Experimental Determination of the ^{24}Mg I $(3s3p)^3P_2$ Lifetime

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We present the first experimental determination of the electric-dipole forbidden $(3s3p)^3P_2 \rightarrow (3s^2)^1S_0$ ($M2$) transition rate in ^{24}Mg and compare to state-of-the-art theoretical predictions. Our measurement exploits a magnetic trap isolating the sample from perturbations and a magneto-optical trap as an amplifier converting each $^3P_2 \rightarrow {}^1S_0$ decay event into millions of photons readily detected. The transition rate is determined to be $(4.87 \pm 0.3) \times 10^{-4}$ s^{-1} corresponding to a $^3P_2$ lifetime of 2050^{+140}_{-110} sec. This value is in agreement with recent theoretical predictions, and to our knowledge the longest lifetime ever determined in a laboratory environment.

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Alkaline earth elements have attracted considerable attention, theoretical as a well as experimental, over the past years. One of the main motivations for studying these atoms is their use for optical atomic clocks and high precision measurements [1–4]. Especially the ^{87}Sr atom has enjoyed success and now forms a basis for neutral atoms clocks [4,5] that surpass the best ^{133}Cs primary standards [6–8]. The high precision achieved relies on long-lived metastable states where a doubly forbidden intercombination transition $^1S_0 \rightarrow {}^3P_0$ provides a narrow resonance with a mHz natural linewidth.

Effective isolation of a quantum system dramatically increases the ability to make accurate measurements. Magnetic, optical, and magneto-optical trapping of neutral atoms in ultrahigh vacuum environments form an important basis for many of today's precision measurements. Recently, several groups have even succeeded in creating a Bose-Einstein condensate (BEC) with alkaline earth elements, such as ^{40}Ca [9] and ^{84}Sr [10,11], by trapping the atoms in crossed dipole traps. Unlike these two elements, magnesium has the attractive property that evaporative cooling seems more likely to be feasible for atoms trapped magnetically in the long-lived $^3P_2$ state [12], even for the most abundant isotope ^{24}Mg, thereby opening up the possibility of creating a BEC this way with an alkaline earth element. The long lifetime and possibility to evaporatively cool ^{24}Mg magnetically trapped in the $^3P_2$ state also offer interesting perspectives for quantum logic operations and quantum computing [13].

Despite considerable efforts, the lifetimes of forbidden transitions of alkaline earth atoms remain relatively unknown, in particular, for magnesium [14]. Unlike the energy separation between atomic levels, weak transition rates offer a more challenging test of modern quantum theory, where an accurate determination of matrix elements can serve as a benchmark test of atomic structure models. Today's atomic structure calculations based on modern *ab initio* methods aim at predicting basic properties of low-lying states with an accuracy better than 1% [15]. In this connection, reliable experimental values for allowed—and, in particular, weak—transition rates play a key role for the further advancement of modern quantum theory. The measurement presented in this work provides a stringent test of state-of-the-art theoretical predictions.

The very long lifetime of the $^3P_2$ state (see Fig. 1) is a result of quantum mechanical selection rules, which exclude direct single photon decay to first order by dipole transitions ($E1$) and to second order by magnetic dipole transitions ($M1$) or electric quadrupole transitions ($E2$). By a third order process single photon decay to the ground state is allowed via a magnetic quadrupole ($M2$) transition. Decay probabilities for ($M1$, $E2$) transitions $^3P_2 \rightarrow {}^3P_1$, $^3P_0$ are lower by many orders of magnitude [14].

In a pioneering work, ^{88}Sr atoms in the $^3P_2$ state were magnetically trapped [16], and in a later work the lifetime was measured to be 520^{+310}_{-140} s [17], about a factor of 2 below theoretical predictions. Similar results for other alkaline earth elements are, to our knowledge, not reported in the literature. The measurements using ^{88}Sr atoms where complicated by quenching induced by room temperature blackbody radiation (BBR). Optical pumping at 2 μm connecting the $^3P_2$ state to the ^3D_J manifold resulted in an unwanted depletion of trapped atoms and a complex data analysis was required. This temperature dependence is absent in the magnesium system. Here, resonant transitions are considerably more energetic, rendering the system practically immune to blackbody effects at room temperature. This is also a key feature behind proposals of magnesium for a future atomic clock since it has one of the lowest BBR contributions among optical atomic clock candidates [18].

