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Frequency measurement of the $^3P_1$-$^3P_0$ fine-structure transition of ^{24}Mg

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A frequency measurement of the fine-structure transition $^3P_1$-$^3P_0$, $\Delta m_j=0$ in the first metastable triplet of ^{24}Mg is reported. The experiment has been performed on a metastable atomic beam interacting with the submillimetric radiation ($\lambda \simeq 0.5$ mm) of a phase-locked backward wave oscillator. The transition is observed via the fluorescence emission at 4571 Å of the intercombination line $^3P_1$-$^1S_0$. The center frequency was evaluated to be $\nu = 601\,277.16 \mp 0.05$ MHz with precision limited by first-order Doppler effect.

High spectroscopic interest is currently associated with the observation and frequency measurement of fine-structure transitions within metastable triplets of alkaline earths. This is mainly due to the fact that the development of sources, signal handling, and measurement techniques in the submillimeter wavelength region is quite recent and a whole new field of precision spectroscopy is now open in a range where mostly theoretical data were available in the past. Experimental determinations of energy separations, dipole moments, Landè interval rule anomalies, gyromagnetic factors, hyperfine splittings, and isotope shifts are to be expected, among others. The ability to perform precise absolute frequency measurements with the aid of signals synthesized with reference to microwave standards offers ground for comparison with sophisticated theories.

The case of Mg is particularly interesting. The presence of three stable isotopes (^{24}Mg, ^{25}Mg, and ^{26}Mg), of which the two even ones do not show hyperfine structure ($I=0$) while the odd one does ($I=\frac{5}{2}$), forms a nice system for the study of isotopic shifts; uncertain knowledge of the departure from pure Russel-Saunders *LS* coupling demands precise measurements of energy separations; and the twelve-year-old proposal[1] for a frequency standard based on the $^3P_1$-$^3P_0$, $\Delta m_j=0$ transition of ^{24}Mg can only now be analyzed with proper technology.[2-4]

A simplified diagram of the first energy levels of Mg is shown in Fig. 1. Transitions from the $^3P_2$ and $^3P_0$ levels to ground are strictly forbidden for even isotopes, while decay from $^3P_1$ is weakly allowed, due to incomplete Russel-Saunders (RS) coupling, and produces the well-known intercombination line of alkaline earths. Decay time for Mg is estimated to be $\tau = 2.4$ ms (Ref. 5) [4 ms (Ref. 6)], and the natural linewidth of the $^3P_1$-$^3P_0$ transition is therefore expected to be smaller than 1 kHz, limited by the lifetime of $^3P_1$. For this reason, among others, this transition ($\nu \simeq 0.6$ THz) was proposed for use as clock transition in a frequency standard.[1] The magnetic dipole moment μ of the π ($\Delta m_j = 0$) transition is $\sqrt{2/3}\mu_B$ in the RS approximation, μ_B being the Bohr magneton.

The block diagram of the system used for the observation of the 0.5-mm Mg transition is reported in Fig. 2.

A tantalum oven operating above 500 °C produces a beam of Mg atoms in the ground level; excitation to the ^3P levels is obtained by means of an electrical discharge sustained by the beam itself right near the oven aperture and stabilized by a magnetic field. The full divergence of the atomic beam after collimation is 10 mrad; at the normal operating temperature of 520 °C the atomic flux has been evaluated to be $\Phi_0 = 8\times 10^{13}$ atoms/s on the basis of the Taylor-Langmuir formulas, taking into account collision scattering at the oven aperture.[2] From the evaluation of the fluorescence light intensity at 4571 Å it has been possible to estimate an efficiency $\eta \sim 30\%$ in the production of metastable atoms.[2] This quite satisfactory value has been obtained by optimizing discharge parameters; in particular, it must be taken into account that the cross section for atom-electron collisions populating the ^3P levels is narrowly peaked at 6 eV [the levels mainly involved in this process are ^3S and ^3D (Fig. 1)]. The ratio between the fluorescence signal and its shot noise observed in the experimental setup with a photomultiplier of 5 cm diam at 10 cm from the beam, 1.5 m from the discharge, was 2×10^4; long-term fluctuations of the signal level are smaller than 1%.

Atomic transitions from $^3P_0$ to $^3P_1$ are induced in the metastable atoms 1.1 m downstream after partial depletion

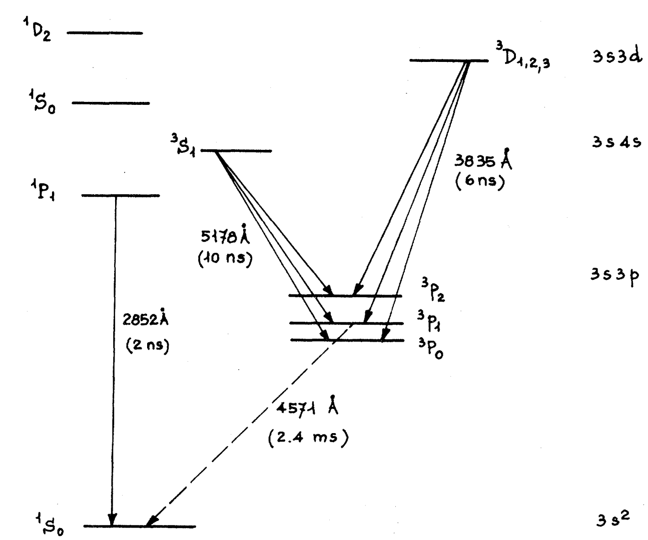

FIG. 1. Energy-level diagram of Mg. The value 2.4 ms is taken from Ref. 5.

