Measurement of the infrared $^2F_{7/2}$-$^2D_{5/2}$ transition in a single $^{171}Yb^+$ ion

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The $^2F_{7/2}(F=3)-^2D_{5/2}(F=2)$ 3.43-μm transition in $^{171}Yb^+$ has been measured to be 87 366 282 258(91) kHz. This transition is of interest as an infrared frequency standard. A subsidiary measurement of the 3.43-μm ($F=4$-$F'=3$) transition yields a frequency of 87 362 471 500(800) kHz. Taken with a previous measurement of the $^2D_{5/2}$ level hyperfine splitting, a $^2F_{7/2}$ level hyperfine splitting of 3620(2) MHz and an A factor of 905.0(0.5) MHz are deduced. In addition, a 3.43-μm transition isotope shift of $\nu_{171}-\nu_{172}=+4048(4)$ MHz is obtained from these experiments and a known value for the transition in the 172 isotope. These measurements taken with previous work give a value for the $^2S_{1/2}(F=0)-^2F_{7/2}(F=3)$ electric octupole transition of 642 121 497 308(178) kHz, which will be used as a starting point in the search for this extremely weak transition. [S1050-2947(99)04610-7]

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I. INTRODUCTION

The ion trapping technique offers the possibility of frequency standards of exceptional reproducibility. A single laser-cooled ion, contained in the low perturbation environment of an ion trap, allows good reproducibility to be attained. The choice of ion also has an influence on the reproducibility of the standard. The linear Zeeman shift of a state is given by $\Delta E=g_F\mu_B B m_F$, and hence transitions from $m_F=0$ to $m_F'=0$ states are free from this effect, making them much less sensitive to magnetic fields. Both $^{199}Hg^+$ and $^{171}Yb^+$ have such transitions and the relatively simple hyperfine structure given by a spin half-nucleus. This, and the amenability of these ions to laser cooling makes them attractive as potential frequency standards [1–6]. Other ions are also of interest, for example In^+ [7,8] has a clock transition insensitive to the effect of the quadrupole shift—another important systematic effect in an ion trap standard.

Ytterbium is perhaps the most versatile of all the ion species under investigation, having clock transitions in the microwave [3], infrared [9], and optical [4,5,10] regions of the spectrum. This work investigates the infrared transition at 3.43 μm for use as a frequency standard. Previous work on this transition [9,11] has used the technically easier 172 isotope, which has no hyperfine structure. Here the more difficult though metrologically advantageous 171 isotope is used. The occurrence of hyperfine structure complicates both laser cooling and the observed spectra.

The 3.43-μm $^2F_{7/2}$-$^2D_{5/2}$ transition in Yb^+ has significant potential as a midinfrared frequency standard. One feature of this transition is its proximity to the 3.39-μm methane stabilized He-Ne laser frequency. Currently this laser system is used in many frequency chains as a bridge between the microwave and optical regions of the spectrum [12–14]. Since the $^2F_{7/2}$-$^2D_{5/2}$ transition in ytterbium is less than one terahertz from this standard, it should be relatively easy to incorporate it into current frequency chains. A standard based on the 3.43-μm transition in ytterbium should have improved performance over the 3.39-μm He-Ne due to its higher Q of 4×10^{12} and the low perturbation environment of the ion trap.

The $^2F_{7/2}$-$^2D_{5/2}$ 3.43-μm transition is unusual for an ion trap standard since the transition is formed between two excited states (see Fig. 1). To use this reference transition, the ion must be prepared in the lower of these states, prior to interrogation by the 3.43-μm laser. In the current work this is achieved by driving the 411-nm $^2S_{1/2}$-$^2D_{5/2}$ transition, and subsequently populating the $^2F_{7/2}$ level by spontaneous decay. An alternative would be to prepare the initial state by driving the 467-nm $^2S_{1/2}$-$^2F_{7/2}$ transition. This transition is also under investigation at the National Physical Laboratory as a potential optical frequency standard [5]. This leads to the intriguing possibility of combining both an optical and infrared standard in a single device [9]. Using the 467-nm transition for state preparation would also have considerable benefit to the infrared standard. Since, in this case, the ion is prepared in the $^2F_{7/2}$ ($m_F=0$) state, it would then be possible to selectively drive the 3.43-μm $m_F=0$ to $m_F'=0$ transition. As mentioned, the insensitivity of such transitions to magnetic fields would improve the infrared standard's reproducibility. In addition, the 467-nm $^2S_{1/2}$-$^2F_{7/2}$ transition has an exceptionally narrow natural linewidth, due to the 10-year lifetime of the excited state. This will allow an optical standard of unprecedented stability.

