Limits on the temporal variation of the fine structure constant, quark masses and 
strong interaction from quasar absorption spectra and atomic clock experiments

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(Dated: March 26, 2022)

We perform calculations of the dependence of nuclear magnetic moments on quark masses and 
and obtain limits on the variation of \( \frac{m_q}{\Lambda_{QCD}} \) from recent measurements of hydrogen hyperfine (21 cm) 
and molecular rotational transitions in quasar absorption systems, atomic clock experiments 
with hyperfine transitions in H, Rb, Cs, Yb\(^+\), Hg\(^+\) and optical transition in Hg\(^+\). Experiments with 
Cd\(^+\), deuterium/hydrogen, molecular SF\(_6\) and Zeeman transitions in \(^3\)He/Xe are also discussed.

PACS numbers: 06.20.Jr, 06.30.Ft, 12.10.-r

I. INTRODUCTION

Interest in the temporal and spatial variation of major constants of physics has been recently revived by 
astrophysical data which seem to suggest a variation of the electromagnetic constant \( \alpha = e^2/\hbar c \) at the \( 10^{-5} \) level for the 
time scale 10 billion years, see [1] (a discussion of other limits can be found in the review [2] and references 
therein). However, an independent experimental confirmation is needed.

The hypothetical unification of all interactions implies that variation of the electromagnetic interaction constant \( \alpha \) 
should be accompanied by the variation of masses and the strong interaction constant. Specific predictions need 
a model. For example, the grand unification model discussed in Ref. [3] predicts that the quantum chromody-
namic (QCD) scale \( \Lambda_{QCD} \) (defined as the position of the Landau pole in the logarithm for the running strong 
coupling constant) is modified as follows: \( \delta \Lambda_{QCD}/\Lambda_{QCD} \approx 34 \delta \alpha/\alpha \). The variation of quark and electron masses in 
this model is given by \( \delta m/m \sim 70 \delta \alpha/\alpha \). This gives an estimate for the variation of the dimensionless ratio

\[
\frac{\delta (m/\Lambda_{QCD})}{(m/\Lambda_{QCD})} \sim 35 \frac{\delta \alpha}{\alpha} \quad (1)
\]

This result is strongly model-dependent (for example, the coefficient may be an order of magnitude smaller 
and even of opposite sign [4]). However, the large coefficients in these expressions are generic for grand uni-
fication models, in which modifications come from high energy scales: they appear because the running strong 
coupling constant and Higgs constants (related to mass) run faster than \( \alpha \). This means that if these models are 
correct the variation of masses and the strong interaction scale may be easier to detect than the variation of \( \alpha \).

One can only measure the variation of dimensionless quantities and therefore we want to extract from the 
measurements the variation of the dimensionless ratio \( m_q/\Lambda_{QCD} \) – where \( m_q \) is the quark mass (with the dependence 
on the renormalization point removed). A number of limits on the variation of \( m_q/\Lambda_{QCD} \) have been ob-
tained recently from consideration of Big Bang Nucleosynthesis, quasar absorption spectra and the Oklo nat-
ural nuclear reactor, which was active about 1.8 billion years ago [5, 6, 7, 8] (see also [9, 10, 11, 12, 13]). Below we 
consider the limits which follow from quasar absorption radio spectra and laboratory atomic clock compar-
isons. Laboratory limits with a time base of the order one year are especially sensitive to oscillatory variations 
of fundamental constants. A number of relevant measurements have been performed already and even larger 
numbers have been started or are planned. The increase in precision is happening very fast.

It has been pointed out by Karshenboim [14] that measurements of ratios of hyperfine structure intervals in diff-
ferent atoms are sensitive to any variation of nuclear magnetic moments. First rough estimates of the dependence 
of nuclear magnetic moments on \( m_q/\Lambda_{QCD} \) and limits on the variation of this ratio with time were obtained in 
Ref. [2]. Using H, Cs and Hg\(^+\) measurements [14], we obtained an upper limit on the variation of \( m_q/\Lambda_{QCD} \) of about 
0.5 \( \times 10^{-13} \) per year. Below we calculate the dependence of nuclear magnetic moments on \( m_q/\Lambda_{QCD} \) and obtain the 
limits from recent atomic clock experiments with hyperfine transitions in H, Rb, Cs, Yb\(^+\), Hg\(^+\) and the optical 
transition in Hg\(^+\). It is convenient to assume that the strong interaction scale, \( \Lambda_{QCD} \), does not vary, so we will 
speak about the variation of masses (this means that we measure masses in units of \( \Lambda_{QCD} \)). We shall restore the 
explicit appearance of \( \Lambda_{QCD} \) in the final answers.