Similar to the setup for ^{88}Sr described in [17], our setup to measure the $^3P_2$ lifetime of ^{24}Mg features a magnetic trap for storing the $^3P_2$ atoms and a magneto-optical trap (MOT) collecting atoms that decay to the ground state thereby amplifying each decay event considerably. The magnetic trap is formed by the MOT quadrupole field

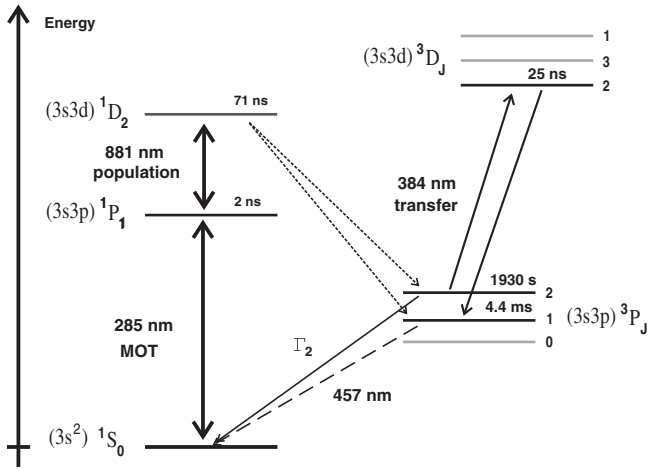

FIG. 1. Energy level diagram for ^{24}Mg. Atoms magnetically trapped in the $^3P_2$ state decay to the $^1S_0$ state by a magnetic quadrupole ($M2$) transition.

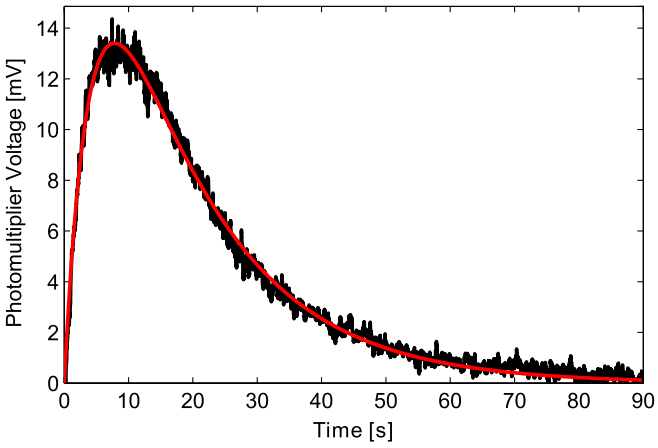

FIG. 2 (color online). Experimental data for $^3P_2$ decay back to the $^1S_0$ ground state. Here, atoms are recaptured in the MOT and a strong fluorescence signal emerges. The red (or gray) curve is a fit of Eq. (2) to the data consistent with no intratrap collisions.

and ensures optimal overlap. Each decay event $^3P_2 \rightarrow {}^1S_0$ is amplified by the MOT to millions of photons which we record. The principle of our measurement relies on counting the number of decayed $^1S_0$ atoms relative to the remaining $^3P_2$ atoms after a time interval t. This ratio is independent of the initial population in the magnetic trap and yields the transition rate directly. Depending on the observation time t, small corrections must be included. These corrections are discussed more in detail below.

A sample of $^3P_2$ atoms will be depleted as a result of two effects. First, atoms decay to the ground state $^1S_0$ with a rate Γ_2. Second, atoms will be ejected out of the magnetic trap as a result of collisions with the background gas with a rate α_2. By operating the traps at low enough densities we can exclude collisions between the trapped atoms. In this case we find the rate equations for the number N_2 of $^3P_2$ atoms (in the magnetic trap) and the number N_0 of $^1S_0$ atoms (in the MOT),

$$\frac{dN_2}{dt} = -(\alpha_2 + \Gamma_2)N_2, \qquad \frac{dN_0}{dt} = -\alpha_0 N_0 + \Gamma_2 N_2. \tag{1}$$

Note that the magnetic trap lifetime $1/\alpha_2$ is generally different from the MOT lifetime $1/\alpha_0$ since additional two photon ionization losses are taking place in the MOT [19]. The solution to Eq. (1), assuming initially no population in the MOT, is then given by

$$N_0(t) = \frac{N_2^0 \cdot \Gamma_2}{\alpha_2 + \Gamma_2 - \alpha_0}(e^{-\alpha_0 t} - e^{-(\alpha_2+\Gamma_2)t}), \tag{2}$$

where N_2^0 is the initial magnetic trap population.

