## <sup>3</sup>He/<sup>129</sup>Xe Clock Comparison Experiment: Search for Spin-Dependent Short-Range Interaction

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We report on the search for a new spin–dependent interaction  $\vec{\sigma} \cdot \hat{r}$  which causes a shift in the precession frequency of nuclear spin polarized gases. Therefore we use a comagnetometer that is based on the detection of freely precessing nuclear spins from <sup>3</sup>He and <sup>129</sup>Xe gas samples using SQUIDs as low–noise magnetic flux detectors. As result we could improve the upper bounds of the dimensionless product  $g_s g_p$  of the monopole–dipole coupling of an axion to the spin of a bound neutron in the mass range between 2  $\mu$ eV and 500  $\mu$ eV (corresponding to force ranges between  $3 \cdot 10^{-4}$  m and  $10^{-1}$  m) by up to 4 orders of magnitude.

The existence of a new spin-dependent short-range force may be a signature of pseudoscalar boson particles like the axion, which was proposed by Peccei and Quinn to solve the strong CPproblem [1]. This hypothetical particle could have been created in early stages of the universe and since it is a light and weak interacting particle, it is an attractive candidate to the cold dark matter that could compose up to 1/3 of the ingredients of the universe [2]. An axion or any axion-like particle mediates an interaction between a fermion f and the spin of another fermion  $f_{\sigma}$  which in case of monopole-dipole coupling violates parity and time symmetries. The Yukawa-type potential of this monopole-dipole interaction with range  $\lambda$  is given by [3]

$$V_{\rm sp}(r) = \frac{\hbar^2 g_s^f g_p^{f\sigma}}{8\pi m} (\vec{\sigma} \cdot \hat{r}) \left(\frac{1}{\lambda r} + \frac{1}{r^2}\right) e^{-r/\lambda} \quad , \tag{1}$$

where  $g_s^f$  and  $g_p^{f\sigma}$  are dimensionless scalar and pseudoscalar constants for the axion-fermion vertices, which in our case correspond to the scalar coupling of an axion or an axion-like particle to a nucleon  $(g_s^f = g_s^N)$  and its pseudoscalar coupling to a polarized bound neutron  $(g_p^{f\sigma} = g_p^n)$ .  $\hat{r}$  is the unit distance vector pointing from the polarized fermion to the unpolarized fermion, respectively from the polarized sample to the unpolarized sample.  $\lambda$  is the range of the Yukawa force with  $\lambda = \hbar/m_a c$ ,  $m_a$  is the mass of the axion and m is the mass of the fermion which carries the spin  $\sigma$ . The potential given by Eq. 1 effectively acts near the surface of a massive unpolarized sample  $(r \leq \lambda)$  as a pseudo-magnetic field and gives rise to a shift  $\Delta\omega_{\rm sp} = 2\pi\Delta\nu_{\rm sp} = V_{\Sigma}/\hbar$ in the precession frequency of nuclear spin polarized gases. The potential  $V_{\Sigma}$  is obtained by integration of  $V_{\rm sp}(r)$  from Eq. 1 over the volume of the massive unpolarized sample averaged over the volume of the polarized sample.

Our approach to search for non-magnetic spin-dependent interactions is to use an ultrasensitive low-field comagnetometer which is based on simultaneous detection of free spin precession of gaseous, nuclear spin polarized <sup>3</sup>He and <sup>129</sup>Xe atoms. The Larmor frequencies of helium and xenon in a constant magnetic guiding field *B* are given by  $\omega_{L,He(Xe)} = \gamma_{He(Xe)} \cdot B$ , whereby  $\gamma_{He(Xe)}$  are the gyromagnetic ratios of the according gas species. Hence, the influence of ambient magnetic fields cancels in the weighted difference of Larmor frequencies, respectively, the corresponding time integral, the weighted difference of Larmor phases

$$\Delta \omega = \omega_{\rm He} - \frac{\gamma_{\rm He}}{\gamma_{\rm Xe}} \cdot \omega_{\rm Xe} = 0 , \quad \Delta \Phi(t) = \Phi_{\rm He}(t) - \frac{\gamma_{\rm He}}{\gamma_{\rm Xe}} \cdot \Phi_{\rm Xe}(t) = const .$$
 (2)

For the gyromagnetic ratios of helium and xenon we took the literature values given by  $\gamma_{\text{He}}/\gamma_{\text{Xe}} = 2.75408159(20)$  [5, 6]. The quantities  $\Delta \omega$  and  $\Delta \Phi(t)$  are sensitive to anomalous frequency shifts due to non-magnetic spin interactions. According to the Schmidt model [4], in the nuclei of helium and xenon the spin of 1/2 is carried by a neutron only. Thus, the frequency shift  $\Delta \nu_{\text{sp}}$  due to the spin-dependent short-range force (Eq. 1) is expected to be similar for <sup>3</sup>He and <sup>129</sup>Xe. Hence, the frequency shift  $\Delta \nu_{\text{sp}}$  does not drop out in the the weighted frequency, respectively, weighted phase difference (Eq. 2). A detailed description of this comagnetometer can be found in [7].