The hyperfine structure constant can be presented in the following form

\[
A = \text{const} \times \left[ \frac{m_e e^4}{\hbar^2} \right] [\alpha^2 F_{ref}(Z\alpha)] \left[ \frac{m_e}{m_p} \right] \quad (2)
\]

The factor in the first bracket is an atomic unit of energy. The second “electromagnetic” bracket determines
the dependence on $\alpha$. An approximate expression for the relativistic correction factor (Casimir factor) for an s-wave electron is the following

$$ F_{\text{rel}} = \frac{3}{\gamma(4\gamma^2 - 1)}, \quad (3) $$

where $\gamma = \sqrt{1 - (Z\alpha)^2}$ and $Z$ is the nuclear charge. Variation of $\alpha$ leads to the following variation of $F_{\text{rel}}$:

$$ \delta F_{\text{rel}} = K \frac{\delta \alpha}{\alpha}, \quad (4) $$

$$ K = \frac{(Z\alpha)^2(12\gamma^2 - 1)}{\gamma^2(4\gamma^2 - 1)}. \quad (5) $$

More accurate numerical many-body calculations of the dependence of the hyperfine structure on $\alpha$ have shown that the coefficient $K$ is slightly larger than that given by this formula. For Cs ($Z=55$) $K = 0.83$ (instead of 0.74), for Rb $K=0.34$ (instead of 0.29) and finally for Hg$^+$ $K=2.28$ (instead of 2.18).

The last bracket in Eq. (2) contains the dimensionless nuclear magnetic moment $\mu$ (i.e., the nuclear magnetic moment $M = \mu \frac{2}{\sqrt{2}m_{\pi}^2}$), electron mass $m_e$ and proton mass $m_p$. We may also include a small correction arising from the finite nuclear size. However, its contribution is insignificant.

Recent experiments measured the time dependence of the ratios of the hyperfine structure intervals of $^{199}$Hg$^+$ and H $^{15}$, $^{133}$Cs and $^{87}$Rb $^{18}$ and the ratio of the optical frequency in Hg$^+$ to the hyperfine frequency of $^{133}$Cs $^{20}$. In the ratio of two hyperfine structure constants for different atoms time dependence may appear from the ratio of the factors $F_{\text{rel}}$ (depending on $\alpha$) as well as from the ratio of nuclear magnetic moments (depending on $m_q/A_{\text{QCD}}$). Magnetic moments in a single-particle approximation (one unpaired nucleon) are:

$$ \mu = (g_s + (2j - 1)g_l)/2, \quad (6) $$

for $j = l + 1/2$.

$$ \mu = \frac{j}{2(j + 1)}(-g_s + (2j + 3)g_l) \quad (7) $$

for $j = l - 1/2$. Here the orbital g-factors are $g_l = 1$ for a valence proton and $g_l = 0$ for a valence neutron. The present values of the spin g-factors, $g_s$, are $g_p = 5.586$ for proton and $g_n = -3.826$ for neutron. They depend on $m_q/A_{\text{QCD}}$. The light quark masses are only about 1% of the nucleon mass ($m_q = (m_u + m_d)/2 \approx 5$ MeV) and the nucleon magnetic moment remains finite in the chiral limit, $m_u = m_d = 0$. Therefore, one might think that the corrections to $g_s$ arising from the finite quark masses would be very small. However, through the mechanism of spontaneous chiral symmetry breaking, which leads to contributions to hadron properties from Goldstone boson loops, one may expect some enhancement of the effect of quark masses $^{12}$. The natural framework for discussing such corrections is chiral perturbation theory and we discuss these chiral corrections next.