This work presents a measurement of the 3.43-μm $^2F_{7/2}$-$^2D_{5/2}$ transition in the 171 isotope of ytterbium. This measurement is part of an ongoing program to locate the 467-nm $^2S_{1/2}$-$^2F_{7/2}$ transition in the 171 isotope of ytterbium. When combined with a previous measurement of the $^2S_{1/2}$-$^2D_{5/2}$ transition [15], it provides a starting frequency for a search for the very weak 467-nm transition. Measurement of a second hyperfine transition also allows the hyperfine splitting of the $^2F_{7/2}$ level to be deduced.

The following section contains a description of the experimental arrangement and procedure. Next is a description of the measurement itself, focusing on the metrology used to determine the 3.43-μm frequency and the errors involved. The final section concerns the hyperfine structure of the $^2F_{7/2}$ level and the isotope shift of the 3.43-μm transition.

II. EXPERIMENTAL ARRANGEMENT

The aim of this experiment is to measure the frequency of the $^2F_{7/2}$-$^2D_{5/2}$ transition at 3.43 μm (see Fig. 1). This in-

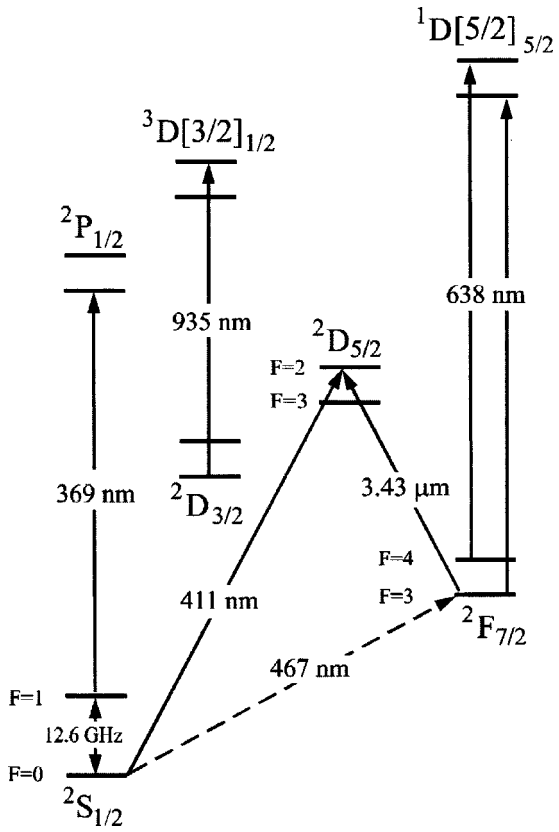

FIG. 1. Partial term scheme of $^{171}Yb^+$. The 369-nm, 935-nm, and 12.6-GHz transitions are used for laser cooling. The 411-nm transition is used to prepare the $^2F_{7/2}$ level. The 3.43-μm reference transition is being probed. The 638-nm transition is used to return the ion to the cooling cycle, should the 3.43-μm transition not be driven.

frared reference transition occurs between two excited states. The lower of these, the $^2F_{7/2}$ level, has a lifetime of 10 years [5] and so forms an effective ground state for the infrared transition. A variation of the quantum jump technique is used to detect the infrared reference. A single ion of ytterbium is held in a Paul trap and laser-cooled by driving a strong transition, from which fluorescence is used to monitor the state of the ion. The $^2F_{7/2}$ level is first populated by driving a visible transition. An absence of fluorescence for an extended period indicates that the ion is trapped in this long-lived level. Once prepared, the infrared reference is driven and detected by the return of fluorescence. If the fluorescence does not resume, a further laser is used to quickly return the ion to the cooling cycle via a high-lying level.

