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## On the existence of a quadrupole moment in muonium

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It is shown that even in its ground-state, muonium (hydrogen, mu-nucleonic atom) has a quadrupole moment which is comparable in magnitude to nuclear quadrupole moments. This leads to a pronounced effect of inhomogeneous intracrystalline fields on the spin precession and relaxation of  $\mu^+$  mesons and may serve as a basis for carrying out experiments with muonium (mu-nucleonic atoms) which are analogous to quadrupole resonance and relaxation.

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It is well known that the principal quantum characteristic of muonium is its magnetic moment. It is specifically the existence of this magnetic moment that gave rise to the meson method for studying properties of matter, a distinctive analogue of NMR and EPR, and to its intensive development in recent times (cf., the review article<sup>[1]</sup>). It will be shown below that muonium in its ground state has yet another quantum characteristic-a quadrupole moment. The existence of a quadrupole moment in muonium opens up new possibilities in the meson method associated with the investigation of quadrupole splitting of muonium levels and of the mechanism of its quadrupole relaxation in matter. In this sense the meson method becomes similar to nuclear quadrupole resonance and can be utilized to study not only magnetic but also inhomogeneous intracrystalline electric fields.

At first sight the assertion made above contradicts the well-known circumstance that as a result of spherical symmetry of the Coulomb interaction the ground 1S-state of muonium (of a hydrogen atom, and of other hydrogen-like systems) is described by a spherically symmetric wave function. As a consequence of this the aforementioned systems should not have any electric multipole moments. However, it is necessary to note that a violation of central symmetry, although insignificant at first sight, arises as a result of the hyperfine interaction between the spins of the electron and of the  $\mu^*$  meson in

muonium ( $\mu^-$  meson and the nucleus in a mesic atom, etc.). As a result of this the ground state does not have spherical symmetry and the appearance becomes possible of a quadrupole moment<sup>[2]</sup> comparable, as it turns out, in order of magnitude with nuclear quadrupole moments. Indeed, the energy of the hyperfine interaction, for example, in the system  $\mu^+e^-$  can be written in the form<sup>[3]</sup>

$$(\mathbf{r}, \mathbf{\bar{s}}, \mathbf{\bar{i}}) = V_{\mathbf{i}}(\mathbf{r}) \sigma_{\mathbf{i}} \sigma_{\mathbf{2}} + V_{\mathbf{2}}(\mathbf{r}) \{3\sigma_{\mathbf{i}}\mathbf{n}\} (\sigma_{\mathbf{2}}\mathbf{n}) - \sigma_{\mathbf{i}} \sigma_{\mathbf{2}}\}', \qquad (1)$$

where

V

$$V_1(\mathbf{r}) = -\frac{8}{3}\pi\mu_1\mu_2\delta(\mathbf{r}), \quad V_2(\mathbf{r}) = -\frac{\mu_1\mu_2}{r^3}, \quad \mathbf{n} = \mathbf{r}/r,$$

 $\sigma$  are the Pauli matrices,  $\mathbf{s} = \frac{1}{2}\sigma_1$ ,  $\mathbf{i} = \frac{1}{2}\sigma_2$ ,  $\mu_1$  is the magnetic moment of  $e^-$ ,  $\mu_2$  is the magnetic moment of  $\mu^+$ .

In the usual analysis of the hyperfine splitting of levels of atoms which are in the ground 1S state the second term in (1) is not taken into account since on averaging over a spherically symmetric state it is equal to zero.<sup>[31]</sup> However, the interaction proportional to  $V_2$  can admix to the triplet state of muonium, for example, the  ${}^{3}D_{1}$  state. As a result of this, without practically altering the energy of the triplet level the small admixture to it of the *D* state will lead to a qualitatively new result: to the appearance in the system of a quadrupole moment  $Q^{[2]}$ (cf., with the analogous mechanism for the appearance of a quadrupole moment in a deuteron<sup>[41</sup>; we also note that the spin structure of the second term in (1) is analogous to the spin structure of the tensor part of the nuclear potential responsible for the appearance of Q in the case of a deuteron).