We operate a MOT on the $^1S_0 \rightarrow {}^1P_1$ transition at 285 nm (see Fig. 1), which captures approximately 4×10^7 ^{24}Mg atoms at a temperatures of 5 mK from a hot magnesium beam. By optical pumping on the $(3s3p)^1P_1 \rightarrow (3s3d)^1D_2$ transition at 881 nm, atoms may decay to $(3s3p)^3P_2$ at a rate of about 50 s^{-1} [20]. We prepare a sample of 10^6 $^3P_2$ atoms magnetically trapped in the MOT quadrupole field. We use a B-field gradient of 150 G/cm (symmetry axis) corresponding to a magnetic trap size of 2 mm, significantly smaller than the 8 mm MOT beam diameter.

Figure 2 shows experimental data of atoms decaying from $^3P_2$ to $^1S_0$ recorded over a 90 s period. The measured signal is the MOT fluorescence, which is proportional to $N_0(t)$, of atoms initially prepared in $^3P_2$. The red or gray line in Fig. 2 is a fit to Eq. (2). The goodness of the fit (adjusted R-square $= 0.988$) indicates that the assumption of no intratrap collisions is justified. At short time scales (up to 2 s) the signal is dominated by the $^3P_2$ decay rate. For longer time scales the finite lifetime of the MOT and magnetic trap dominate. The fit gives the lifetimes $1/\alpha_0 = 3$ s and $1/\alpha_2 = 16$ s for the MOT and magnetic trap, respectively. Even though the measurement of decaying $^3P_2$ atoms gives a nonzero signal for times of up to more than 50 s, the decay rate Γ_2 cannot be accurately extracted from this data by fitting Eq. (2) since the value is expected to be dwarfed by the other dominating decay rates, $\Gamma_2 \ll \alpha_0$, α_2, and the signal-to-noise ratio is too low for an accurate determination of Γ_2 this way.

To extract the decay rate Γ_2 in a different way, we have adopted an experimental procedure consisting of a preparation stage where we populate the $^3P_2$ magnetic trap, and three MOT stages. First, a counting stage where each $^3P_2$ decay is recorded as fluorescence using the MOT as an amplifier (A), then a transfer stage where the remaining $^3P_2$ atoms are transferred to the ground state and counted (B), and finally, a background stage where atoms loaded from the background vapor are counted (C).

The experimental cycle begins by loading the magnetic trap for 5 s, after which the Mg atomic beam is blocked by

a mechanical shutter. When the trap has been loaded we switch off the MOT light for 200 ms, which is sufficient to remove all atoms in $^1S_0$, and then turn it back on again. During the next 1000 ms (stage A) we monitor the decay of the $^3P_2$ atoms to the $^1S_0$ state through the MOT fluorescence. In this period, about 500 atoms are reloaded into the MOT. Then, using optical pumping via the $^3D_2$ state, we transfer all the remaining $^3P_2$ atoms to the $^1S_0$ ground state while detecting the MOT fluorescence (stage B). Resonant 384 nm light couples the $^3P_2$ state with the $^3D_2$ state. Since the involved branching ratio is 3:1, it takes only two 384 nm photons on average to transfer an atom back to the $^3P_1$ state where it decays back to the $^1S_0$ state in about 4 ms [21]. Finally, we measure the load from background gas during 1 s (stage C) starting with an empty magnetic trap and MOT. This allows us to correct for residual atoms recaptured during the 1 s decay recording initiating our measurement sequence. We have carried out more than 90 of these measurement cycles, which were distributed over several days of data acquisition.

Figure 3 shows a typical data recording cycle of the MOT fluorescence induced by spontaneous decay to $^1S_0$ state (A), after optical pumping back to $^1S_0$ (B), and finally a background load measurement (C).