II. CHIRAL PERTURBATION THEORY

RESULTS FOR NUCLEON MAGNETIC MOMENTS AND MASSES

In recent years there has been tremendous progress in the calculation of hadron properties using lattice QCD. Moore’s Law, in combination with sophisticated algorithms, means that one can now make extremely accurate calculations for light quark masses ($m_q$) larger than 50 MeV. However, in order to compare with experimental data, it is still necessary to extrapolate quite a long way as a function of quark mass. This extrapolation is rendered non-trivial by the spontaneous breaking of chiral symmetry in QCD, which leads to Goldstone boson loops and, as a direct consequence, non-analytic behaviour as a function of quark mass $^{21}$. Fortunately the most important nonanalytic contributions are model independent, providing a powerful constraint on the extrapolation procedure.

In the past few years the behaviour of hadron properties as a function of quark mass has been studied over a much wider range than one needs for the present purpose $^{22, 23, 24, 25, 26, 27}$. One can therefore apply the successful extrapolation formulas developed in the context of lattice QCD with considerable confidence.

The key qualitative feature learnt from the study of lattice data is that Goldstone boson loops are strongly suppressed once the Compton wavelength of the boson is smaller than the source. Inspection of lattice data for a range of observables, from masses to charge radii and magnetic moments, reveals that the relevant mass scale for this transition is $m_q \sim 50$ MeV – i.e., $m_\pi \sim 400 – 500$ MeV $^{22, 27}$. The challenge of chiral extrapolation is therefore to incorporate the correct, model independent non-analytic behaviour dictated by chiral symmetry while ensuring excellent convergence properties of the chiral expansion in the large mass region, as well as maintaining the model independence of the results of the extrapolation. Considerable study of this problem has established that the use of a finite range regulator (FRR) fulfils all of these requirements $^{30, 31, 32}$. Indeed, in the case of the mass of the nucleon, it has been shown that the extrapolation from $m_\pi^2 \sim 0.25$ GeV$^2$ to the physical pion mass – a change of $m_q$ by a factor of 10 – can be carried out with a systematic error less than 1% $^{31}$. In the following we apply this same method to calculate the change in the nucleon mass, corresponding to quark mass changes at the level of 0.1% or less, as required in the present context.

A. Variation of the nucleon mass with quark mass

The expansion for the mass of the nucleon given in Ref. $^{31, 32}$ is:

$$ M_N = a_0 + a_2 m_\pi^2 + a_4 m_\pi^4 + a_6 m_\pi^6 + \sigma_{N\pi} + \sigma_{\Delta\pi} + \sigma_{\text{tad}}, \quad (8) $$
where the chiral loops which given rise, respectively, to the leading and next-to-leading nonanalytic (LNA and NLNA) behaviour are:

$$\sigma_{N\pi} = -\frac{3}{32 \pi f_\pi^2} g_A^2 I_M(m_\pi, \Delta_{NN}, \Lambda)$$

(9)

$$\sigma_{\Delta\pi} = -\frac{3}{32 \pi f_\pi^2} \frac{32}{29} g_A^2 I_M(m_\pi, \Delta_{NN}, \Lambda)$$

(10)

$$\sigma_{\pi\Delta} = -\frac{3}{16 \pi f_\pi^2 c_2 m_\pi^2} I_T(m_\pi, \Lambda)$$

(11)

and the relevant integrals are defined (in heavy baryon approximation) as:

$$I_M(m_P, \Delta_{BB'}, \Lambda) = \frac{2}{\pi} \int_0^\infty dk \frac{k^4 u^2(k, \Lambda)}{\omega_k (\Delta_{BB'} + \omega_k)}$$

(12)

$$I_T(m_\pi, \Lambda) = \int_0^\infty dk \left( \frac{2k^2 u^2(k)}{\sqrt{k^2 + m_\pi^2}} - t_0 \right)$$

(13)

with $$\omega_k = \sqrt{k^2 + m_\pi^2}$$ and $$\Delta_{BB'}$$ the relevant baryon mass difference (i.e., $$M_{BB'} - M_P$$). We take the $$\Delta-N$$ mass splitting, $$\Delta = M_\Delta - M_N$$, to have its physical value (0.292 GeV), while $$g_A = 1.26$$. The regulator function, $$u(k, \Lambda)$$, is taken to be a dipole with mass $$\Lambda = 0.8$$ GeV.

In Eq. (13) $$t_0$$, defined such that $$I_T$$ vanishes at $$m_\pi = 0$$, is a local counter term introduced in FRR to ensure a linear relation for the renormalisation of $$c_2$$.