In the absence of the hyperfine interaction the wave functions  $|nlsjiFM\rangle$  and the energy spectrum of a hydrogenlike atom are characterized by the quantum numbers nlsijFM (n is the principal quantum number, 1 is the orbital angular momentum, j=1+s is the angular momentum of the atomic shell, i is the angular momentum of  $\mu^{*}$ (proton, atomic nucleus),  $\mathbf{F}=\mathbf{j}+\mathbf{i}$  is the total angular momentum of the atom, M is the magnetic quantum number corresponding to the angular momentum F).<sup>[5]</sup>

The hyperfine interaction V admixes to the ground triplet state of muonium (hydrogen) characterized by the quantum numbers n=1, l=0,  $s=\frac{1}{2}$ ,  $j=i=\frac{1}{2}$ , F=1, M, excited states  $|nlsjiFM\rangle$  allowed by the law of conservation of F and M and having the same parity as the ground state. As a result of this in first order in V the wave function of the ground triplet state can be written in the form

$$\psi_{15}^{\alpha} = |10^{1}/_{2}^{1}/_{2}^{1}/_{2}^{1}/_{2}^{1}M\rangle + \sum_{n=2}^{\infty} \beta_{n}^{(D)} |n2^{1}/_{2}^{1}/_{2}^{1}/_{2}^{1}M\rangle, \qquad (2)$$

where  $\beta_n^{(S)}$  and  $\beta_n^{(D)}$  are the mixing coefficients of the *nS* and *nD* states:

$$\beta_{n}^{(s)} = \frac{\langle n0^{1}/_{2}^{1}/_{2}^{1}/_{2} \frac{1}{1} \frac{1}{M} | V | 10^{1}/_{2}^{1}/_{2} \frac{1}{M} \rangle}{E_{1s} - E_{ns}} = -\frac{2}{3} \mu_{1} \mu_{2} \frac{R_{n0}(0)R_{10}(0)}{E_{1s} - E_{ns}},$$

$$\beta_{n}^{(D)} = \frac{\langle n2^{1}/_{2}^{3}/_{2} \frac{1}{2} \frac{1}{M} | V | 10^{1}/_{2}^{1}/_{2} \frac{1}{M} \rangle}{E_{1s} - E_{nD}} = -\frac{\sqrt{8}}{8} \mu_{1} \mu_{2} \frac{\langle n2 | r^{-1} | 10 \rangle}{E_{1s} - E_{nD}}.$$
(4)

Here the radial matrix element is taken between the ground, n=1, l=0, and the excited, n, l=2, D states (cf., below for its explicit form)  $E_{1S}$  and  $E_{nD}$  are the fine structure energies corresponding to them,  $R_{n0}(0)$  and  $R_{10}(0)$  are the radial Coulomb functions for r=0 (cf., Refs. 3 and 5 for their form).

The quadrupole moment of the system in the ground (triplet) state of a hydrogenlike atom of interest to us is determined by the expression

$$Q = \langle \psi_{1s}^{M} | Q_{1s} | \psi_{1s}^{M} \rangle_{M-1}, \qquad (5)$$

where  $Q_{zz}$  is the operator for the zz component of the quadrupole moment. In the center of inertia system for the atom we have

$$Q_{zz} = \frac{Zm_1^2 - m_2^2}{(m_1 + m_2)^2} (3z^2 - r^2), \qquad (6)$$

where Z and  $m_2$  are the charge and the mass of the atomic nucleus (in muonium  $\mu^*$  plays the role of the nucleus),  $m_1$  is the mass of the electron ( $\mu^-$  meson, if we are dealing with a mesic atom,  $\mu^-$ +nucleus). From (6) it can be seen that positronium (Z = 1,  $m_1 = m_2$ ) does not have a quadrupole moment.

Substituting (2-4) into (5) one can obtain for the quadrupole moment of the system in the ground triplet state in first order with respect to mixing the following expression:

$$Q = -\frac{8}{5} \frac{Zm_1^2 - m_3^2}{(m_1 + m_2)^2} \mu_1 \mu_2 \sum_{n=3}^{\infty} \frac{\langle n2|r^{-3}|10\rangle \langle n2|r^2|10\rangle}{E_{18} - E_{nD}},$$
(7)

where the radial matrix elements are taken, just as in (4), between the ground and the excited D states of the atom and are equal to

$$\langle n2|r^{-3}|10\rangle = \frac{2}{15} \frac{1}{n^2 (n+1)^2} \left(\frac{(n+2)!}{(n-3)!}\right)^{\frac{1}{2}} F\left(-(n-3), 2, 6, \frac{2}{n+1}\right) a^{-3},$$
  
$$\langle n2|r^2|10\rangle = 96 \frac{n^3}{(n+1)^7} \left(\frac{(n+2)!}{(n-3)!}\right)^{\frac{1}{2}} F\left(-(n-3), 7, 6, \frac{2}{n+1}\right) a^2,$$
 (8)

F are hypergeometric functions,  $a = (\hbar^2/Ze)(m_1 + m_2)/m_1m_2$  is the Bohr radius of the system under consideration.