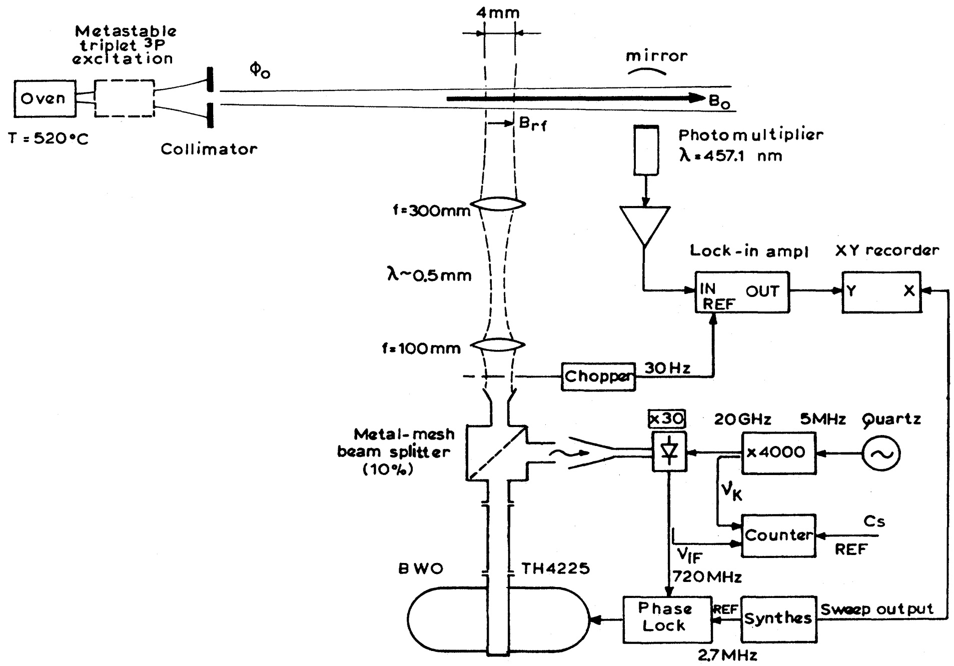

FIG. 2. Experimental setup for the observation of the $^3P_1$-$^3P_0$ Mg transition.

through decay of the $^3P_1$ level, and are detected from an increase of the intercombination line fluorescence, which is expected to be about 20% at optimum microwave power in the conditions of the present experiment. Optimum power density is 2.3 mW/mm^2 for an interaction time of 10 μs; the power necessary for saturating the transition with single pass in the used configuration can be evaluated to be about 140 mW.[2]

The sub-mm radiation at 0.5 mm is generated by a backward wave oscillator (BWO) especially designed for the experiment (Thompson TH4225); its frequency ranges from 590 to 610 GHz and at 601 GHz the output power is $\simeq 5$ mW. A telescopic system couples the output overmoded BWO waveguide (2×1 mm) to the atomic beam; a single-pass interaction was adopted in order to simplify the measurement procedure since the frequency to be measured was not known better than 0.3%.[7] The half-power Gaussian beam diameter in the interaction region is 4 mm and the interrogating power is lower than 1 mW. The quantization axis, provided by a static magnetic field B_0 as shown in Fig. 2, is parallel to the rf H field for the observation of the π transition. The relative frequency instability of the BWO when free running is $\simeq 10^{-6}$ for measuring times from 10 ms to 100 s; it needs therefore to be locked to a stable

