Absolute frequency measurement of the $7s^2\, ^1S_0 - 7s\, ^1P_1$ transition in $^{225}\Ra$

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Transition frequencies were determined for transitions in Ra in an atomic beam and for reference lines in $\text{Te}_2$ molecules in a vapor cell. The absolute frequencies were calibrated against a GPS stabilized Rb-clock by means of an optical frequency comb. The $7s^2\, ^1S_0(F = 1/2)-7s\, ^1P_1(F = 3/2)$ transition in $^{225}\Ra$ was determined to be 621 042 124(2) MHz. The measurements provide input for designing efficient and robust laser cooling of Ra atoms in preparation of a search for a permanent electric dipole moment in Ra isotopes.

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Radium (Ra) is the heaviest alkaline earth metal and it offers unique possibilities for measuring parity and time reversal symmetry violation. The particular atomic and nuclear [1-8] structure in Ra isotopes cause the largest enhancement for permanent electric dipole moments (EDMs) [9] in any atom. This arises from the close proximity of the $7s\, ^1P_1$ and $7s\, ^3D_2$ states [1]. The exploitation of the enhancement from this 5 cm$^{-1}$ separation requires precise knowledge of Ra atomic properties such as the absolute frequencies of transitions that are relevant for laser cooling and state manipulation (see Fig. 1). Many isotopes of Ra are available from radioactive sources such as $^{229}\text{Th}$ [10,13], or at online isotope production facilities such as ISOLDE, CERN, Switzerland [14,15].

A sensitive search for EDMs requires efficient collection of the atoms in an optical trap because of the low abundance of Ra isotopes. A strategy for efficient laser cooling and trapping has been developed with the chemical homologue barium (Ba). Exploiting the strong $6s^2\, ^1S_0-6s\, ^1P_1$ transition [16,17] resulted in an efficiency of $\sim 1\%$ for slowing and capturing Ba from an atomic beam, whereas capture efficiencies of below $10^{-6}$ were reported for Ra when using the weak intercombination transition $7s^2\, ^1S_0-7s\, ^3P_1$ [13].

The optical spectrum of Ra was first studied by Ras-mussen [18]. This identified Ra as an alkaline earth metal. Hyperfine splittings and isotope shifts were determined for the $7s^2\, ^1S_0-7s\, ^7p\, ^1P_1$ transitions by collinear laser spectroscopy [14,15] with intense Ra ion beams at ISOLDE, lifetimes of low lying states were determined in ANL, USA [9,19]; no absolute frequencies are quoted.

Here we report on a laser spectroscopy measurement of the frequency of the strong $7s^2\, ^1S_0-7s\, ^7p\, ^1P_1$ transition in $^{225}\Ra$ in an atomic beam. The laser frequency was recorded with an optical frequency comb. A set of reference lines in molecular tellurium ($^{130}\text{Te}_2$) were measured simultaneously. An uncertainty of about 1 MHz is achieved. This is sufficiently small for efficient laser cooling on that transition compared to the natural linewidth of 30 MHz [4].

Doppler-free saturated absorption spectroscopy of $^{130}\text{Te}_2$ provides a reliable secondary frequency standard over a wide range in the visible spectrum. The absence of nuclear spin results in a spectrum without hyperfine structure. Many of the linear absorption lines are listed in the $^{130}\text{Te}_2$ atlas [20] which reports the analysis of Fourier transform spectroscopy in the wavelength range of 450 nm to 600 nm. Several lines have been independently calibrated to MHz accuracy in interferometric measurements for particular experiments, e.g. for $1S-2S$ transitions in hydrogen, deuterium, positronium and muonium [21] or strong lines around 500 nm [22]. Deviations of up to 0.003 cm$^{-1}$ (100 MHz) from the values in reference [20] have been found. This makes independent calibration of individual reference lines indispensable. The strongest $\text{Te}_2$ line in the vicinity of the $7s^2\, ^1S_0(F = 1/2)-7s\, ^7p\, ^1P_1(F = 3/2)$ transition in $^{225}\Ra$ is at 20 715.477 7 cm$^{-1}$ (Line 2004 in [20]).

Isotopically pure $^{130}\text{Te}_2$ contained in a quartz cell of dimensions (100 mm $\times$ 25 mm) is heated to temperatures between 700 K and 800 K for sufficient vapor density. The temperature is maintained by a Watlow heater element of inner diameter 50 mm and length 150 mm. It is monitored by a K-type thermocouple placed next to the cell. Light for the transitions in $^{225}\Ra$ and $^{130}\text{Te}_2$ is obtained through second harmonic