## **1** Experimental Setup and Principle of Measurements

The experiment was performed inside the magnetically shielded room BMSR-2 at the Physikalisch Technische Bundesanstalt Berlin (PTB). BMSR-2 has a passive shielding factor, which exceeds  $10^8$  above 6 Hz. A homogeneous guiding magnetic field of about 350 nT - with maximum field gradients of about 33 pT/cm - was provided inside the shielded room by means of a square coil pair ( $B_x$ -coils). With a second square coil pair ( $B_y$ -coils) which was arranged perpendicular to the first one it was possible to manipulate the precession of the spin samples, e.g.,  $\pi/2$ spin flip by nonadiabatic switching [7]. For detection of the spin precession we used a low- $T_c$  DC-SQUID vector magnetometer system [8, 9]. The pick-up coils of the SQUIDs were sensitive to the vertical magnetic field component of the two precessing nuclear spin species  $(^{3}\text{He}, ^{129}\text{Xe})$ . The system noise of the SQUIDs was about 2.3 fT/ $\sqrt{\text{Hz}}$  in the range of the precession frequencies. Most part of the environmental noise was caused by dewar vibrations relative to the  $B_x$ -coils. Here, combining two SQUIDs to a gradiometer helps to suppress this effect down to a level of  $2.5 \,\mathrm{fT}/\sqrt{\mathrm{Hz}}$ . As gas container we used a cylindrical cell made of aluminosilicate glass (GE180) with a diameter of 58 mm and a length of 60 mm.  ${}^{3}$ He and  ${}^{129}$ Xe were nuclear spin polarized outside the shielding by means of optical pumping. Afterwards the cell was filled with a mixture of polarized <sup>3</sup>He, polarized <sup>129</sup>Xe ( $\approx 2 \text{ mbar}$ ,  $\approx 8 \text{ mbar}$ ) and N<sub>2</sub>  $(\approx 35 \text{ mbar})$ . The nitrogen was added to suppress xenon relaxation due to the van der Waals molecular coupling. After transportation of the measurement cell into the BMSR-2 the cell was mounted directly beneath the dewar which houses the SQUIDs. A cylindrical glass tube with a length of 1 m and an inner diameter of 60 mm was placed on a separate support with its axis aligned with the axis of the cylindrical measurement cell. At its open end towards the polarized sample cell a BGO–crystal (BGO =  $Bi_4Ge_3O_{12}$ ,  $\rho$ =7.13 g/cm<sup>3</sup>, diameter 60 mm and length 70 mm) was installed. The tube and with it the BGO–crystal could be moved horizontally from "close"-position to "distant"-position and vice versa, whereby the movement of the glass tube was possible without opening the door of the magnetic shielded room, i.e., without interruption of the <sup>3</sup>He and <sup>129</sup>Xe spin precession. At "close"–position we had a minimum gap of 2.2 mm between the BGO-crystal and the polarized gases. The BGO-crystal was used, since a nonconducting material prevented us from additional noise sources.

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If the spin-dependent axion fermion interaction exist it will cause a shift  $\Delta \omega_{sp}^w$  in the weighted precession frequency difference (Eq. 2) when the BGO-crystal is close ("close"-position) to the measurement cell. If the BGO-crystal is far away ("distant"-position) from the measurement cell this frequency shift should vanish. To measure this frequency shift  $\Delta \omega_{sp}^w$  the measurement procedure was as follows: The spin precession signals were measured whereby the BGO-crystal was installed close to the cell ("close"-position). After 10800 s the BGO-crystal was moved away ("distant"-position). The spin precession signals were then measured again for about 21500 s. It was also possible to do measurements where the BGO-crystal was first in the "distant"-position and in the second part of the measurement in the "close"-position. These measurements were repeated several times whereby for systematic checks, the BGO-crystal could be placed left and right with respect to the <sup>3</sup>He/<sup>129</sup>Xe sample cell.

