

Isotope shifts of the $(3s3p) {}^3P_{0,1,2}-(3s4s) {}^3S_1$ Mg I transitions

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We report measurements of the isotope shifts of the $(3s3p) {}^3P_{0,1,2}-(3s4s) {}^3S_1$ Mg I transitions for the stable isotopes ${}^{24}\text{Mg}$ ($I=0$), ${}^{25}\text{Mg}$ ($I=5/2$), and ${}^{26}\text{Mg}$ ($I=0$). Furthermore, the ${}^{25}\text{Mg}$ 3S_1 hyperfine coefficient $A({}^3S_1)=(-321.6 \pm 1.5)$ MHz is extracted and found to be in excellent agreement with state-of-the-art theoretical predictions giving $A({}^3S_1)=-325$ MHz and $B({}^3S_1)\approx 10^{-5}$ MHz. Compared to previous measurements, the data presented in this work are improved up to a factor of 10.

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I. INTRODUCTION

Accurate measurements of atomic transitions play an important role in many parts of physics and astronomy. They form a basis for state-of-the-art atomic structure calculations and provide important reference data for spectroscopic measurements. Accurate spectroscopic measurements have a wide range of applications covering astrophysical [1] and laboratory-based experiments including optical cooling schemes and atomic clocks [2,3]. With the access to new and improved spectroscopic data, detailed models using relativistic many-body methods of atomic structure calculations and models may advance significantly [4,5]. For the alkaline-earth elements, some transitions are very well known, but most transitions are relatively unknown or known only with a modest accuracy.

The magnesium atom is interesting for a number of reasons. Magnesium has a particularly simple level structure among the two electron systems studied such as Ca, Sr, Ba, and Yb. This opens for detailed comparison to theoretical models [6,7]. Most of the transitions are in the blue part of the spectrum which makes the magnesium atom ideal for optical clock purposes. The black body shift, currently limiting the Sr atomic clock [3], is expected to be factor of 100 lower compared to the Sr case [8]. However, exploiting the magnesium system for an optical clock has been restricted due to lack of ultracold magnesium atoms. In the magnesium system, cooling to low temperatures using the intercombination line is not feasible and other methods has failed to efficiently cool samples below one mK [9,10]. The 517 nm transitions studied in this paper are suited for repumping purposes and can in combination with a cooling laser on the $(3s3p) {}^3P_2-(3s3d) {}^3D_3$ be used to efficiently cool magnesium atoms to micro-kelvin temperatures.

In the search for a possible temporal drift of the fine-structure constant $\alpha = \frac{e^2}{4\pi\epsilon_0\hbar c}$, accurate spectroscopic measurements on distant quasars have been carried out [12]. One of the sources of possible systematic effects is the difference

between isotopic abundance ratios in gas clouds in the early universe compared to those on earth. To test this possibility, it is necessary to have accurate isotope shifts for the relevant atomic transitions. Here, the $(3s3p) {}^3P_{0,1,2}-(3s4s) {}^3S_1$ transitions (see Fig. 1) play an important role and new data on the isotope shifts will be helpful [4]. An improvement of the experimental determination of the isotope shift will reflect on the precision of theoretical calculations, since magnesium is often used as a test ground for different methods of atomic calculations. The need for accurate isotope shifts is further motivated by the wish to study the isotopic evolution of the universe. The isotopic abundances of gas clouds may be measured independently of a variation of α [11]. This is important for testing models of nuclear reactions in stars and supernovae and of the chemical evolution of the universe.

In this Brief Report, we present measurements for the isotope shift and hyperfine structure splitting of the $(3s3p) {}^3P_{0,1,2}-(3s4s) {}^3S_1$ Mg I transitions around 517 nm in a metastable atomic magnesium beam. We improve previous