The quadrupole moment of muonium (of a hydrogen atom) is obtained from (7), if we set Z=1, and is equal to  $Q_{Mu}=2.2\times10^{-25}$  cm<sup>2</sup> ( $Q_{H}=0.7\times10^{-25}$  cm<sup>2</sup>). We recall for comparison that the quadrupole moment of a deuteron is equal to  $Q_{d}=0.28\times10^{-26}$  cm<sup>2</sup>.

The existence of a nonvanishing quadrupole moment leads to the fact that even in the absence of a magnetic field the polarization vector of a  $\mu^*$  meson in muonium ( $\mu$ <sup>-</sup> in a mesic atom) whose surroundings do not have cubic symmetry, will oscillate at a frequency determined by the quadrupole splitting of the levels with M=0and M = +1. Since the magnitude of Q is of the order of nuclear quadrupole moments one should expect that also the magnitude of the quadrupole splitting will be of the order of splitting observed in NQR, i.e., the beat frequency for  $\mu^{\pm}$  will be of the order of  $10^6 - 10^7 \text{ sec}^{-1}$ , and this lies in the range of frequencies typical for experiments with muonium. If a magnetic field is turned on, then the levels with M = +1 will be split and the  $\mu^{\pm}$  meson will oscillate at two frequencies determined by the energy difference of levels with M = 0 and M = -1 (M = 0and M=1). Explicit expressions for the magnitude of the splitting of the levels of the triplet state of an atom (i.e., the splitting of levels of a particle with unit spin) in a magnetic and an inhomogeneous electric field of arbitrary orientation can be easily obtained if one utilizes the known solution of a similar problem in NQR (cf., for example, Ref. 6).

The hyperfine interaction leads to the appearance of Q also in excited triplet S states of atoms (this circumstance for  $n \ge 3$  was noted in Ref. 7).

In obtaining the quadrupole moment in an excited state one should take into account the fact that for large n the hyperfine interaction can be comparable with the spacing between the level nS and the level of the fine structure nD closest to it. For this reason the mixing of wave functions by the hyperfine interaction must be investigated with the aid of perturbation theory in the presence of closely spaced levels. Simple, but sufficiently awkward calculations yield the following expression for the quadrupole moment of levels which, as the hyperfine interaction tends to zero, go over into nS levels (the contribution of only the nD levels closest to the nS level under consideration is taken into account):

$$Q_{ns} = \frac{\sqrt{8}}{5} \frac{Zm_1^2 - m_2^2}{(m_1 + m_2)^2} \frac{H_{ns} - E_{ns}'}{V_{ns,nD}} \langle n2 | r^2 | n0 \rangle, \qquad (9)$$

$$\Pi_{ns} = \mathcal{L}_{ns} + \langle \mathcal{H} 0 \rangle_2 / 2 / 2 \Pi u | v | \mathcal{H} 0 / 2 / 2 / 2 \Pi u \rangle = \mathcal{L}_{ns} - / 3 \mu_1 \mu_2 \mathcal{H}_{n0} (0),$$

 $E_{ns}' = \frac{1}{2} (H_{ns} + H_{nD}) + \frac{1}{2} [(H_{ns} - H_{nD})^{2} + 4 |V_{ns, nD}|^{2}]^{\frac{1}{2}},$   $H_{nD} = E_{nD} + \frac{1}{2} \frac{1}{2} \frac{3}{4} \frac{1}{4} \frac{1}{2} \frac{1}{4} \frac{1}{2} \frac{1}{2} \frac{1}{2} \frac{3}{4} \frac{1}{4} \frac{1}{2} \frac{1}{2} \frac{1}{4} \frac{$ 

We note that the hyperfine interaction does not mix the 3S and 3D levels, and also 4S and 4D levels (for n=3 this fact was noted by Berestetskii<sup>[8]</sup>). For this reason the quadrupole moment in these states is possible only as a consequence of taking into account the admixture to them of states with different n.