The model independence of the expansion given in Eq. (3) is ensured by fitting the unknown coefficients to the physical nucleon mass and lattice data from the CP-PACS Collaboration [33], yielding: $$a_0 = 1.22, a_2 = 1.76, a_4 = -0.829, a_6 = 0.260$$ (with all parameters expressed in the appropriate powers of GeV). With these parameters fixed one can evaluate the rate of change of the mass of the nucleon with quark or pion mass at the physical pion mass:

$$m_q \frac{\partial}{\partial m_q} M_N = m_\pi^2 \frac{\partial}{\partial m_\pi^2} M_N = 0.035 \text{ GeV}$$

(14)

a quantity commonly known as the pion-nucleon sigma commutator. Using Eq. (14) one finds the relationship (in terms of dimensionless quantities):

$$\frac{\delta M_N}{M_N} = \frac{m_\pi^2}{M_N} \frac{\partial M_N}{\partial m_\pi^2} \frac{\delta m_q}{m_q}$$

(15)

$$= 0.037 \frac{\delta m_q}{m_q}$$

(16)

The extension of this procedure to the effect of a variation in the strange quark mass is similar, but one must include the variation arising from $$\eta$$-Nucleon loops, as well as Kaon loops with intermediate $$\Sigma$$ or $$\Lambda$$ baryons.

$$\sigma_{\Sigma\eta} + \sigma_{\Lambda\eta} + \sigma_{\Sigma\Lambda}$$

(17)

These contributions can be expressed as

$$\sigma_{BB'}^{P} = -\frac{3}{32 \pi f_\pi^2} G_{BB'}^{P} I_M(m_P, \Delta_{BB'}, \Lambda)$$

(18)

with $$G_{BB'}^{P}$$ the associated coupling squared. Once again we select the dipole regulator:

$$u(k, \Lambda) = \left( \frac{\Lambda^2}{k^2 + \Lambda^2} \right)^2$$

(19)

For the relevant diagrams, $$N \to \Sigma K$$, $$N \to K \Lambda$$ and $$N \to N\eta$$, we have

$$G_{N\Sigma}^{K} = \frac{1}{3} (D - F)^2$$

$$G_{N\Lambda}^{K} = \frac{1}{9} (3F + D)^2$$

$$G_{NN}^{\eta} = \frac{1}{9} (3F - D)^2$$

(20)

where we take $$F = 0.50$$ and $$D = 0.76$$. We use the Gell Mann-Oakes-Renner relation in the SU(2) chiral limit to relate the variation of the kaon mass in the chiral SU(2) limit, $$m_K = \sqrt{m_K^2 - \frac{1}{2} F_K^2} = 0.484 \text{ GeV}$$ (with $$\mu_{\pi}(K)$$), the physical pion{kaon} mass, to the variation of the strange quark mass ($$\delta m_K^2/m_K^2 = \delta m_s/m_s$$). Hence the variation of the nucleon mass with strange quark mass is given by:

$$\frac{\delta M_N}{M_N} = \left\{ \frac{\delta m_K^2}{M_N} \frac{\partial}{\partial m_K^2} (\sigma_{N\Sigma}^K + \sigma_{N\Lambda}^K + \sigma_{NN}^\eta) \right\} \frac{\delta m_s}{m_s}$$

(21)

Using the dipole regulator mass, $$\Lambda = 0.8$$ GeV, Eq. (21) leads to the result:

$$\frac{\delta M_N}{M_N} = 0.011 \frac{\delta m_s}{m_s}$$

(22)

B. Variation of proton and neutron magnetic moments with quark mass

The treatment of the mass dependence of the nucleon magnetic moments is very similar to that for the masses. Once again the loops which give rise to the LNA and NLNA behaviour are evaluated with a FRR, while the smooth, analytic variation with quark mass is parametrized by fitting relevant lattice data with a finite number of adjustable constants.

For the lattice data we use CSSM Lattice Collaboration results [34] of nucleon 3-point functions. Results are obtained using established techniques in the extraction of form factor data [35]. Similar calculations have also been recently reported by the QCDSF Collaboration [28]. We use the two heaviest simulation results, $$m_s^2 \sim 0.6-0.7 \text{ GeV}^2$$ [34]. These simulations were performed with the FLIC fermion action [36] on a $$20^3 \times 40$$ lattice at $$a = 0.128 \text{ fm}$$.

