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ENHANCEMENT OF NUCLEAR POLARIZATION WITH FREQUENCY MODULATED MICROWAVES

The Spin Muon Collaboration (SMC)

Abstract

We report our discovery of a large increase in deuteron polarization up to values of about 0.5 due to frequency modulation of the polarizing microwaves in a 2 l polarized target using the method of dynamic nuclear polarization, 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 the new phenomenon is not quantitatively understood, it may provide a useful testing ground for the deeper understanding of dynamic nuclear polarization in polarized targets.

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Measurements of deep inelastic scattering of polarized muons off polarized protons and deuterons determines the spin dependent structure functions of the nucleon which allow fundamental tests of quantum chromodynamics and of models of nucleon structure [1]. Our discovery of a large enhancement of our deuteron target polarization occurred during the data-taking for deep inelastic scattering [2] and was associated with the failure of a regulator circuit on the high voltage power supply for the microwave source. After controllable frequency modulation (FM) on the microwave tube was implemented, a gain by a factor of 1.7 in the maximum deuteron polarization and of 2.0 in the polarization growth rate was achieved. These increases have been of crucial importance to our experiment at CERN because data-taking extends over many months and the statistical error is proportional to $1/PN^{1/2}$, in which P is the target polarization and N is the number of scattered events. Similar although less dramatic effects due to microwave frequency modulation were observed for our polarized proton target [3].

In order to study the FM effect extensive measurements have been made of the electron paramagnetic resonance (EPR) absorption spectra by a differential bolometric technique. We present these observations below and discuss briefly effects which may contribute to the FM phenomenon.

The polarized target [4, 5] consists of two cells 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 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 $\approx 10^{-4}$ over the 2 l 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 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 due to the calibration error is 5% and the reproducibility is $\delta\mathrm{P} = 10^{-3}$ after averaging 400 signals [9].

The microwave power for DNP is produced by two extended interaction oscillator (EIO) klystrons 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 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 [10, 11] 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 [12]. In the glassy alcohol materials with Cr(V) complexes, where the dynamic nuclear cooling [13] is the dominant mechanism for DNP, hole burning is not expected. FM has been applied in tests of small samples of such materials and provided noticeable improvement in the final polarization only when a low (< 10%) final DNP was obtained [14]. FM has been used also to compensate for magnetic field inhomogeneity thereby improving the final polarization by less than 5% [15].

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The large polarization enhancement in our target due to FM came therefore as a surprise. It was first produced by the instability of the high voltage which modulated the 69 GHz microwave source at 1.2 kHz frequency with a 20 MHz amplitude and led to a relative increase in deuteron polarization by about 50% . After implementing controllable cathode voltage modulation circuits, FM was routinely used. Figure 1 shows the typical time evolution of the deuteron polarization $P_D = < I_z > / I$ with and without FM. The maximum deuteron polarizations obtained with FM 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 (SP) composite carbon resistor as a bolometer, located in the dilute phase of the mixing chamber outside the target material [16]. 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 [17]. 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 structures in the wings of the spectrum are not well understood. 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 $\Delta T_{SP}^4 = (T_{SP}^{\mathrm{off}})^4 - (T_{SP}^{\mathrm{on}})^4 = (\dot{Q}_{MAT}^{\mathrm{on}} - \dot{Q}_{MAT}^{\mathrm{off}})/c$. 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 of ≈ 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 dP_D/dt 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 dP_D/dt 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 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 was investigated throughout the target volume. Two radially superimposed coils measuring polarization at different radii showed a deuteron polarization ratio of 1.20 before and 1.06 after applying FM [9]. A study of the deuteron NMR line asymmetry [9] allowed us evaluate the inhomogeneity of polarization which was reduced by a factor of more than 2 when using FM. We conclude that FM leads to more uniform polarization.

All the above results have been confirmed recently in the new 2.5 l target [18] where maximum deuteron polarizations of 0.46 and -0.60 were observed using FM. For protons the FM increased polarization, typically from 0.75 to 0.85, and to maximum values as high as 0.95 [3]. The increase in the polarization growth rate was of about 20%.