The MOT fluorescence signal at time t_{ref} in stage (A), $t_{\mathrm{ref}} \ll 1/\alpha_0$, is given by $S_A = \eta\Gamma_2 N_2^0 t_{\mathrm{ref}}$, while the remaining atoms transferred to the MOT in (B) give the signal $S_B = \eta(1 - \alpha_2 t_{\mathrm{ref}})N_2^0 \cong \eta N_2^0$. Here, η represents the fluorescence detector efficiency and N_2^0 the initial $^3P_2$ sample size. Consequently, the ratio of the signals S_A and S_B should yield the decay rate Γ_2 independently of N_2^0.

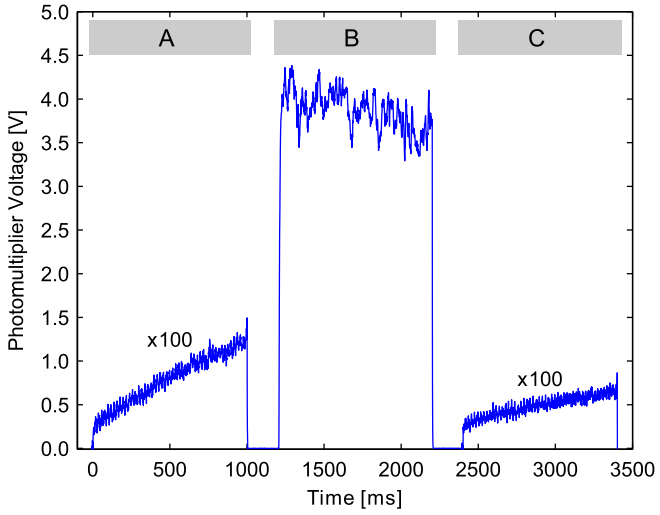

FIG. 3 (color online). Raw fluorescence signal from MOT atoms after $^3P_2$ decay to $^1S_0$ (A), optical transfer (B) to the $^1S_0$ ground state, and background load of the MOT (C). The decay (A) and background signal (C) are multiplied with a factor of 100 to fit all three curves on the same plot. From the measured signal we extrapolate the signals to a time $t_{\mathrm{ref}} = 500$ ms for a more accurate ratio.

This ratio can be measured with high precision but small corrections must be applied to account for the decay of the MOT and the magnetic trap sample during measurements and also from possible background load of the MOT. We have measured these corrections to be typically on the order of 3%.

In principle the lifetime is then given by the ratio $\Gamma_2 = \frac{\tilde{S}_A - \tilde{S}_C}{\tilde{S}_B t_{\mathrm{ref}}}$ of the background corrected load signal to the signal of back transferred population, where the tilde denotes a correction for the decay of the MOT and the magnetic trap during the measurement. This technique has been used in other experiments to determine lifetimes [17].

However, given the complicated dynamics and the different behavior of the various magnetic substates of $^3P_1$ ($m_F = 0, \pm 1$), it is necessary to measure directly the recapture fraction for the transition $^3P_1 \rightarrow {}^1S_0$. Atoms decaying from the $^3P_2$ manifold to $^1S_0$ state are assumed captured with unit efficiency as they are tightly confined within the MOT range. The recapture coefficient is deduced from three steady-state operations of the MOT. At the first stage, the MOT fluorescence is detected with signal M_1. For the second stage, light at 881 nm (optical pumping to $^3P_{1,2}$, see Fig. 1) is superimposed on the atoms giving a MOT fluorescence signal M_2. Finally, a third source of light at 384 nm (repumping to the ground state) is added giving a signal M_3. By writing down the steady-state equations in each case we find a recapture fraction β based on these measurements as

$$\beta = \frac{\Phi - \Theta}{\Phi - \Theta/(1 + R)}, \tag{3}$$

with $\Phi = \frac{M_1}{M_2} - 1$ and $\Theta = \frac{M_1}{M_3} - 1$. $R = 2.4^{-1}$ is the branching ratio of decay from $^1D_2 \rightarrow {}^3P_2$ and $^1D_2 \rightarrow {}^3P_1$ (with the latter dominating) determined experimentally in [20]. Uncertainties in the values of $M_{1,2,3}$ produce an error of 3.5% in β. From a series of 11 measurements of $M_{1,2,3}$ we find a recapture fraction of $\beta = 0.60 \pm 0.03$, including the 10% uncertainty in R from [20]. This value of β is consistent with 50% of the nontrapped $^3P_1$ atoms ($m_F = 0, -1$) escaping the MOT region before decaying due to their mean velocity of 1.44 m/s at 3 mK.