In the magnetic moment case the formulae are a little more complicated, so we leave the details for the Appendix. Suffice it to say here that the relevant processes are shown in Fig. 3.
moments. For all even Z nuclei with valence neutron

\[ \frac{\delta \mu}{\mu} = \delta m_{q}/m_{q} \]

The numerical results may then be summarised as:

\[ \frac{\delta \mu_{p}}{\mu_{p}} = -0.087 \frac{\delta m_{q}}{m_{q}} \]

\[ \frac{\delta \mu_{p}}{\mu_{p}} = -0.013 \frac{\delta m_{s}}{m_{s}} \]

\[ \frac{\delta \mu_{n}}{\mu_{n}} = -0.118 \frac{\delta m_{q}}{m_{q}} \]

\[ \frac{\delta \mu_{n}}{\mu_{n}} = 0.0013 \frac{\delta m_{s}}{m_{s}} \]

\[ \frac{\delta (\mu_{p}/\mu_{n})}{(\mu_{p}/\mu_{n})} = 0.031 \frac{\delta m_{q}}{m_{q}} \]

\[ \frac{\delta (\mu_{p}/\mu_{n})}{(\mu_{p}/\mu_{n})} = -0.015 \frac{\delta m_{s}}{m_{s}} \].

### III. DEPENDENCE OF ATOMIC TRANSITION FREQUENCIES ON FUNDAMENTAL CONSTANTS

Using the results of the previous section we can now use Eqs. (6,7) to study the variation of nuclear magnetic moments. For all even Z nuclei with valence neutron

\( ^{199}\text{Hg},^{171}\text{Yb},^{111}\text{Cd}, \text{etc.} \) we obtain \( \delta \mu = \delta m_{QCD} \). For \(^{133}\text{Cs} \) we have a valence proton with \( j=\frac{7}{2}, l=4 \) and

\[ \frac{\delta \mu}{\mu} = 0.110 \frac{\delta m_{q}}{m_{q}} + 0.017 \frac{\delta m_{s}}{m_{s}} \]

For \(^{87}\text{Rb} \) we have valence proton with \( j=3/2, l=1 \) and

\[ \frac{\delta \mu}{\mu} = -0.064 \frac{\delta m_{q}}{m_{q}} - 0.010 \frac{\delta m_{s}}{m_{s}} \]

As an intermediate result it is convenient to present the dependence of the ratio of the hyperfine constant, \( A \), to the atomic unit of energy \( E = \frac{m_{e}c^{2}}{\alpha} \) (or the energy of the 1s-2s transition in hydrogen, which is equal to \( 3/8 E \)) on a variation of the fundamental constants. We introduce a parameter \( V \) defined by the relation

\[ \frac{\delta V}{V} = \frac{\delta (A/E)}{A/E} \]

We start from the hyperfine structure of \(^{133}\text{Cs} \) which is used as a frequency standard. Using Eqs. (31,32) we obtain

\[ V^{(133}\text{Cs}) = \alpha^{2.83}(\frac{m_{q}}{\Lambda_{QCD}})^{0.110}(\frac{m_{s}}{\Lambda_{QCD}})^{0.017}\frac{m_{e}}{m_{p}} \]

The factor \( \frac{m_{e}}{m_{p}} \) will cancel out in the ratio of hyperfine transition frequencies. However, it will survive in comparison between hyperfine and optical or molecular transitions (see below). According to Eqs. (33) and (34), the relative variation of the electron to proton mass ratio can be described by the parameter

\[ X(m_{e}/m_{p}) = (\frac{m_{q}}{\Lambda_{QCD}})^{-0.037}(\frac{m_{s}}{\Lambda_{QCD}})^{-0.011}\frac{m_{e}}{\Lambda_{QCD}} \]

which can be substituted into Eq. (35) instead of \( m_{e}/m_{p} \). This gives an expression which is convenient to use for comparison with optical and molecular vibrational or rotational transitions

\[ V^{(133}\text{Cs}) = \alpha^{2.83}(\frac{m_{q}}{\Lambda_{QCD}})^{0.073}(\frac{m_{s}}{\Lambda_{QCD}})^{0.006}\frac{m_{e}}{m_{p}} \]