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 detailed study of the effect of FM on the target polarization. Our observations of the EPR absorption were used primarily as a guide to optimize the parameters of the FM. Since some of the results reported here may be related to the size of our target, additional studies of smaller samples should also be undertaken.

In conclusion, we discovered a large increase in deuteron polarization and a smaller increase in proton polarization due to frequency modulation which were of great value for our high energy experiment. We found that using an amplitude of FM of ≈ 30 MHz and a frequency of 1 kHz improved the deuteron polarization growth rate by a factor 2 and resulted in a record of deuteron polarization which exceeded $P_D = 50\%$ along with a much improved spatial uniformity over the volume of the largest existing target. Relations of this new FM phenomenon to features of the EPR absorption mechanism were found and may provide useful information for a deeper understanding of dynamic nuclear polarization.

References

[1] V.W. Hughes and J. Kuti, Ann. Rev. Nucl. Part. Sci. **33** (1983) 611.

[2] Spin Muon Collaboration, B. Adeva et al., Phys. Lett. **B 302** (1993) 533.

[3] Spin Muon Collaboration, D. Adams et al., Phys. Lett. **B 329** (1994) 395.

[4] T. Niinikoski Nucl. Inst. Meth. **192** (1982) 151.

[5] S. C. Brown et al., in Proceedings of the 4th Int. Workshop on Polarized Target Material and Techniques, Bad Honnef (Germany), edited by W. Meyer (3-6 Sept. 1984) p. 102.

[6] M. Krumpolc, D. Hill and H.B. Stuhrmann, in Proceedings of the 9th Int. Symposium on High Energy Spin Physics, volume 2 Bonn (Germany), edited by W. Meyer, E. Steffens and W. Thiel (6-15 Sept. 1990) p. 340.

[7] S.Bültmann. 7th Workshop On Polarized Target Materials and Techniques, Bad Honnef (Germany), 3-6 June 1994, to be published in Nucl. Inst. Meth.

[8] G.R. Court et al., Nucl. Inst. Meth. **A324** (1993) 443.

[9] Spin Muon Collaboration, B. Adeva et al., Nucl. Inst. Meth. **A349** (1994) 334.

[10] A. Abragam and M. Goldman, Rep. Prog. Phys. **41** (1978) 395.

[11] T. J. Schmugge and C. D. Jeffries, Phys. Rev. **138** (1965) A1785.

[12] H.W. van Kesteren, W.Th. Wenckebach and J. Schmidt, Chem. Phys. Lett. **121** (1985) 440.

[13] A. Abragam and M. Borghini, *Dynamic Polarization of Nuclear Targets*, in Progress in Low-Temperature Physics, Ed. C. J. Gorter (Interscience Publ., 1964) p. 384.

[14] D. Hill and M. Krumpolc, in Proceedings of the 2nd Workshop on Polarized Target Material, Rutherford Laboratory, RL-80-080 (1980) p.66.

[15] S. Ishimoto et. al., Jap. J. Appl. Phys. **28** (1989) 1963.

[16] T.O. Niinikoski and J.M. Rieubland, Phys. Lett. **72A** (1979) 141.

[17] O.V. Lounasmaa, *Experimental Principles and Methods Below 1K*, (Academic Press, London 1974) p. 265.

[18] J. Kyynäräinen. 7th Workshop On Polarized Target Materials and Techniques, Bad Honnef, Germany, 3-6 June 1994, to be published in Nucl. Inst. Meth.

[19] B.N. Provotorov, Soviet Phys. JETP **14** (1962) 1126.

[20] B.N. Provotorov, Soviet Phys. JETP **15** (1962) 611.

[21] M.A. Kozhushner and B.N. Provotorov Soviet Phys. Solid State **6** (1964) 1152.

[22] T.O. Niinikoski, 10th International Symposium on High Energy Spin Physics, Nagoya, Japan, 9-14 November (1992).