In this experiment the ion is prepared in the $^2F_{7/2}$ level by driving the 411-nm $^2S_{1/2}$-$^2D_{5/2}$ transition. From the $^2D_{5/2}$ level, the branching ratio [10] to the $^2F_{7/2}$ level is 83%. The $^2D_{5/2}$ level has a lifetime [10] of 7 ms, thus if fluorescence has not returned after 70 ms, the ion is established to be in the $^2F_{7/2}$ level with 99.995% certainty. Next the 3.43-μm laser is used to interrogate the $^2F_{7/2}$-$^2D_{5/2}$ transition. If fluorescence returns after the interrogation cycle, the infrared transition has been successfully driven and the event is recorded as a quantum *on* jump. If fluorescence does not return, the ion is still in the $^2F_{7/2}$ level. Either the transition has not been driven, or having been driven the ion has decayed back to the $^2F_{7/2}$ level. In either case, no quantum jump is recorded and the ion is quickly returned to the cooling cycle by driving the $^2F_{7/2}$-$^1D[5/2]_{5/2}$ transition at 638 nm. A quantum jump profile is built up by performing a number of 3.43-μm interrogations and recording the number of *on* jumps at each frequency step.

In this experiment the $^2F_{7/2}$ level is prepared by decay from the $^2D_{5/2}$ level and hence it is impossible to control which m_F sublevel becomes populated. It is therefore impossible to selectively drive the $m_F=0$-$m_F'=0$ component. However, in order to simply observe the 3.43-μm transition and measure its frequency at the part in 10^{10} level, this is not necessary. It is, however, expedient to work in a small magnetic field so that all the Zeeman components lie within the laser linewidth allowing the transition to be driven with high efficiency on resonance. A magnetic field is applied to the ion using three pairs of orthogonal coils. The ion does not fluoresce in zero magnetic field, due to coherent population trapping [15]. This effect is used to null out the ambient laboratory field. A single $^{172}Yb^+$ ion is used to null the field before the main experiment is started, since the fluorescence null is about 100 times narrower in this isotope than in ytterbium 171 [15]. In order to select the $\Delta m_F=0$ components of the 3.43-μm transition, a small field of 14 μT is then applied in an appropriate direction. This field also selects the $\Delta m=\pm 1$ components of the 411-nm transition during the state preparation of the $^2F_{7/2}$ level via the $^2D_{5/2}$ level. During the cooling cycle, a further large magnetic field of 1 mT is applied so that appreciable fluorescence is observed. This large field, applied via a fourth pair of coils, is switched off during the clock interrogation. The field is applied at 45° to the 369-nm laser polarization to ensure that both $\Delta m_F=0$ and ± 1 transitions are driven.

A schematic diagram of the experimental apparatus is shown in Fig. 2. A single ion of $^{171}Yb^+$ is confined in an electrodynamic (Paul) trap. The trap consists of a ring and two endcap electrodes with an ac voltage applied to the ring, forming a pseudopotential well. In addition, a dc voltage is applied to the ring to make the axial and radial secular frequencies approximately equal at $2\pi\times 1.1$ MHz. The rf-photon correlation technique [16] is used to minimize the ion's excess micromotion in the direction of the cooling laser beam. Tight confinement of the ion in the Lamb-Dicke regime eliminates the first-order Doppler effect. The ion is laser cooled by driving the $^2S_{1/2}(F=1)$-$^2P_{1/2}(F=0)$ transition at 369 nm. The $^2P_{1/2}$ level can decay into the metastable $^2D_{3/2}$ level, therefore to maintain the cooling cycle this level is depopulated by driving a transition at 935 nm [17]. To depopulate the $^2S_{1/2}(F=0)$ state, the ground-state microwave transition at 12.6 GHz is driven by the amplified output from an HP83732B synthesizer. The fluorescence from the cooling transition is collected with a high numerical aperture lens and detected by a photomultiplier.

The 3.43-μm-light is generated by difference-frequency-mixing 1064- and 812-nm laser radiations in a crystal of $AgGaS_2$. This system is described in more detail in Ref. [9]. The narrow-linewidth 1064-nm light is generated with a Lightwave Nd:YAG laser and the 812-nm light by a grating-stabilized diode laser (SDL5422-H1). The frequency stability