The dependence on the strange quark mass is relatively weak. Therefore, it may be convenient to assume that the relative variation of the strange quark mass is the same as the relative variation of the light quark masses (this assumption is motivated by the Higgs mechanism of mass generation) and to use an approximate expression

\[ V^{(133}\text{Cs}) \approx \alpha^{2.83}(\frac{m_{q}}{\Lambda_{QCD}})^{0.13}\frac{m_{e}}{m_{p}} \]

For hyperfine transition frequencies in other atoms we obtain

\[ V^{(87}\text{Rb}) = \alpha^{2.34}(\frac{m_{q}}{\Lambda_{QCD}})^{-0.064}(\frac{m_{s}}{\Lambda_{QCD}})^{-0.010}\frac{m_{e}}{m_{p}} \]

\[ V^{(1}\text{H}) = \alpha^{2}(\frac{m_{q}}{\Lambda_{QCD}})^{-0.087}(\frac{m_{s}}{\Lambda_{QCD}})^{-0.013}\frac{m_{e}}{m_{p}} \]
\[ V(Hg) = \frac{V(Cs)}{V(Rb)} = \frac{1}{X(Cs/Rb)}\frac{dX(Cs/Rb)}{dt} = (0.2 \pm 7) \times 10^{-16}/\text{year}. \] (46)

Note that if the relation (14) were correct, the variation of \(X(Cs/Rb)\) would be dominated by variation of \(m_q/\Lambda_{QCD}\). The relation (14) would give \(X(Cs/Rb) \propto \alpha^8\).

For \(A^{133}\text{Cs}^4)/A(H)\) we have
\[ X(Cs/H) = \frac{V(Cs)}{V(H)} = \alpha^{0.83}[m_q/\Lambda_{QCD}]^{0.196}[m_s/\Lambda_{QCD}]^{0.030} \] (47)
and the result of the measurements in Ref. [16] may be presented as
\[ \left| \frac{1}{X(Cs/H)} \frac{dX(Cs/H)}{dt} \right| < 5.5 \times 10^{-14}/\text{year}. \] (48)

For \(A^{199}\text{Hg})/A(H)\) we have
\[ X(Hg/H) = \frac{V(Hg)}{V(H)} \approx \alpha^{2.3}[m_q/\Lambda_{QCD}]^{-0.031}[m_s/\Lambda_{QCD}]^{0.015}. \] (49)

The result of the measurement in Ref. [17] may be presented as
\[ \left| \frac{1}{X(Hg/H)} \frac{dX(Hg/H)}{dt} \right| < 8 \times 10^{-14}/\text{year}. \] (50)

Note that because the dependence on masses and strong interaction scale is very weak here, this experiment may be interpreted as a limit on the variation of \(\alpha\).

In Ref. [17] a limit was obtained on the variation of the ratio of hyperfine transition frequencies \(171\text{Yb}^+/^{133}\text{Cs}\) (this limit is based on the measurements of Ref. [18]). Using Eqs. (34, 11, 11) we can present the result as a limit on \(X(Yb/Cs) = \alpha^{0.7}[m_q/\Lambda_{QCD}]^{-0.228}[m_s/\Lambda_{QCD}]^{-0.015}:\)
\[ \left| \frac{1}{X(Yb/Cs)} \frac{dX(Yb/Cs)}{dt} \right| \approx -1(2) \times 10^{-13}/\text{year}. \] (51)

The optical clock transition energy \(E(Hg) \ (\lambda=282 \text{ nm})\) in the Hg\(^+\) ion can be presented in the following form:
\[ E(Hg) = \text{const} \times \frac{m_e e^4}{\hbar^2} F_{rel}(Z \alpha). \] (52)

Numerical calculation of the relative variation of \(E(Hg)\) has given [17]:
\[ \frac{\delta E(Hg)}{E(Hg)} = -3.2 \frac{\delta \alpha}{\alpha}. \] (53)
This corresponds to $V(HgOpt) = \alpha^{-3.2}$. Variation of the ratio of the Cs hyperfine splitting $A(Cs)$ to this optical transition energy is described by $X(Opt) = V(Cs)/V(HgOpt)$:

$$X(Opt) = \alpha^6 \left( \frac{m_q}{\Lambda_{QCD}} \right)^{0.073} \left( \frac{m_s}{\Lambda_{QCD}} \right)^{0.066} \left( \frac{m_e}{\Lambda_{QCD}} \right)^{0.066} .$$