[23] M. Goldman et al., J. Physique **30** (1969) 427.

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

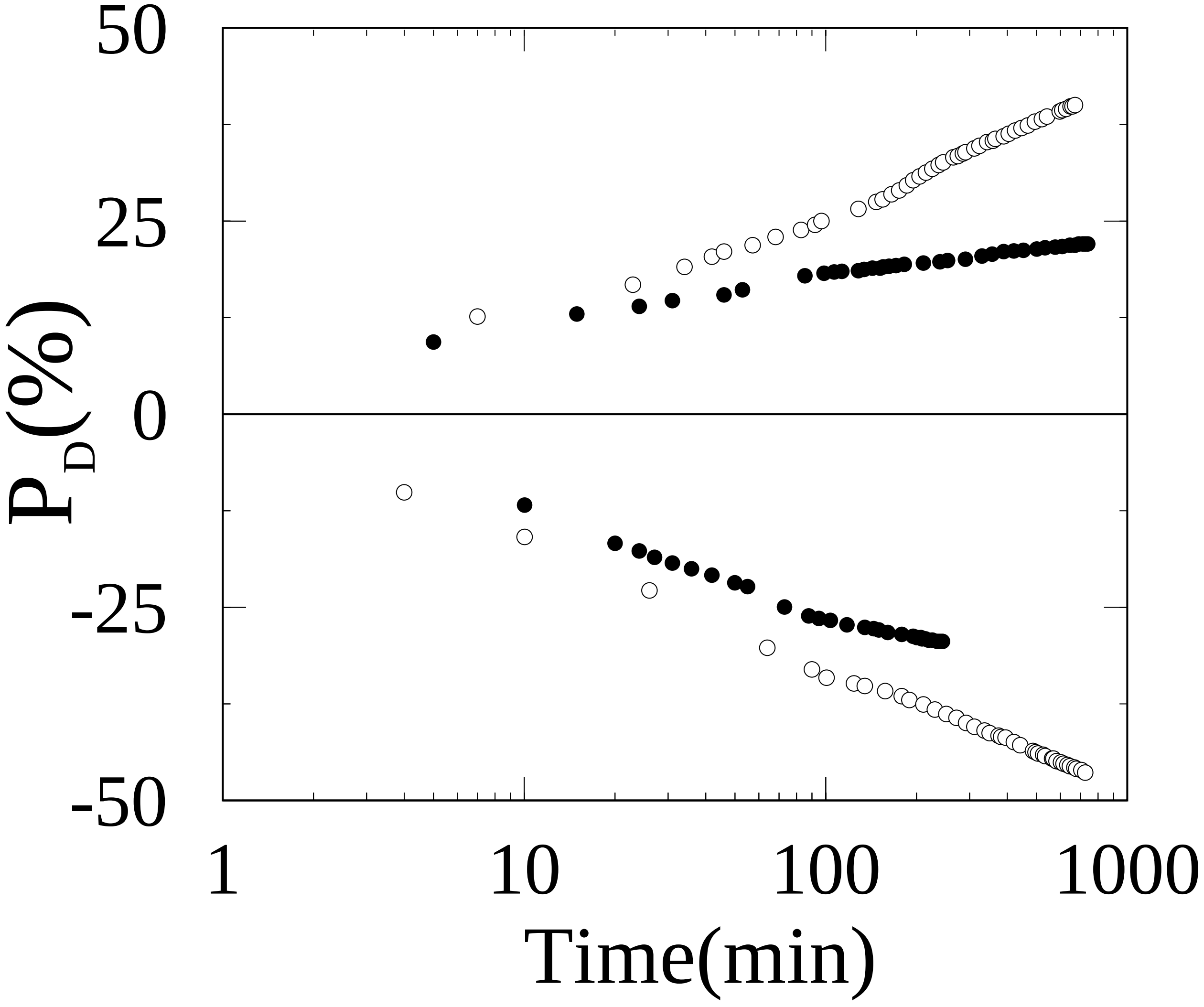


FIGURE 2