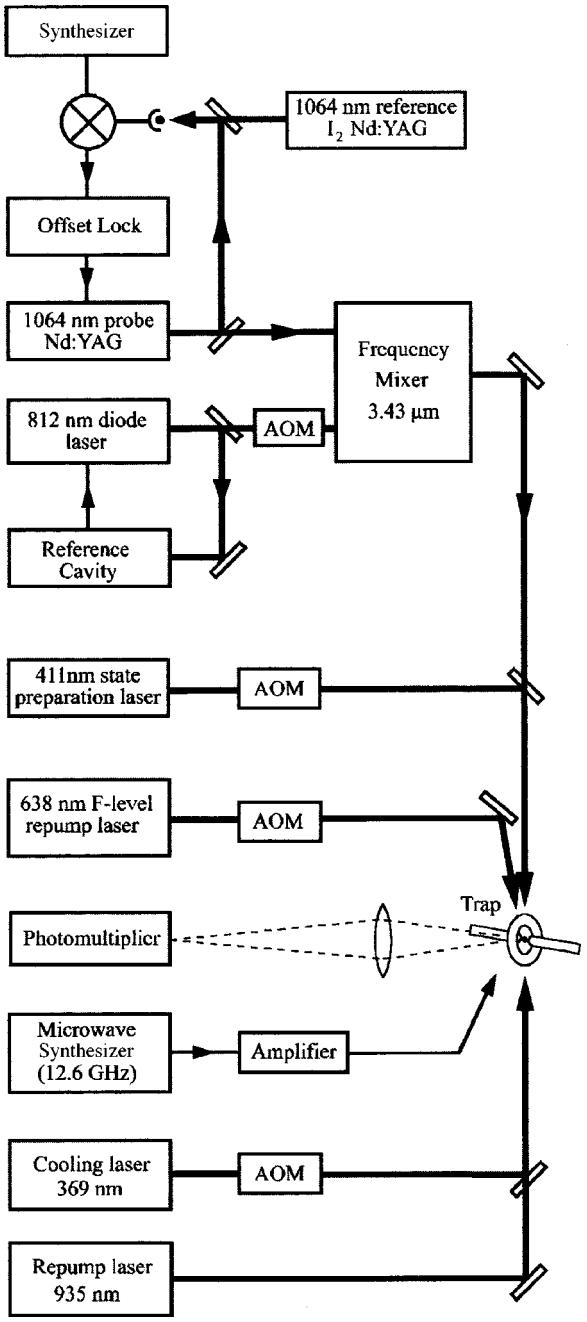

FIG. 2. Experimental arrangement. AOM denotes acousto-optic modulator.

of the 3.43-μm output is improved by individually stabilizing the 812- and 1064-nm radiations, as described in the next section. The overall frequency linewidth of the 3.43-μm source, estimated to be a few hundred kilohertz, is dominated by the diode laser linewidth.

The experiment is controlled by a computer, which is used to count fluorescence, scan the 3.43-μm laser frequency, and sequence the lasers, microwaves, and magnetic field. Acousto-optic modulators are used to switch the laser radiations; in particular, the 3.43-μm radiation is switched by gating the 812-nm light. The large 1-mT magnetic field, microwaves, 411-nm light, and cooling radiation are switched off during the measurement cycle so that the ion is not significantly perturbed.

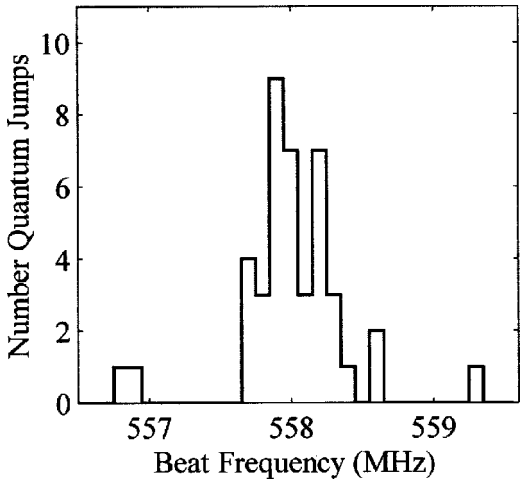

FIG. 3. Quantum jump profile of the 3.43-μm $^2F_{7/2}(F=3)$-$^2D_{5/2}(F=2)$ transition in $^{171}Yb^+$.

III. MEASUREMENT OF THE $^2F_{7/2}$-$^2D_{5/2}$ TRANSITION

To record the absolute frequency of the 3.43-μm transition, a quantum jump profile was taken of the relevant hyperfine transition. The 3.43-μm laser frequency was deduced from individual measurements of the 812- and 1064-nm laser frequencies.

The frequency of the 812-nm diode laser was measured by interferometric comparison with a 633-nm iodine-stabilized He-Ne laser [18], using the NPL 1-m etalon intercomparator [19,20]. The 812-nm laser was stabilized to an ultralow-expansivity (ULE) glass reference cavity. The laser frequency was measured at the beginning and end of a data-taking session to record any temperature-induced frequency drift of the reference cavity and hence the 812-nm frequency.

