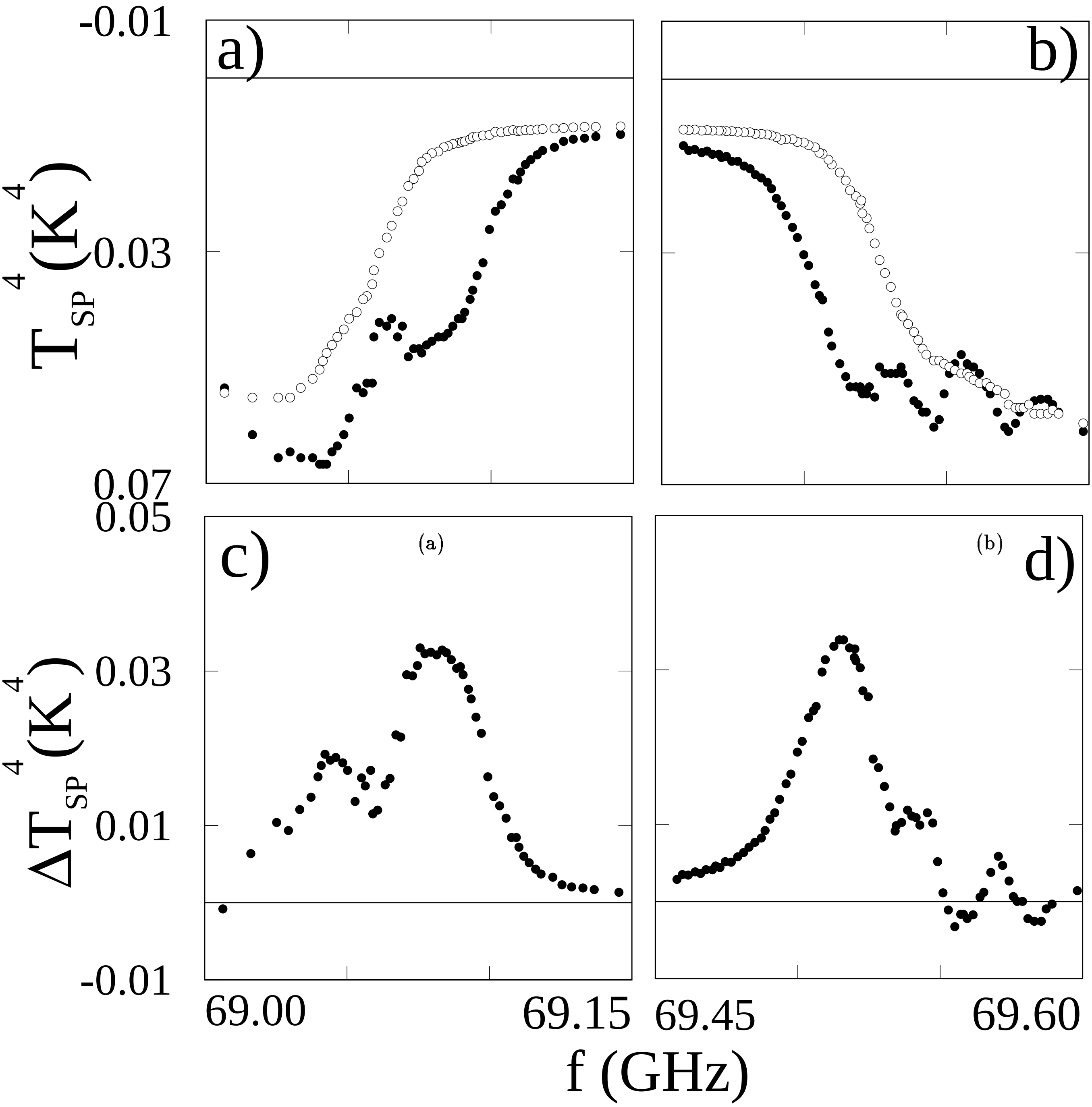
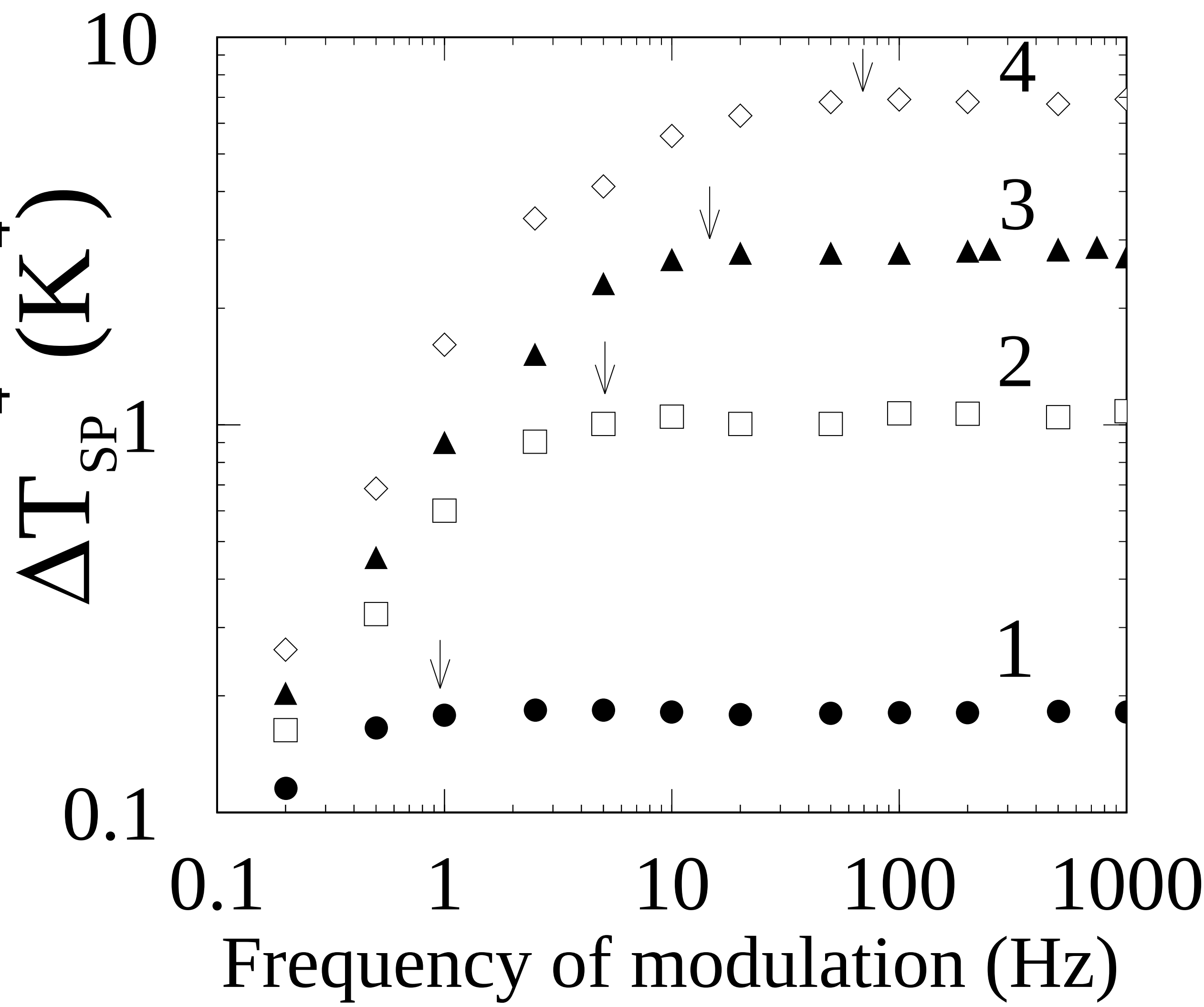


FIGURE 3


-0.01
0.03
0.07
0.05
0.03
0.01
-0.01
a)
b)
c)
d)
(a)
(b)
$T_{SP}^4 (K^4)$
$\Delta T_{SP}^4 (K^4)$
69.00
69.15
69.45
69.60
f (GHz)

(c) (d)

FIGURE 4


10
1
0.1
0.1
1
10
100
1000
Frequency of modulation (Hz)
ΔT_SP^4 (K^4)
4
3
2
1