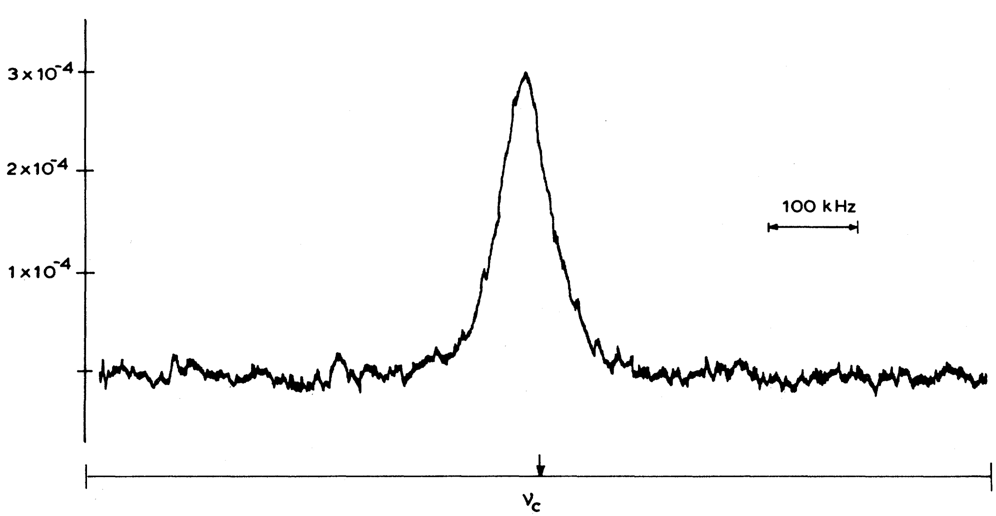

FIG. 3. $^3P_1$-$^3P_0$ ^{24}Mg π transition. Center sweep frequency $\nu_c = 601\,277.200$ MHz; vertical axis: fluorescence level relative to background; equivalent video bandwidth: 3 mHz.

reference for use in sub-Doppler spectroscopy. This is realized in this system by phase-locking the BWO to a 5-MHz quartz-crystal oscillator via a low-noise-frequency multiplication chain. The reference and the i.f. signals are sent to an electronic counter referred to a Cs atomic clock. More details about the BWO phase lock and the problems related to high-order frequency multiplication are reported in Refs. 3 and 4. The coherent linewidth of the phase stabilized submm radiation is smaller than the spectrum analyzer selectivity (100 Hz); the signal-to-phase noise ratio at 600 GHz is 33 dB in 1-kHz bandwidth.

Coherent detection of the fluorescence light intensity versus the BWO radiation frequency is used as indicated in Fig. 2. In fact, the resonance signal with the available power is expected not to exceed 0.1% of the background fluorescence; signal-to-noise ratio in 1-Hz bandwidth is therefore lower than 10 in our configuration. The resonance signal is reported in Fig. 3 as observed with an equivalent detection bandwidth of 3 mHz. The frequency at the peak is 601 277.18 MHz and the full width at half maximum of the line is 70 kHz, in good agreement with the estimated transit-time broadening.

The first-order Doppler shift due to imperfect orthogonality has been experimentally evaluated from the shape asymmetry between the two curves obtained by tilting the BWO beam the same amount on either side.

The resulting best estimate of the unperturbed transition is 601 277.16 $\mp$0.05 MHz.

The error bar was taken as the distance between two opposite Doppler-shifted and obviously broadened curves ($\pm$2.3° from the narrowest observed line).

Magnetic field dependence of the observed transition was evaluated by superimposing first derivative curves for two B_0 field values, with slightly different amplitudes so that they would cross (Fig. 4). No shift bigger than 2 kHz for a field change from 10 to 20 G was observed, in agreement with the expected behavior of hyperfine-structure-free even isotopes.[1]

Search for other lines of higher intensity was carried out as far as 80 MHz each side of the first one, with no result, unambiguously indicating that the observed resonance is due to the most abundant isotope (^{24}Mg). In fact, isotopic shift is expected to be well within this interval for these transitions.[8]

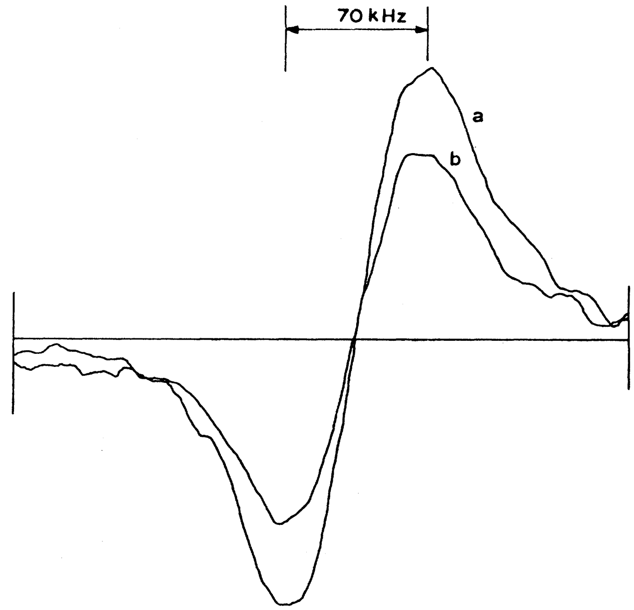

FIG. 4. First derivative of the resonance for two field values: (a) $B_0=20$ G; (b) $B_0=10$ G.

Precision improvements in the determination of unperturbed frequency include use of a standing wave in the cavity for Doppler-free interrogation and optimum power operation, two-zone Ramsey-type interaction for linewidth reduction, and increase in detection efficiency.

Possible applications are high-resolution spectroscopic measurements as indicated above and frequency metrology.

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