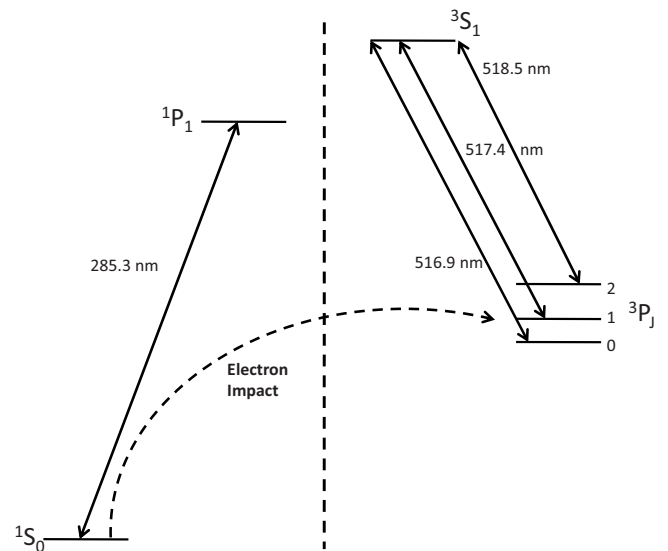


FIG. 1. Level diagram for ${}^{24}\text{Mg}$ showing transitions relevant for this Brief Report. There are three stable magnesium isotopes ${}^{24}\text{Mg}$, ${}^{25}\text{Mg}$, and ${}^{26}\text{Mg}$ with nuclear spins of $I=0$, $I=5/2$, and $I=0$, respectively. Dashed line indicates the separation between the singlet and triplet spin states. Note that for ${}^{25}\text{Mg}$, hyperfine structure is present.

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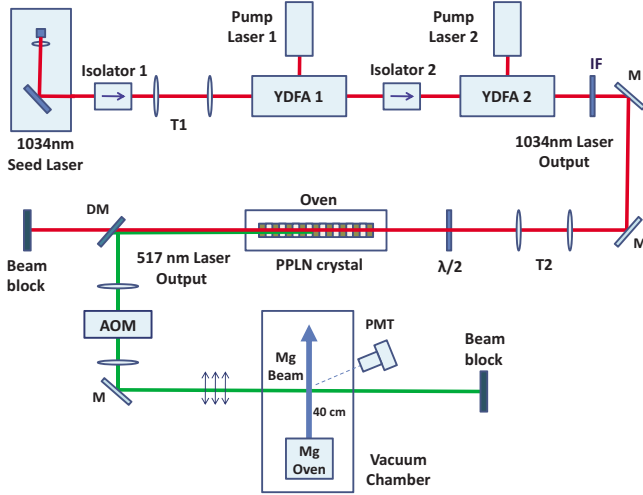


FIG. 2. (Color online) Schematic diagram of the laser system: T, telescope; DM, dichroic mirror; IF, interference filter; M, mirror; $\lambda/2$, half wave plates; BS, beam splitter; AOM, acousto-optic modulator; PMT, photomultiplier tube. A two-stage YDFA system is used for 1034 nm laser amplification and is single pass frequency doubled in a PPLN crystal. Fluorescence spectroscopy is performed on a metastable magnesium beam, approximately 40 cm from the oven orifice.

measurements by up to a factor of 10 for the stable isotopes ^{24}Mg ($I=0$), ^{25}Mg ($I=5/2$), and ^{26}Mg ($I=0$). For ^{25}Mg 3S_1 , we extract the hyperfine coefficient $A(^3S_1)$ and compare the result to state-of-the-art relativistic many-body calculations.

II. EXPERIMENTAL SETUP

Figure 2 shows the experimental setup used for spectroscopy on metastable magnesium atoms. The Mg oven is operated around 520 °C and produces an effusive magnesium beam with mean velocity 1000 m/s and flux of $\sim 10^{13}$ atoms/s. Electron impact initiates the discharge which runs at a stable current of about 1 A in a setup similar to the one described in [13]. Using the main 285.3 nm ($3s^2$) 1S_0 –($3s3p$) 1P_1 transition and switching the discharge, we estimate about 40% of the atoms are in a metastable state, distributed among the $^3P_{0,1,2}$ levels. The 517 nm light is produced from a fiber amplified diode laser centered at 1034 nm [14]. We use an external cavity diode laser in a Littrow configuration followed by a 40 dB optical isolator. Typically, the output after the isolator is 15 mW and about 10 mW is injected into a two-stage Yb-doped fiber amplifier (YDFA) system. The fiber consists of a highly doped single-mode core of 10 μm diameter with a peak absorption of 1200 dB/m at 976 nm and a large multimode pump guiding cladding of 125 μm in diameter. The fibers are pumped with up to 5 W at 976 nm. After the amplifier stage, 1.5 W of 1034 nm light is generated. The 1034 nm light is single pass frequency doubled in a periodically poled lithium niobate (PPLN) crystal generating up to 40 mW at 517 nm. The domain period of the PPLN crystal is 6.37 μm with a quasi-phase-matching temperature around 35 °C and temperature coefficient of 0.09 nm/K. An oven is used to stabilize the

PPLN temperature within 0.01 °C in order to achieve optimal phase matching. After the frequency doubling, a dichroic mirror is used to separate the 517 nm light from the 1034 nm light.