## 2 Data Analysis and Results

A detailed despription of the data analysis is described in [10, 11]. Briefly, for each measurement run – in total we did 10 runs – the accumulated phases and with it the corresponding phase difference  $\Delta \Phi(t)$  was determined, which is not a constant in time, as Eq. 2 may suggest. Instead higher order effects, as Earth rotation, chemical shift and the generalized Ramsey-Bloch-Siegert-Shift [11], have to be taken into account. These effects can be parameterized by

$$\Delta\Phi(t) = \Phi_0 + \Delta\omega_{\rm lin} \cdot t - \epsilon_{\rm He} \cdot T^*_{2,\rm He} \cdot A_{\rm He} \cdot e^{-\frac{t}{T^*_{2,\rm He}}} + \epsilon_{\rm Xe} \cdot T^*_{2,\rm Xe} \cdot A_{\rm Xe} \cdot e^{-\frac{t}{T^*_{2,\rm Xe}}} .$$
(3)

If the pseudoscalar Yukawa potential  $V_{\rm sp}(r)$  would occur at an instant  $t = t_0$  due to the movement of the BGO-crystal, an additional linear phase drift  $\Delta \omega_{\rm sp}^w \cdot (t - t_0) = 2\pi \cdot \Delta \nu_{\rm sp}^w \cdot \Theta(\pm (t - t_0)) \cdot (t - t_0)$  in Eq. 3 is expected<sup>1</sup>. The frequency shift  $\Delta \nu_{\rm sp}$  is then extracted from

$$\Delta \nu_{\rm sp} = \frac{\Delta \omega_{\rm sp}^w}{2\pi \cdot \left(1 - \frac{\gamma_{\rm He}}{\gamma_{\rm xe}}\right)} \ . \tag{4}$$

For each measurement run  $\Delta \nu_{\rm sp}$  was determinded. From the calculation of the weighted mean, one gets  $\overline{\Delta \nu}_{\rm sp} = (-2.9 \pm 2.3 \pm 0.1)$  nHz. The last value corresponds to the systematic error<sup>2</sup>. The  $\chi^2/d.o.f$  of the data to their weighted mean  $\overline{\Delta \nu}_{\rm sp}$  gives 2.29, indicating that the errors on the measured frequency shifts are somewhat underestimated. In order to take this into account, the errors were scaled to obtain a  $\chi^2/d.o.f$  of one, as recommended, e.g., by [12, 13]. At the 95% C.L., our result for the measured frequency shift is

$$\overline{\Delta\nu}_{\rm sp} = (-2.9 \pm 6.9 \pm 0.2) \,\mathrm{nHz} \,.$$
 (5)

The result of  $\overline{\Delta\nu}_{sp}$  indicates that we find no evidence for a pseudoscalar short–range interaction mediated by axions or axion–like particles.

From the total error  $\delta(\overline{\Delta\nu_{sp}}) = \pm 7.1 \text{ nHz}$  exclusion bounds for  $|g_s^N g_p^n|$  can be derived by calculating  $V_{\Sigma}$  and using  $|\delta(\overline{\Delta\nu_{sp}})| \geq 2 \cdot V_{\Sigma}/h$ . The results are shown in Fig.1. We substantially improved the bounds on a spin-dependent short-range interaction between polarized (bound) neutrons and unpolarized nucleons over most of the axion window. Existing constraints on axions or axion-like particles heavier than 20  $\mu$ eV could be tightened by up to four orders of magnitudes.

 $<sup>^{1}(\</sup>pm)$  in the argument of the Heaviside step function has to be set (-) for the sequence  $c \rightarrow d$  and (+) for the reverse one  $d \rightarrow c$ .

<sup>&</sup>lt;sup>2</sup>A detailed description of the calculation of the systematic error is given in [11].



Figure 1: The experimental 95% confidence upper limit on  $|g_s^N g_p^n|$  plotted versus the range  $\lambda$  of the Yukawa-force with  $\lambda = \hbar/(m_a c)$ . The light gray area indicates the axion window. (1): result of [14], (2): result of [15], (3): result of [16], (4): this experiment ( $\Delta x = 2.2 \text{ mm}$ ), (5): expected results for  $\Delta x \approx 0 \text{ mm}$  using  $\delta(\overline{\Delta \nu_{sp}}) = \pm 7.1 \text{ nHz}$  demonstrates the gain in measurement sensitivity for  $\lambda < 10^{-3} \text{ m}$ . For bounds on the pseudoscalar short–range force between polarized electrons and unpolarized nucleons see [17].

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