We add, that in the case of  $\mu^-$  +a nucleus there is yet another important mechanism for the formation of a quadrupole moment in a mesic atom produced by the violation of the spherical symmetry of the Coulomb field of nuclei the spin of which is  $J \ge 1$ . In order to estimate the magnitude of Q in this case we consider the ground state of a  $\mu^-$  mesic atom, for example, with a nucleus characterized by J=3/2. For the nuclear model we choose the rigid deformed rotator, symmetric with respect to some axis. The nuclear state in this model can be specified by the quantum numbers  $J, M_J, K$  (the total angular momentum of the nucleus, its component along the z axis and its component along the symmetry axis). We assume that the deviation from spherical symmetry of the Coulomb field due to the presence of an internal quadrupole moment of the nucleus  $Q_0$  is small, i.e., the quadrupole interaction  $V_Q$  of the  $\mu^-$  meson with the nucleus can be regarded as a perturbation of the fine structure of the mesic atom.<sup>[9]</sup> In the absence of the quadrupole interaction the wave functions  $|JK, nlj, FM\rangle$  and the energy spectrum of the atom are characterized by the quantum numbers JKnljFM (n is the principal quantum number, l is the orbital angular momentum, j=l+sis the angular momentum of the  $\mu^{-}$  meson,  $\mathbf{F} = \mathbf{j} + \mathbf{J}$  is the total angular momentum of the atom, M is its component along the z-axis). In first order with respect to  $V_{0}$ the wave function of the ground (K = J) triplet state of the atom can be written in the form

$$\psi_{10}{}^{M} = |{}^{3}/{}_{2}{}^{3}/{}_{2}, 10{}^{1}/{}_{2}, 1M\rangle + \sum_{n=3}^{\infty} \beta_{n}^{(D{}^{N}{}_{1})} |{}^{3}/{}_{2}{}^{3}/{}_{2}, n2{}^{3}/{}_{2}, 1M\rangle + \sum_{n=3} \beta_{n}^{(D{}^{3}/{}_{2})} |{}^{3}/{}_{2}{}^{3}/{}_{2}, n2{}^{3}/{}_{2}, 1M\rangle,$$
(11)

where  $\beta_n^{(D_j)}$  are the mixing coefficients of the *D* states with j = 3/2 and j = 5/2. In (11) we have omitted the excited *S* states which gave no contribution in first order in *Q*. In the model chosen by us<sup>[9]</sup> we have

$$V_{\mathbf{q}} = -\frac{e^2 Q_{\mathbf{q}}}{2} f(r) P_{\mathbf{1}}(\cos \theta), \qquad (12)$$

where  $P_2(\cos\theta)$  is a Legendre polynomial,  $\theta$  is the angle

between the position vector of the  $\mu^-$  meson and the nuclear symmetry axis, the function f(r) is defined in Ref. 9. Using the standard method for dealing with tensor operators,<sup>[5]</sup> and also the explicit form for the mixing coefficients  $\beta$  (cf., Ref. 9), we obtain for the quadrupole moment of the mesic atom in the ground triplet state

$$Q_{\mu^{-}\text{nuc}} = Q_{*p} + \frac{2}{125} e^{2} Q_{0} \sum_{n=3}^{\infty} \frac{\langle n2|r^{2}|10\rangle \langle n2|f(r)|10\rangle}{E_{15'h} - E_{nD'h}} + \frac{3}{125} e^{2} Q_{0} \sum_{n=3}^{\infty} \frac{\langle n2|r^{2}|10\rangle \langle n2|f(r)|10\rangle}{E_{15'h} - E_{nD'h}}$$
(13)

where  $Q_{sp}$  is the observed so-called spectroscopic quadrupole moment of the nucleus, the radial matrix elements are taken between the ground and the excited Dstates of the  $\mu^-$  atom ( $Q_{sp}$  takes into account the contribution of the  $\mu^-$ ).

In the model based on a surface quadrupole charge distribution<sup>(9)</sup> we obtain for the quadrupole moment due to the mechanism under consideration the estimate  $Q' \sim Q_{sp} \sim 10^{-25} - 10^{-24}$  cm<sup>2</sup>. It is of interest that if the so-called dynamic E2 effects<sup>(9)</sup> are taken into account this leads to the appearance of a quadrupole moment in the ground states of mesic atoms whose nuclei also have a spin lower than unity.