Spectroscopy is performed 40 cm from the oven orifice using linearly polarized light. The imaging system collects fluorescence from an area of about 8 mm² within the atomic beam limiting the residual Doppler effect to 60 MHz. A 275–400 MHz acousto-optic modulator (AOM) is used for frequency scale calibration. The absolute AOM frequency is controlled below 1 kHz rms and was verified using a precision counter. For calibration, both zero- and first-order beams from the AOM are overlapped producing a double set of spectra. Changing the dc offset of the AOM enables us to test the degree of linearity of the frequency scan. The intrinsic linewidth of the 517 nm light has been measured to be below 3 MHz using a high finesse cavity. Each spectrum is averaged 32 times and 30 different spectra are recorded for each transition.

III. METHOD OF CALCULATION

The method used for calculation of the magnetic-dipole and electric-quadrupole hyperfine structure constants A and B for the 3S_1 state is a combination of the configuration-interaction (CI) method with many-body perturbation theory (MBPT) [15]. Initially, the method was developed for calculating energy levels. The MBPT was used to construct an effective Hamiltonian for valence electrons. Then the multi-particle Schrödinger equation for valence electrons was solved in the frame of the CI method. Following the earlier works, we refer to this approach as the CI+MBPT formalism.

In this approach, the energies and wave functions are determined from the equation

$$H_{\text{eff}}(E_n)\Phi_n = E_n\Phi_n,$$

where the effective Hamiltonian is defined as

$$H_{\text{eff}}(E) = H_{\text{FC}} + \Sigma(E).$$

Here, H_{FC} is the Hamiltonian in the frozen core approximation and Σ is the energy-dependent correction, which takes into account virtual core excitations.

In order to calculate other atomic observables, one needs to construct the corresponding effective operators for valence electrons [16–18]. In particular, the effective operator of the hyperfine interaction used in this work accounts for the core-valence and core-core correlations. To account for shielding of an externally applied field by core electrons, we have solved random-phase approximation (RPA) equations, summing a certain sequence of many-body diagrams to all orders of MBPT [16,19].

We consider Mg as a two-electron atom with the core [$1s^2, \dots, 2p^6$]. On the whole, the one-electron basis set for Mg consists of $1s$ – $13s$, $2p$ – $13p$, $3d$ – $12d$, and $4f$ – $11f$ orbitals, where the core and $3, 4s$, $3, 4p$, $3, 4d$, and $4f$ orbitals are Dirac-Hartree-Fock (DHF) ones, while all the rest are the virtual orbitals. The orbitals $1s$ – $3s$ are constructed by solving the DHF equations in V^N approximation, $3p$ orbital is

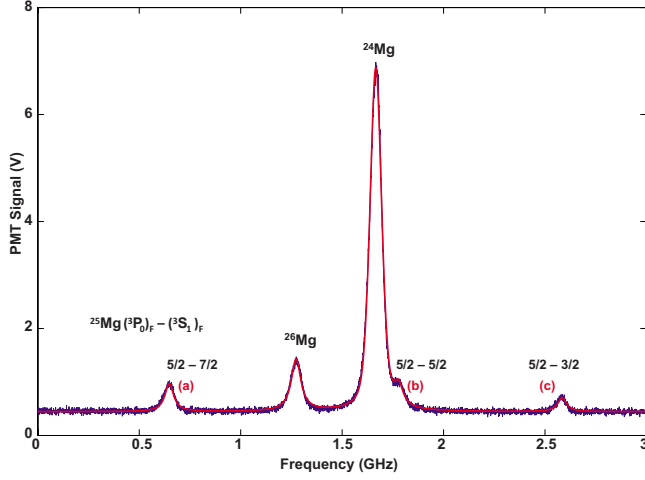


FIG. 3. (Color online) Fluorescence signal from the 3P_0 - 3S_1 transition. Blue curve represents the experimental data and red curve is a fit to the data. The hyperfine splittings of $^{25}\text{Mg}(^3P_0)_F - (^3S_1)_F$ are indicated as (a)–(c).