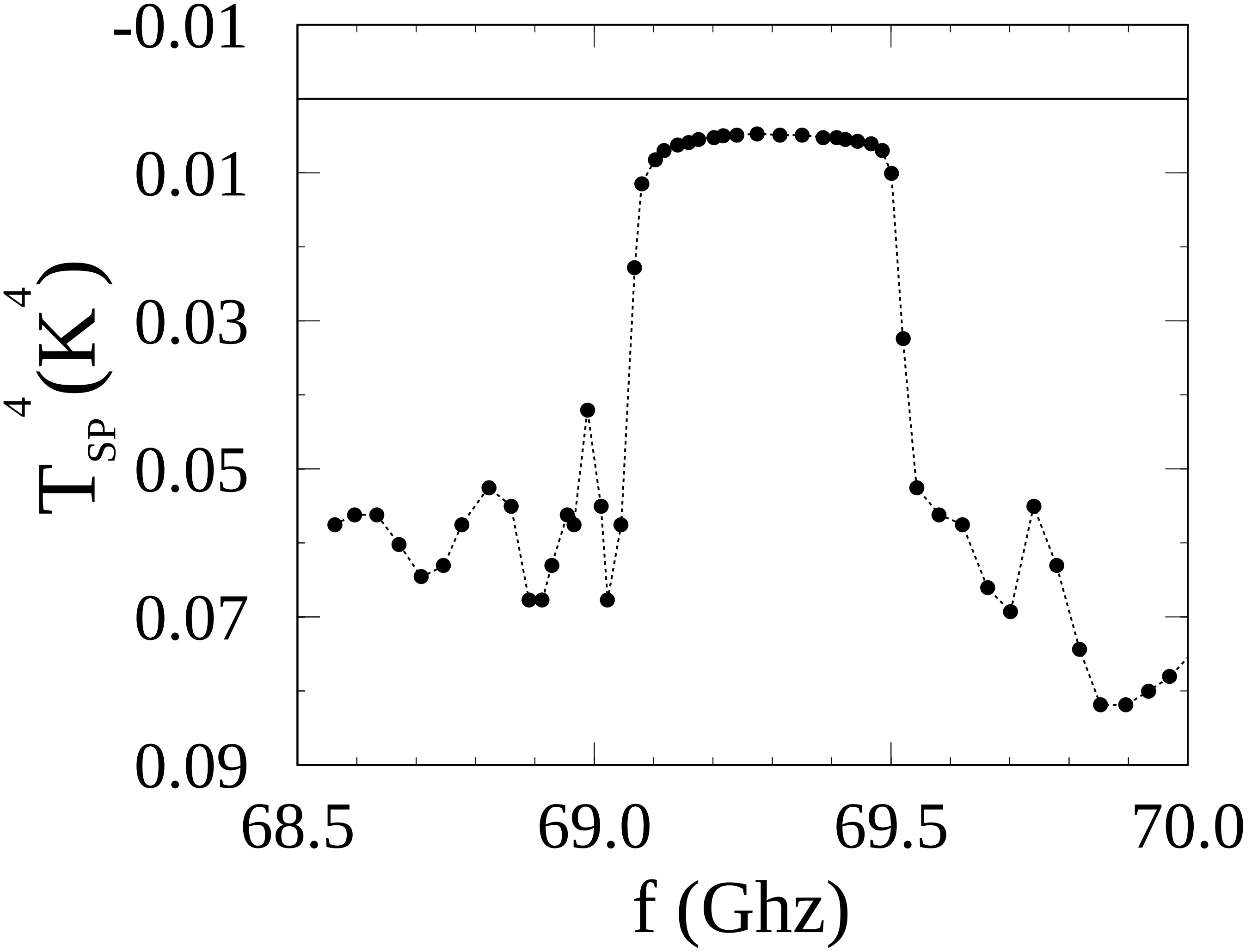
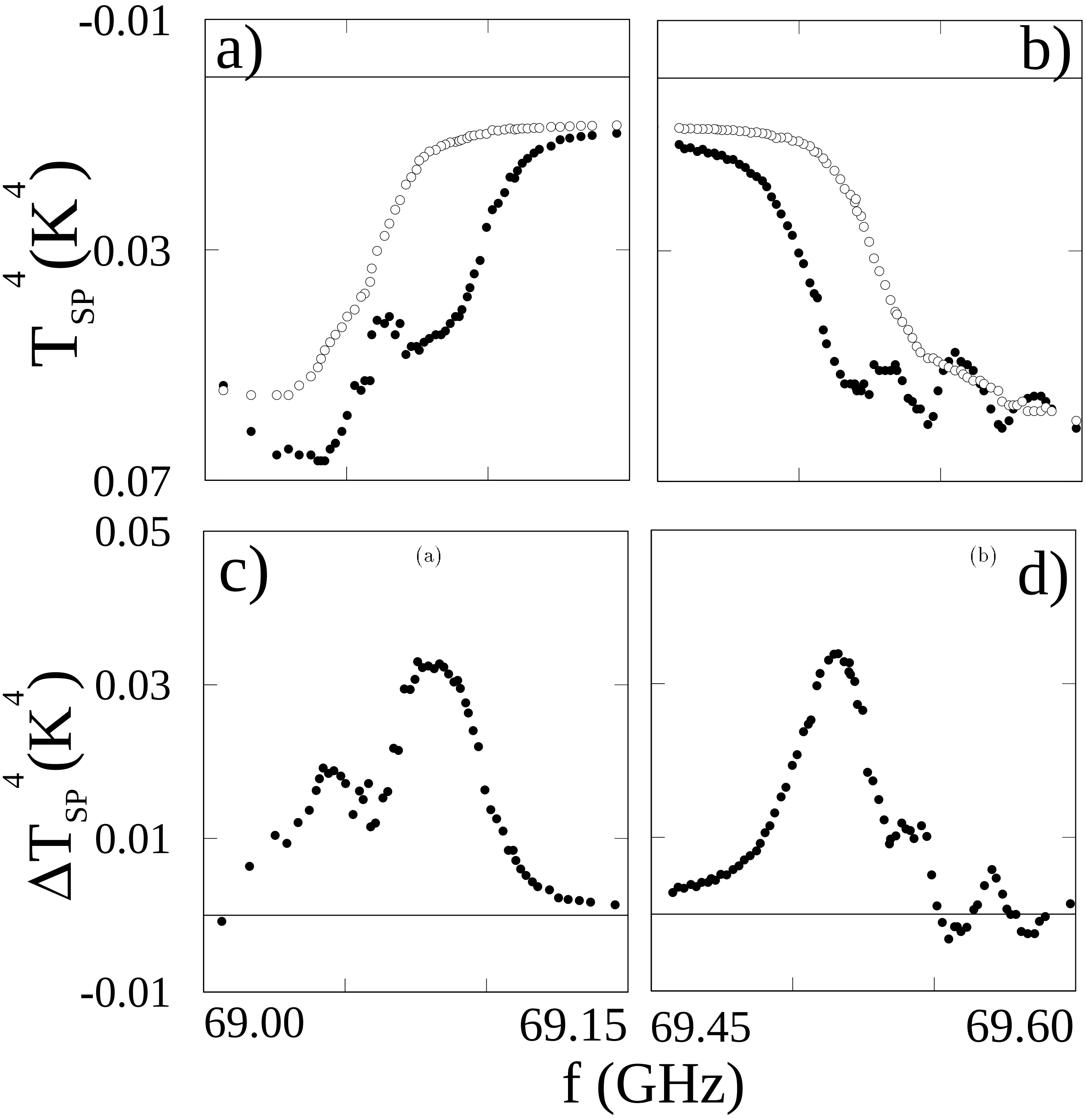