Going over to the discussion of the possibility of an experimental observation of the quadrupole moment of mesic atoms we first of all turn our attention to  $\mu^-$  atoms, i.e., to the system  $\mu$ <sup>-</sup> + a nucleus. The dimensions of such mesic atoms are significantly smaller than the dimensions of ordinary atoms and interatomic distances. For this reason the expressions given above, in which the distortions of the excited states by the medium are not taken into account, are applicable to them. If a mesic atom has a unit spin and is situated in an inhomogeneous intracrystalline field characterized by axial symmetry, then, as has already been pointed out, the levels with M = 0 and M = +1 undergo a splitting and the polarization vector for the  $\mu^-$  mesons will in the absence of a magnetic field exhibit beats at the splitting frequency. But if the field does not have axial symmetry then the levels with M = +1 and M = -1 will also be split. As a result of this even in the absence of a magnetic field in consequence of the quadrupole splitting of the levels it turns out to be possible to observe two-frequency beats of the polarization vector of the  $\mu^-$  mesons over a wide range of frequencies of the order of  $10^6 - 10^9 \text{ sec}^{-1}$  (cf., above). It is important to note that such experiments in the absence of a magnetic field can be carried out using polycrystalline samples. But if a magnetic field is applied to the target then as a result of the dependence of the magnitude of the level splitting on the orientation of the crystal with respect to the magnetic field it is necessary for the observation of the effect of beats due to quadrupole splitting to use single crystals as targets. The situation here is completely analogous to quadrupole resonance.<sup>[6]</sup>

One can also observe quadrupole splitting of lines of hydrogenlike atoms in vacuum in an inhomogeneous electric field produced, for example, by a standing laser wave.<sup>[2]</sup> The magnitude of the splitting of excited states in the field of a wave of  $\sim 10^4$  V/cm turns out to be of the order of  $10^7-10^8$  sec<sup>-1</sup>, and this can be recorded by modern methods of laser spectroscopy (cf., for example, Ref. 10).

Thus, the discussion given above indicates completely realistic (particularly in the case of  $\mu$ -mesic atoms) experimental possibilities of observing the rotation and relaxation of spin of  $\mu^*$  mesons due to a quadrupole moment under very varied conditions, and this can be utilized for a further extension of the possibilities of the  $\mu$ -meson method for investigation of matter. The mechanism for the formation of a quadrupole moment noted above should also be taken into account in investigating the quadrupole and nuclear  $\gamma$  resonances in the case of ions which have common electron-nucleus levels.

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## ful remarks.

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## Study of the temperature dependence of the probabilities of emission and absorption of resonance $\gamma$ rays by <sup>182</sup>W without loss of energy to recoil

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The current method of  $\gamma$ -ray detection has been used to measure the Mössbauer effect in the 100.1-keV level of <sup>182</sup>W in the absorber temperature range 109–255 K at a constant source temperature of 89 K and in the range of source temperatures 93.5–160 K at a constant absorber temperature of 101 K. We have determined the temperature dependence of the probability of absorption and emission of resonance  $\gamma$  rays by <sup>182</sup>W without loss of energy to recoil, and the Debye temperatures of metallic tungsten and tantalum.

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The further extension of the region of nuclei in which it is possible to observe and study the Mössbauer effect depends, on the one hand, on the possibility of selecting sources and absorbers with a large ratio  $\Theta/T$ , where  $\Theta$ is the Debye temperature and T is the absolute temperature, and on the other hand—on an increase in the accuracy of the measurements, which is achieved by detection of a very large number of events in relatively short time interval which depends on the stability of the apparatus. However, in the latter case experimenters encounter the problem of dead time in the measuring system. not the pulse method of  $\gamma$ -ray detection, but the current method.<sup>[1-3]</sup> This method provides the possibility of investigating nuclei with small expected Mössbauer effect, whose study by the usual pulse method of  $\gamma$ -ray spectrometry is extremely difficult. In order to use the current method for Mössbauer spectroscopy, we have developed an apparatus based on an LP-4050 512-channel analyzer and a scintillation detector.

The temperature dependence of the Mössbauer effect in the 100.1-keV level of <sup>182</sup>W in the temperature range roughly from 20 to 160 K has been studied only once,<sup>[4]</sup> by detection of conversion electrons by a  $\beta$  spectrometer of the Kofoed-Hansen type. However, in that work as a

As a result there has recently been a tendency to use

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