Here we used Eq. (30) for $V(Cs)$. The work of Ref. 20 gives the limit on variation of this parameter:

$$\frac{1}{X(Opt)} \frac{dX(Opt)}{dt} < 7 \times 10^{-15}/\text{year} .$$

Molecular vibrational transitions frequencies are proportional to $(m_e/m_p)^{1/2}$. Based on Eq. (31) we may describe the relative variation of vibrational frequencies by the parameter

$$V(vib) = \left( \frac{m_q}{\Lambda_{QCD}} \right)^{-0.018} \left( \frac{m_s}{\Lambda_{QCD}} \right)^{-0.005} \left( \frac{m_e}{\Lambda_{QCD}} \right)^{0.5} .$$

Comparison of the Cs hyperfine standard with $SF_6$ molecular vibrational frequencies was discussed in Ref. 31. In this case $X(Cs/Vibrations) = \alpha^{2.8}[m_e/\Lambda_{QCD}]^{0.5}[m_q/\Lambda_{QCD}]^{0.093}[m_s/\Lambda_{QCD}]^{0.011}$.

The measurements of hyperfine constant ratios in different isotopes of the same atom depends on the ratio of magnetic moments and is therefore sensitive to $m_q/\Lambda_{QCD}$. For example, it would be interesting to measure the rate of change for hydrogen/deuterium ratio where $X(H/D) = [m_q/\Lambda_{QCD}]^{-0.068}[m_s/\Lambda_{QCD}]^{0.032}$.

Walsworth has suggested that one might measure the ratio of the Zeeman transition frequencies in noble gases in order to explore the time dependence of the ratio of nuclear magnetic moments. Consider, for example $^{129}$Xe/$^3$He. For $^3$He the magnetic moment is very close to that of neutron. For other noble gases the nuclear magnetic moment is also given by the valence neutron, however, there are significant many-body corrections. For $^{129}$Xe the valence neutron is in an $s_1/2$ state, which corresponds to the single-particle value of the nuclear magnetic moment, $\mu = \mu_n = -1.913$. The measured value is $\mu = -0.778$. The magnetic moment of the nucleus changes most efficiently through the spin-spin interaction, because the valence neutron transfers a part of its spin, $<s_z>$, to the core protons and the proton magnetic moment is large and has the opposite sign. In this approximation $\mu = (1 - b)\mu_n + b\mu_p$. This gives $b=0.24$ and the ratio of magnetic moments $Y = \mu(^{129}$Xe)/$\mu(^3$He) $\approx 0.76 + 0.24 g_p/g_n$. Using Eqs. (23, 24, 25, 26) we obtain an estimate for the relative variation of $\mu(^{129}$Xe)/$\mu(^3$He), which can be presented as variation of $X = [m_q/\Lambda_{QCD}]^{-0.027}[m_s/\Lambda_{QCD}]^{0.012}$. Here again $\delta Y/Y = 6X/X$.

Note that the accuracy of the results presented in this paper depends strongly on the fundamental constant under study. The accuracy for the dependence on $\alpha$ is a few percent. The accuracy for $m_q/\Lambda_{QCD}$ is about 30% – being limited mainly by the accuracy of the single-particle approximation for nuclear magnetic moments. (For comparison, the estimated systematic error associated with the calculation of the effect of the quark mass variation is less than 10%) Finally, we stress that the relation between the variation of $\alpha$ and $m/\Lambda_{QCD}$ has been used solely for purposes of illustration.

**Acknowledgments**

V.F. is grateful to C. Chardonnet, S. Karshenboim and R. Walsworth for valuable discussions and to the Institute for Advanced Study and the Monell foundation for hospitality and support. This work is supported by the Australian Research Council.

**APPENDIX A: MAGNETIC MOMENTS**

As explained in the text, we explicitly include the processes shown in Fig. 1 which give rise to the leading and next-to-leading nonanalytic behaviour as a function of quark mass.