The frequency of the 1064-nm Nd:YAG probe laser was measured by beating it against the fundamental output of a Nd:YAG reference laser [21]. The reference laser has both 1064-nm fundamental and 532-nm frequency doubled outputs. The frequency doubled output is used to stabilize the laser to the a_1 component of the 32-0, $P(54)$ line in $^{127}I_2$. The Bureau International des Poids et Mesures (BIPM) quotes the frequency of this iodine component as 563 212 634 588(40) kHz, when operated under the recommended conditions [22]. When this transition was measured at the NPL [21], essentially the same frequency was measured but with a slightly larger error bar of 70 kHz. This larger error is due in part to the slightly worse reproducibility of the NPL system, and so this error is used in the analysis presented here.

The 1064-nm outputs from both the reference and probe Nd:YAG lasers were focused onto a fast photodiode to form the beat signal. This beat was downshifted by mixing it with a synthesiser and fed to an offset lock box. The offset lock box maintained the downshifted beat at 4.000(1) MHz, by feeding back the integrated error signal to the 1064-nm Nd:YAG probe laser. Scanning the synthesiser frequency scans the frequency of the Nd:YAG probe laser. The synthesiser frequency, and hence the beat frequency, was recorded along with the number of 3.43-μm induced quantum jumps that occurred at each frequency step.

The measurement of the 3.43-μm transition frequency is

TABLE I. Measurement errors (1σ standard uncertainty) for the 3.43-μm $F=3$-$F'=2$ transition. The entry entitled ''determination of the mean'' is the 1σ standard uncertainty of the mean of the data shown in Fig. 3.

Error source	Magnitude (kHz)
812-nm frequency intercomparison	53
Iodine-stabilized Nd:YAG (at 1064 nm)	35
Determination of line center	61
Asymmetry of profile	23
Acousto-optic offset	1
Root sum of squares total	91

made up of a number of parts: the beat frequency between the two Nd:YAG lasers, the frequency of the Nd:YAG reference laser, and the frequency of the 812-nm diode laser. It was also necessary to take into account the $-80.005(1)$-MHz shift induced by the AOM used to chop the 812-nm radiation. The quantum jump profile for the 3.43-μm $^2F_{7/2}(F=3)$-$^2D_{5/2}(F=2)$ transition is shown in Fig. 3. The resonance consists of five unresolved $\Delta m_F=0$ Zeeman components, which have an end-to-end splitting of 90 kHz due to the 14-μT field. The width of the resonance is therefore dominated by the spectral linewidth of the 3.43-μm source. The relatively poor statistics of the data are in part due to the fact that the 3.43-μm transition is observed only 20% of the time on resonance due to the $^2D_{5/2}$ branching ratio. The beat frequency for the line center is derived from the mean of these data. The 1-σ standard deviation of the data is 390 kHz. The standard error of the mean is therefore $\sigma/\sqrt{N-1}$ with $N=42$. This is the ''determination of line center'' error of 61 kHz shown in Table I. The remaining errors for the measurement are also summarized in Table I. The entry ''asymmetry of profile'' is an upper estimate of the possible systematic shift due to an asymmetric initial population of the $^2F_{7/2}(F=3)$ level. The initial population of the $^2F_{7/2}$ state depends on which 411-nm $\Delta m=\pm 1$ transition is driven. Driving the 411-nm $\Delta m=+1$ component populates the $^2F_{7/2}(F=3,m_F=2,1,0)$ states and driving the $\Delta m=-1$ component populates the $^2F_{7/2}(F=3,m_F=-2,-1,0)$ states. Hence the 3.43-μm $\Delta m=0$ line shape can be systematically pulled by up to ± 23 kHz (i.e., the separation of the means of the two groups of components, given an end-to-end splitting of 90 kHz).

The 3.43-μm $F=3$-$F'=2$ transition was measured on two separate days. The results from each day's measurements were consistent. The first day's results had a much larger error due to imperfect temperature control of the 812-nm reference cavity. This temperature control was improved significantly for the second day's measurement. The weighted mean of the two measurements is therefore dominated by the second day's results. The results of this measurement give a 3.43-μm $F=3$-$F'=2$ transition frequency of 87 366 282 258(91) kHz.