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generation (SHG) by feeding light from a Ti:Sapphire laser (Coherent MBR110) at wavelength 966 nm into a KNbO₃ crystal inside a linear enhancement cavity (see Fig. 2). The light at wavelength 483 nm is focused into an acousto-optic modulator (AOM1, MT350, AA Optoelectronics) operated at frequency \( f_{\text{AOM1}} = 450 \text{ MHz} \). It provides a frequency offset, which bridges the separation between the nearest line in \(^{130}\text{Te}_2\) and the transition in \(^{225}\text{Ra}\). The 0\(^{1}\)st-order beam from AOM1 serves for saturated absorption in \(^{225}\text{Te}_2\) and the minus first-order beam is used for spectroscopy on \(^{225}\text{Ra}\). For the latter the light is overlapped with an effusive \(^{225}\text{Ra}\) atomic beam from an oven filled with 30 \(\mu\)g of \(^{229}\text{Th}\) (half-life 7340 y), corresponding to source strength 10 \(\mu\)Ci. The oven is contained inside a UHV vacuum chamber with residual gas pressure below \(10^{-9}\) mbar. Typically \(^{225}\text{Ra}\) atoms were accumulated for one week inside the cavity of the oven, which when heated to temperature 900 K produced a flux of \(10^6 \text{s}^{-1} \text{cm}^{-2}\) for about one hour. Optical windows for laser beam access enable the alignment of the laser beam orthogonal to the atomic Ra beam. Fluorescence light is collected through an interference filter under right angle to both beams with a photomultiplier tube (PMT).

The light for \(^{130}\text{Te}_2\) spectroscopy is passed through a thick beam splitter plate (BS) to provide two probe beams and one pump beam. The parallel probe beams are passed through the \(^{130}\text{Te}_2\) cell. The pump beam at intensity 400(50) \(\mu\)W/mm\(^2\) is...
focused into an acousto-optic modulator (AOM2) operated at $f_{\text{AOM2}} = 60$ MHz. The driving rf power is chopped at $f_{\text{mod}} \approx 13$ kHz to provide amplitude modulation of the beam. This light is overlapped on a polarizing beam splitter cube (PBS) with one of the probe beams in the $^{130}$Te$_2$ cell. The difference in absorption for both probe beams is measured on a balanced amplifier to obtain the saturated absorption signal. The vapor pressure in the $^{130}$Te$_2$ cell is monitored by linear absorption with light at sufficiently known frequency. $n_1$ and $n_2$ are the sideband diffraction orders from AOM1 and AOM2, while $f_1$ and $f_2$ are the respective operating frequencies. Fig. 4 and Fig. 5 display saturated (top) and linear absorption (bottom) spectra in $^{130}$Te$_2$. The repetition rate $f_{\text{rep}}$, the offset frequency $f_{\text{CEO}}$ and the operation parameters for AOM1 and AOM2 were all kept constant while the IR light frequency was scanned across the transitions. Six of the recorded lines are displayed in Fig. 6. The measured saturated absorption lines are fitted with Lorentzian line shapes,

$$L(\omega) = \frac{1}{2\pi} \frac{\Gamma}{(\omega - \omega_0)^2 + \Gamma^2/4},$$

where $\omega_0$ is the center frequency and $\Gamma$ is the width of the profile.

Linear absorption of the pump beam was measured by transmitting light through the $^{130}$Te$_2$ cell for vapor pressures ranging from 0.2 mbar to 1.6 mbar. Saturated absorption was measured at cell temperature 804(5) K and Te$_2$ vapor pressure $\sim$1.6 mbar. For all individual saturated absorption lines frequency uncertainties due to the fitting procedure are of order 0.1 MHz. The linewidths of the transitions is between 11 and 29 MHz while the signal strengths varied by a factor 125 in the range from line Te#0 (Fig. 3) to line Te#51 (Fig. 4 (inset)). The frequencies and wavenumbers for the saturated absorption lines are listed in Table I. The pressure shift of similar lines in $^{130}$Te$_2$ has been determined previously to be 1 MHz/mbar [24, 26, 30].

### Table I: Wavenumbers, frequencies and relative strengths of saturated absorption lines in $^{130}$Te$_2$. The uncertainty of the wavenumber measurement for line Te#0 is 0.000 005 cm$^{-1}$ corresponding to 1.5 MHz for the optical frequency. We quote both frequencies and wavenumbers for the transitions to enable comparison with the $^{130}$Te$_2$ atlas [20] and previous measurements [22, 24, 25].