We describe the quark mass dependence of the magnetic moments as:

$$\mu = \frac{\alpha_0}{1 + \alpha_2 m_q^2} + M^L ,$$

where $M^L$ denotes the chiral loop corrections given by

$$M^L = \chi_{\mu(a)} I_{\mu}(m_\pi, 0, \Lambda) + \chi_{\mu(b)} I_{\mu}(m_\pi, \Delta_{N\Delta}, \Lambda) + \chi_{\mu(c)} I_{\mu}(m_K, \Delta_{N\Lambda}, \Lambda) + \chi_{\mu(d)} I_{\mu}(m_K, \Delta_{N\Sigma}, \Lambda).$$

The chiral coefficients of the loop integrals, $\chi_{\mu(\alpha)}$, are given by

$$\chi_{\mu(\alpha)} = \beta_{\mu(\alpha)} \frac{M_N}{8 \pi f^2} ,$$

and are summarised in Table I. Note that the required analytic terms in the chiral expansion to this order have been placed in a Padé approximant designed to reproduce the Dirac moment behaviour of the nucleon at moderate quark mass.

**TABLE I:** Chiral coefficients for various diagrams contributing to proton and neutron magnetic moments. We use $SU(6)$ symmetry to relate the meson couplings to the $\pi N\Delta$ vertex, $C = -2D$.

| Diagram | $\beta_{\mu(\alpha)}$ | $\beta_{\mu(\alpha)}$ |
|---------|-----------------------|-----------------------|
| (a)     | $-(F + D)^2$          | $F + D)^2$            |
| (b)     | $-\frac{\alpha_2}{2\mu} \frac{m_q}{\Lambda_{QCD}}$ | $\frac{\alpha_2}{2\mu} \frac{m_q}{\Lambda_{QCD}}$ |
| (c)     | $-\frac{\alpha_2}{2\mu} (D + 3F)^2$ | 0 |
| (d)     | $-\frac{\alpha_2}{2\mu} (D - F)^2$ | $-(D - F)^2$ |
The corresponding loop integral is given by

\[ I_\mu(m, \Delta, A) = \frac{4}{3\pi} \int_0^\infty dk \frac{(\Delta + 2\omega_k)k^2u^2(k, A)}{2\omega_k^3(\Delta + \omega_k)^2} \]  

(A4)

where the various terms have been defined in Sect. II. We note that in the limit where the mass-splitting vanishes this integral is normalised such that the leading nonanalytic contribution is \( m \).

With the coefficients of the loop integrals defined, we only require determination of the parameters \( \alpha_0 \) and \( \alpha_2 \) in Eq. (A1) to constrain the variation with quark mass. We note also that this form assumes no analytic dependence on the strange quark mass, beyond what is implicitly included in the loop diagrams \((c,d)\). We determine \( \alpha_{0,2} \) for both the proton and neutron by fitting the physical magnetic moment as well as the lattice QCD data. We fit only to the two heaviest simulation results of the CSSM Lattice Collaboration [34], \( m_s^2 \sim 0.6–0.7 \text{ GeV}^2 \). These simulations were performed with the FLIC fermion action [87], on a \( 20^3 \times 40 \) lattice at \( a = 0.128 \text{ fm} \). We select the heaviest two data points, where the effects of quenching are anticipated to be small [43, 44].

The best fits to the physical values and the lattice data give

\[ \alpha_0^p = 2.17 \mu_N \quad \alpha_0^n = 0.817 \text{ GeV}^{-2} \]  

(A5)

\[ \alpha_2^p = -1.33 \mu_N \quad \alpha_2^n = 0.758 \text{ GeV}^{-2} \]  

(A6)

Upon renormalisation of the loop diagrams, the resultant magnetic moments in the SU(2) chiral limit are given by

\[ \mu_0^p = 3.48 \mu_N \quad \text{and} \quad \mu_0^n = -2.58 \mu_N . \]  

(A7)

We now take derivatives of Eq. (A1) at the physical pion mass to determine the variation with quark mass. In particular, we have

\[ \frac{\delta \mu}{\mu} = \left( \frac{m_s^2}{\mu} \frac{d \mu}{d m_s^2} \right) \frac{\delta m_s}{m_s} \]  

(A8)

\[ \frac{\delta \mu}{\mu} = \left( \frac{m_K^2}{\mu} \frac{d \mu}{d m_K^2} \right) \frac{\delta m_K}{m_K} . \]  

(A9)

This yields the results shown in the text.
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