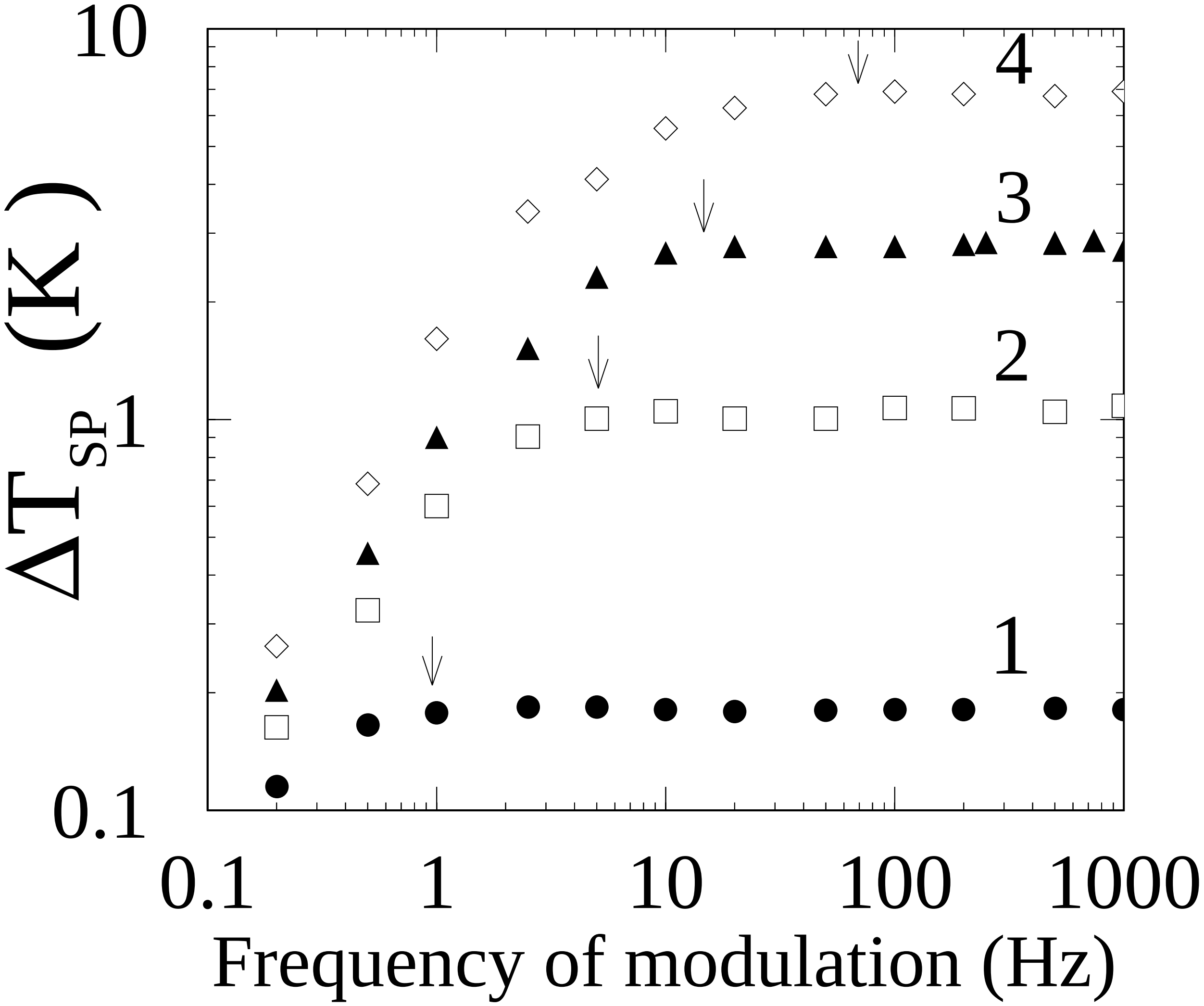
FIGURE 3



(a) (b)

(c) (d)

FIGURE 4


10
1
0.1
0.1
1
10
100
1000
Frequency of modulation (Hz)
$\Delta T_{SP}^4 (K^4)$
4
3
2
1