IV. HYPERFINE SPLITTING AND ISOTOPE SHIFT

A second 3.43-μm measurement was performed, this time on the $F=4$-$F'=3$ hyperfine component. The purpose of this measurement was to deduce a value for the $^2F_{7/2}$ level hyperfine splitting, using the already measured values for the $^2D_{5/2}$ hyperfine splitting. Using this data and previous work, it was also possible to deduce the isotope shift between the 171 and 172 isotopes.

The frequency measurement of the $F=4$-$F'=3$ hyperfine component was carried out in an identical manner to that of the preceding section, yielding a center frequency of 87 362 471 500(800) kHz. The much larger error is due to a failure of the temperature control of the 812-nm cavity, which caused a large frequency drift between the beginning and end of the experiment. The error budget in Table I is modified for this measurement by changing the determination of the line-center error to 800 kHz.

Using the two 3.43-μm transition frequencies along with the previously measured $^2D_{5/2}$ hyperfine splitting [15] yields a value for the $^2F_{7/2}$ hyperfine splitting of 3620(2) MHz. This implies a hyperfine A factor of 905.0(0.5) MHz. There is no hyperfine B factor as the nuclear spin $I=\frac{1}{2}$.

Knowledge of the hyperfine splittings of the $^2F_{7/2}$ and $^2D_{5/2}$ levels allows the centroid frequency of the 3.43-μm transition to be deduced as 87 364 134.8(1.6) MHz. This can be taken along with a previous measurement of the 3.43-μm transition in the 172 isotope [11], to give a 3.43-μm isotope shift of $\nu_{171}-\nu_{172}=+4048(4)$ MHz.

V. CONCLUSION

The $^2F_{7/2}(F=3)$-$^2D_{5/2}(F=2)$ 3.43-μm transition in $^{171}Yb^+$ has been measured to be 87 366 282 258(91) kHz. A subsidiary measurement of the $F=4$-$F'=3$ hyperfine component yields a value of 87 362 471 500(800) kHz. From these measurements and previous work, a $^2F_{7/2}$ level hyperfine splitting of 3620(2) MHz is inferred, which implies an A factor of 905.0(0.5) MHz. A 3.43-μm isotope shift of $\nu_{171}-\nu_{172}=+4048(4)$ MHz has also been deduced.

The $^2S_{1/2}(F=0)$-$^2F_{7/2}(F=3)$ 467-nm transition frequency has been deduced as 642 121 497 308(178) kHz, from the measurement of the $^2F_{7/2}(F=3)$-$^2D_{5/2}(F=2)$ 3.43-μm transition and a previous measurement of the $^2S_{1/2}(F=0)$-$^2D_{5/2}(F=2)$ 411-nm transition [15]. This value defines a small frequency region that can be searched to locate the extremely weak $^2S_{1/2}(F=0)$-$^2F_{7/2}(F=3)$ electric octupole transition in $^{171}Yb^+$. This transition has previously been driven in the technically easier $^{172}Yb^+$ isotope [5]. The switch to the 171 isotope is desirable due to its metrological superiority.

In the present experiment, the preparation of the $^2F_{7/2}$ state was performed via spontaneous decay from the $^2D_{5/2}$ level. This rather uncontrolled means of state preparation made it impossible to selectively populate the $^2F_{7/2}(F=3,m_F=0)$ level and hence drive the Zeeman-free component efficiently. Once the 467-nm $^2S_{1/2}(F=0,m_F=0)$-$^2F_{7/2}(F=3,m_F=0)$ transition has been successfully driven, this route could be used to populate the $m_F=0$ level with 100% efficiency prior to the 3.43-μm interrogation. This leads to the possibility of a high stability and reproducibility optical/infrared standard operating in the same device.

The $^2D_{5/2}$ state lifetime of 7.2 ms gives a natural linewidth of 22 Hz for the $^2F_{7/2}$-$^2D_{5/2}$ transition. Using a high spectral purity laser system and an interrogation time of, say, 1 ms, a fractional frequency stability of $6\times 10^{-14}\tau^{-1/2}$ is

possible. Coupled with a likely reproducibility of better than one part in 10^{15}, the 3.43-μm $^2F_{7/2}$-$^2D_{5/2}$ transition in $^{171}Yb^+$ could make a future replacement for the 3.39-μm methane stabilized He-Ne frequency standard.

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