| Saturated absorption line in Te Atlas | Measured Wave number $\nu$ (cm$^{-1}$) | Measured Frequency $\nu$ (MHz) | Relative strength $\xi$ |
|--------------------------------------|------------------------------------------|-----------------------------|----------------------|
| -3                                   | 20 715.222 52 621 026 747.7(1.0)         | 0.23                        |
| -1                                   | 20 715.440 56 621 033 284.4(1.2)         | 0.37                        |
| 0 # 2004                             | 20 715.468 86 621 034 132.9(1.5)         | 1.00                        |
| 1                                    | 20 715.494 32 621 034 896.2(1.6)         | 0.09                        |
| 2                                    | 20 715.534 21 621 036 092.1(0.7)         | 0.34                        |
| 3                                    | 20 715.593 74 621 037 876.7(1.6)         | 0.19                        |
| 4                                    | 20 715.607 49 621 038 288.8(1.5)         | 0.09                        |
| 5                                    | 20 715.651 12 621 039 596.9(1.6)         | 0.14                        |
| 51                                   | 20 715.721 48 621 041 706.3(1.7)         | 0.008                      |
| 6                                    | 20 715.770 29 621 043 169.4(1.5)         | 0.16                        |
| 80 # 2008                            | 20 716.989 05 621 079 707.1(0.2)         | 0.12                        |
| 81                                   | 20 717.088 18 621 082 678.8(0.7)         | 0.21                        |
| 82                                   | 20 717.150 13 621 084 536.0(0.8)         | 0.03                        |
| 83                                   | 20 717.341 67 621 090 278.2(0.4)         | 0.06                        |
| 90 # 2009                            | 20 717.436 66 621 093 126.1(0.3)         | 0.15                        |
| 91                                   | 20 717.460 39 621 093 837.3(0.4)         | 0.16                        |
During the measurements the maximum observed fluctuation of the $^{130}$Te$_2$ cell temperature was ± 5 K. This limits the frequency uncertainty due to pressure shifts for the lines measured in $^{130}$Te$_2$ to below 0.2 MHz.

![Graph showing fluorescence intensity vs. frequency](image)

**FIG. 7:** (Color online) Doppler-free laser induced fluorescence from the $^1S_0(F = 1/2) - ^1P_1(F’ = 3/2)$ transition in $^{225}$Ra. The data are averaged over five individual scans of duration 50 s. The line is fitted with a Lorentzian function ($\chi^2/1 = 1.256$).

**TABLE II:** The frequency of the $^1S_0 - ^1P_1$ transition in $^{226}$Ra has been determined in different experiments. In this work the frequency of the $^1S_0(F = 1/2) - ^1P_1(F’ = 3/2)$ transition in $^{225}$Ra was measured. The frequency of this transition in $^{226}$Ra is obtained by exploiting this measurement, the known isotope shift in the $^1S_0 - ^1P_1$ transition $\Delta f^{226Ra} = f^{226Ra} - 2236(15)$ MHz [15], and the magnetic dipole interaction constant $A^{(1P_1)} = 2796.5(2.5)$ MHz of $^{225}$Ra [14].

| Isotope | Transition | Frequency (MHz) | Experiment |
|---------|------------|----------------|------------|
| $^{226}$Ra | $^1S_0 - ^1P_1$ | 621 038 489 (15) | This work |
| $^{225}$Ra | $^1S_0 - ^1P_1$ | 621 038 004 (180) | [19] |
| $^{225}$Ra | $^1S_0 - ^1P_1$ | 621 041 362 (1 500) | [18] |

The absolute frequency of the $^1S_0(F = 1/2) - ^1P_1(F' = 3/2)$ transition in $^{225}$Ra is measured with saturated absorption line Te#51 in $^{130}$Te$_2$ as a reference. The transition frequency in Ra is obtained through

$$f_{Ra} = f_{Te} + n_1f_1 - \frac{n_2f_2}{2} + \Delta f_{Te,Ra},$$

(3)

where $\Delta f_{Te,Ra}$ is the observed frequency difference between the reference line and the $^{225}$Ra transition. We find the transition in $^{225}$Ra (see Fig. 7) to be centered 418(1) MHz below the reference. This yields an absolute frequency of $f^{225Ra} = 621 042 124(2)$ MHz, respectively 20715.73542(6) cm$^{-1}$. Together with the isotope shift in this transition of $\Delta f^{226Ra} = 2236(15)$ MHz [15] and the magnetic dipole interaction constant $A^{(1P_1)} = 2796.5(2.5)$ MHz for $^{225}$Ra [14] we have the transition frequency $f^{226Ra} = 621 038 489(15)$ MHz for this transition in $^{226}$Ra (see Table II). The measurement here was performed with a retroreflected beam (see Fig. 2). The overlap angle of both beams was better than 1 mrad, corresponding to a separation $\leq 1$ mm of the counter-propagating beams at distance 1 m from the interaction region. The beam alignment was optimized by minimizing the linewidth of the fluorescence signal. This causes residual first-order Doppler shift $\Delta_{Doppler} \leq 2$ MHz for the thermal atomic beam in our experiment.

We exploited an offline atomic beam of $^{225}$Ra for Doppler-free laser induced spectroscopy on the $^1S_0 - ^1P_1$ transition. It is the main transition for efficient slowing of atoms to within the capture range of a magneto optical trap. Our measurement of the transition frequency with uncertainty 2 MHz is an improvement by some two orders of magnitude over a previous measurement for the same transition in $^{225}$Ra. This together with the calibration of several lines in Te$_2$ molecules to accuracy MHz provides crucial input for the design of a sensitive search for an EDM in atomic Ra.

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