Prior to each experimental cycle, a magnesium beam is present in the main chamber for 5 s. This prompts us to characterize the vacuum pump speed as a function of time and we find it stable below 1%. This leads to corrections in Γ_2 on a 2% level. The trap lifetimes may change during experiments as the background pressure varies. We find that this pressure increase may change lifetimes by 0.3 s corresponding to a 0.5% correction error. The photomultiplier linearity was checked against a photodiode and another photomultiplier. We find it to be 99.5% or better in our operating regime. The complete error budget is presented in Table I.

TABLE I. Relative uncertainty budget for the $^3P_2$ lifetime measurement.

Source	Uncertainty [%]
Recapture fraction	4.7
Background correction	2
Photomultiplier linearity	0.5
Trap lifetimes	0.5
Statistical error	1.8
Total	5.5

A fit of Eq. (2) to the data in the (A) and (B) stages allows for an extrapolation of the signals to a common time reference $t_{\mathrm{ref}} = 500$ ms where the signal ratio is performed. Before the ratio is performed, the signal in (A) is corrected for the background load of the MOT with the signal in (C). However, the background load recorded in (C) does not have the exact same value it has at time t_{ref} since pumping during (A) and (B) has lowered the magnesium background pressure slightly. We correct for this by mapping out the characteristic pump speed function and extrapolate the value measured in (C) to time t_{ref}. Finally, the ratio is corrected with the fraction β and the lifetime is obtained.

In Fig. 4 we have compiled our measurements corrected with the recapture factor.

We obtain a final value of $1/\Gamma_2 = 2050^{+40}_{-30}$ s for the $^3P_2$ lifetime, the error quoted being the statistical uncertainty, corresponding to 1.8%.

The $^3P_2$ lifetime has been predicted theoretically by several groups. In Table II we compare to various calculations from models based on relativistic *ab initio* approaches [14,15,22]. The experimental value we have obtained differs by 2 standard deviations from the most recent theoretical value in [22], but given the complexity of the calculation the agreement between theory and experiment can still be considered as good. We also note that the experimental value is below all the theoretical estimates, which was also the case for the ^{88}Sr measurement reported in [17]. One reason for this could be magnetic quenching of the $^3P_2$ level. However, we have varied the magnetic field strength for the trap from 130 to 216 G/cm without observing any significant change in the value of the lifetime, so quenching effects do not seem to be dominating the lifetime. Another possibility could be a biasing due to ineffectiveness of the repumping to the ground state (via the $^3P_2 \rightarrow {}^3D_2$ transition at 384 nm) in our measurements. To rule this out we varied the frequency of the repump laser by one natural linewidth on either side of the resonance without seeing any effect on the lifetime. A temperature dependence was also tested by cooling the whole vacuum system to well below 0 °C, again with no observed effect on the lifetime. This contrasts the results obtained in [17] for ^{88}Sr, confirming that the BBR has a smaller impact on the Mg system.

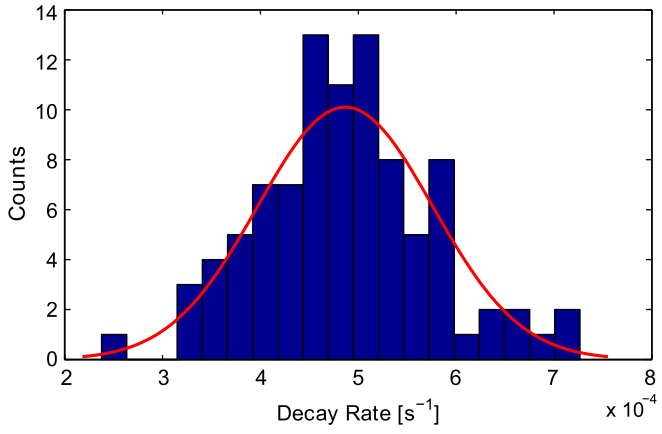

FIG. 4 (color online). A compilation of 93 measurements of the $^3P_2$ lifetime. The red (or gray) curve is a Gaussian fit to the data. We obtain $\Gamma_2 = (4.87 \pm 0.09) \times 10^{-4}$ s^{-1}.