obtained in V^{N-1} approximation, and $4s$, $4p$, $3,4d$, and $4f$ orbitals are constructed in V^{N-2} approximation. We determined virtual orbitals using a recurrent procedure similar to Ref. [20] and described in detail in [17,18]. Configuration-interaction states were formed using this one-particle basis set which is sufficiently large to obtain numerically converged CI results. An extended basis set, used at the stage of MBPT calculations, included $1s$ – $19s$, $2p$ – $19p$, $3d$ – $18d$, $4f$ – $15f$, and $5g$ – $11g$ orbitals.

The results obtained in the frame of the CI+MBPT method for the hyperfine structure (hfs) constants A and B are $A(^3S_1) = -325$ MHz and $B(^3S_1) \approx 10^{-5}$ MHz. The theoretical value for the magnetic-dipole constant A is in a good agreement with the experimental value obtained in this work $A(^3S_1) = -321.6 \pm 1.5$ MHz.

The electric-quadrupole hfs constant $B(^3S_1)$ is very close to zero. In the absence of configuration interaction of the $3s4s$ configuration with other configurations (such as $3snd$ and nd^2 configurations, where $n \geq 3$), the hfs constant B would be exactly equal to zero, but (very weak) configuration interaction leads to a nonzero value of B , though very small.

IV. RESULTS AND DISCUSSION

Figures 3–5 show typical fluorescence spectra as a function of laser frequency for the $^3P_{0,1,2}$ - 3S_1 transitions. The blue curve is the raw data and the red curve is a Voigt profile fit to the data. For ^{24}Mg , we obtain a full width at half maximum (FWHM) linewidth of 55 MHz in agreement with our beam and detector geometry. From the spectra, we clearly identify the isotope shift and the hyperfine components ($^3P_{0,1,2})_F - (^3S_1)_F$ of ^{25}Mg . The hyperfine coefficients of the $^3P_{1,2}$ levels could be extracted from the data, however, we use the values measured in [21] due to the very high precision of these measurements to extract the hyperfine coefficient $A(^3S_1)$. We find the hyperfine coefficient

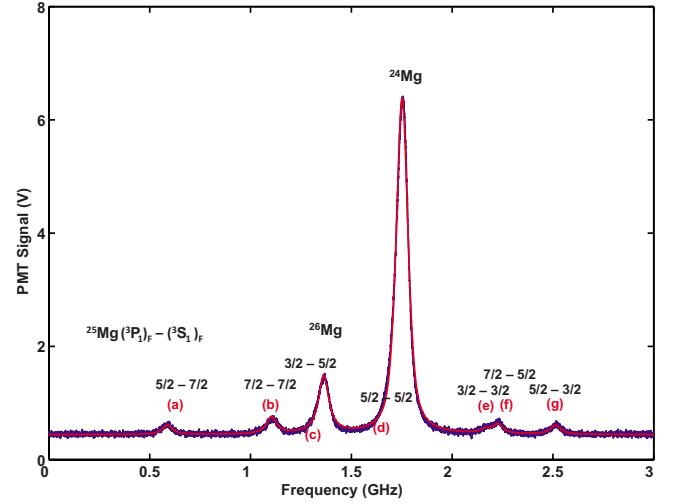


FIG. 4. (Color online) Fluorescence signal from the 3P_1 - 3S_1 transition. Blue curve represents the experimental data and red curve is a fit to the data. The hyperfine splittings of $^{25}\text{Mg}(^3P_1)_F - (^3S_1)_F$ are indicated as (a)–(g).

$A(^3S_1) = (-321.6 \pm 1.5)$ MHz. As described in Sec. III, the electric-quadrupole hfs constant $B(^3S_1)$ is significantly smaller than the resolution of the experiment and is therefore set to zero in the fitting procedure. Table I summarizes our findings and compares to previous measurements. Different systematic errors have been investigated as mentioned in Sec. II, all of which have been determined to be lower than the statistical error. Errors listed in Table I are pure statistical errors. Here, it can be seen that the value for the hyperfine coefficient $A(^3S_1)$ and the isotope shifts agree with previous measurements within the uncertainty.