Using the present setup we were also able to place an upper bound on the $^1P_1$ to $^3P_2$ ($M1, E2$) decay rate for the first time. Repeating the above measurements with no $^3P_2$ population stage we found a rate of $\Gamma \leq 4 \times 10^{-4}$ s^{-1}. Currently there are no published theoretical values on this transition, but estimates point towards $\Gamma \leq 1.4 \times 10^{-4}$ s^{-1} [25].

In conclusion, we have presented the first ever measurement of the ^{24}Mg I $(3s3p)^3P_2$ lifetime, which we find to be 2050^{+140}_{-110} s. This is to our knowledge the longest lifetime ever determined in a laboratory environment. The small measurement error (6%) provides a stringent test of state-of-the-art *ab initio* calculations of a $^3P_2$ lifetime. We find that the measured value is in agreement with the calculations, which not only verifies the models used in the theoretical estimates of the lifetime of the $^3P_2$ state, but also—along with the possibility to trap a large number of ^{24}Mg magnetically—confirms the feasibility of using magnesium in future setups that take advantage of the long $^3P_2$ lifetime, e.g., in BEC and quantum computing experiments.

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TABLE II. Experimental value compared to state-of-the-art theoretical calculations. Uncertainty given when applicable.

Work	Year	Lifetime [s]
Experimental		
This work	2011	2050^{+140}_{-110}
Theoretical		
Santra [22]	2004	2381 ± 117
Derevianko [14]	2001	2268
Jönsson [23]	1997	2510
Garstang [24]	1967	6250

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[1] G. K. Campbell *et al.*, Metrologia **45**, 539 (2008).
[2] N. D. Lemke *et al.*, Phys. Rev. Lett. **103**, 063001 (2009).
[3] J. Lodewyck, P. G. Westergaard, and P. Lemonde , Phys. Rev. A **79**, 061401(R) (2009).
[4] A. D. Ludlow *et al.*, Science **319**, 1805 (2008).
[5] X. Baillard *et al.*, Eur. Phys. J. D **48**, 11 (2007).
[6] S. Bize *et al.*, J. Phys. B **38**, S449 (2005).
[7] T. P. Heavner *et al.*, Metrologia **42**, 411 (2005).
[8] S. Weyers *et al.*, Metrologia **38**, 343 (2001).
[9] S. Kraft *et al.*, Phys. Rev. Lett. **103**, 130401 (2009).
[10] Y. N. Martinez de Escobar *et al.*, Phys. Rev. Lett. **103**, 200402 (2009).
[11] S. Stellmer *et al.*, Phys. Rev. Lett. **103**, 200401 (2009).
[12] A. Derevianko *et al.*, Phys. Rev. Lett. **90**, 063002 (2003).
[13] A. J. Daley *et al.*, Phys. Rev. Lett. **101**, 170504 (2008).
[14] A. Derevianko, Phys. Rev. Lett. **87**, 023002 (2001).
[15] S. G. Porsev *et al.*, Phys. Rev. A **64**, 012508 (2001).
[16] X. Xu *et al.*, J. Opt. Soc. Am. B **20**, 968 (2003).
[17] M. Yasuda and H. Katori, Phys. Rev. Lett. **92**, 153004 (2004).
[18] S. G. Porsev and A. Derevianko, Phys. Rev. A **74**, 020502 (2006).
[19] D. N. Madsen and J. W. Thomsen, J. Phys. B **35**, 2173 (2002).
[20] K. T. Therkildsen *et al.*, Phys. Rev. A **79**, 034501 (2009).
[21] P. L. Hansen *et al.*, Phys. Rev. A **77**, 062502 (2008).
[22] R. Santra, K. V. Christ, and C. H. Greene, Phys. Rev. A **69**, 042510 (2004).
[23] P. Jönsson and C. F. Fischer, J. Phys. B **30**, 5861 (1997).
[24] R. H. Garstang, Astrophys. J. **148**, 579 (1967).
[25] G. Tachiev and C. Froese Fischer, http://nlte.nist.gov/MCHF (MCHF, *ab initio*).