We observe the ^{24}Mg - ^{26}Mg shift to be almost constant which indicates that relativistic isotope shift effects are small or comparable to the statistical error in our measurement. Our measured isotope shifts are accounted for by pure mass effect. In this case, the ratio between the ^{24}Mg - ^{26}Mg and ^{24}Mg - ^{25}Mg shifts can be expressed as [23]

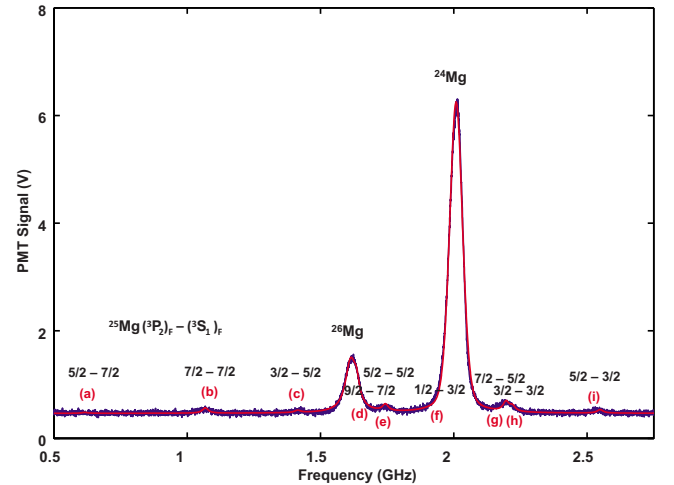


FIG. 5. (Color online) Fluorescence signal from the 3P_2 - 3S_1 transition. Blue curve represents the experimental data and red curve is a fit to the data. The hyperfine splittings of $^{25}\text{Mg}(^3P_2)_F - (^3S_1)_F$ are indicated as (a)–(i).

TABLE I. Measured isotope shift and 3S_1 hyperfine structure constant. The errors listed are the statistical errors.

	$^3P_0-^3S_1$		$^3P_1-^3S_1$		$^3P_2-^3S_1$		$A(^3S_1)$ (MHz)
	Δ^{24-26} (MHz)	Δ^{24-25} (MHz)	Δ^{24-26} (MHz)	Δ^{24-25} (MHz)	Δ^{24-26} (MHz)	Δ^{24-25} (MHz)	
Ref. [22] (1949)	414 ± 9		366 ± 45		414 ± 12		-322 ± 6
Ref. [23] (1978)	396 ± 6	210 ± 36	391 ± 4.5	201 ± 21	393 ± 7.5	204 ± 7.5	-329 ± 6
Ref. [24] (1990)	391 ± 10	214 ± 10	393 ± 10	215 ± 10	397 ± 10	217 ± 10	-322 ± 6
This work (2009)	391.3 ± 1.7	205.7 ± 1.5	390.1 ± 1.4	209.1 ± 1.3	394.4 ± 0.8	205.7 ± 0.8	-321.6 ± 1.5

$$\frac{\Delta(^{24}\text{Mg} - ^{26}\text{Mg})}{\Delta(^{24}\text{Mg} - ^{25}\text{Mg})} = \frac{26 - 24}{26 \cdot 24} \frac{25 \cdot 24}{25 - 24} = \frac{25}{13} \simeq 1.92. \quad (1)$$

We obtain the ratios $^3P_0 \rightarrow ^3S_1$ 1.90 ± 0.02 , $^3P_1 \rightarrow ^3S_1$ 1.87 ± 0.01 and $^3P_2 \rightarrow ^3S_1$ 1.92 ± 0.01 . These values are consistent with previous results [23,24].

V. CONCLUSION

In this paper, we present improved data for the isotope shift of the Mg $^3P_{0,1,2}-^3S_1$ and the hyperfine coefficient

$A(^3S_1)$ for the ^{25}Mg isotope. We find good agreement between state-of-the-art many-body theory and experimental results. Experimental values reported here are improved by up to a factor of 10 compared to previous studies.

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