We investigate a technique of detecting weakly allowed optical transitions that have poor signal-to-noise ratio (SNR) when using conventional frequency sweep methods. This is of importance given the resurgence of isotopic shift spectroscopy used to search for beyond Standard-Model signatures. Forbidding transitions in neutral atoms, such as the $^1S_0 - ^3P_0$ transition in group-II-like atoms may be used in such investigations. Knowledge of isotopic shifts is also relevant to many radiopharmaceutical applications. We demonstrate a means to increase the sensitivity of detecting weak transitions and apply it to the clock transition in the composite fermions of ytterbium. The method is applicable to any transition in atoms that undergo cyclic preparation before probing. With regard to searching for atomic transitions, the technique is appropriate where the ratio of search space in frequency, $\Delta f_s$, to transition linewidth, $\Delta v$, is not excessively large. For example, when the transition frequency can be estimated using the knowledge of isotope shifts or in the case of photoassociation spectroscopy, estimated using a Le Roy–Bernstein type equation. The gain in detection sensitivity is made by cycling the probe light and looking for a corresponding signal in the Fourier domain. For example, if we refer to each atom loading and cooling cycle as one cycle, then for subsequent cycles the probe light alternates between on and off. When on resonance, some fraction of atoms will be excited to the upper state every second cycle. This appears as a modulation of the fluorescent received by the photodetector, which is proportional to the number of atoms in the ground state. The strength of the modulation is extracted through the FFT of a time record of the photoreceiver’s signal. We demonstrate the sensitivity of the method by performing spectroscopy on the $^1S_0 - ^3P_0$ line in $^{171}$Yb and $^{173}$Yb, which have natural linewidths of approximately 40 mHz. A Fourier signal with $\sim 10$ dB SNR is observed for the $^{171}$Yb clock line with $\sim 10^3$ atoms. We note that the technique should be well suited to searching for clock transitions in bosonic isotopes when held in an optical lattice trap. It could also be applied in the search for photoassociation resonances in ultra cold molecules, for example, at predicted frequencies, where otherwise they are not apparent.

The experiment is comprised of a magneto-optical trap (MOT), a frequency comb (MenloSystems FC1500), a hydrogen maser (KVARZ-75A), cooling lasers at 399 nm and 556 nm, and the clock-line laser at 578 nm to probe the $^1S_0 - ^3P_0$ transition. The main components are shown in Fig. 1. The source of ytterbium atoms is provided by an oven at $\sim 400^\circ$C from which atoms effuse through an array of narrow collimation tubes orientated horizontally. Some fraction of atoms...
pass through a narrow conduit downstream that separates the vacuum chamber into two main sections: (i) the oven and (ii) the Zeeman slower and main chamber.

The MOT operates in two stages: first with 399 nm light acting on the $^1S_0 - ^1P_1$ transition, then 556 nm light acting on the $^1S_0 - ^3P_1$ transition to reduce the atomic temperature to ~50 µK. A dual tapered Zeeman slower (crossing zero magnetic field) assists with loading atoms into the MOT. The 399 nm light is generated by frequency doubling 798 nm light from a Ti:sapphire laser in a resonant cavity. The 556 nm light is produced by use of a second frequency doubling cavity, where the incident 1112 nm light originates from a fibre laser (NKT Photonics) that injection locks a semiconductor laser (EYP-RWL-1120). The 399 nm light is stabilized by locking to the $^1S_0 - ^1P_1$ line in atoms progressing to the Zeeman slower. The 556 nm light is stabilized by locking the sub-harmonic at 1112 nm to a mode of the frequency comb via a frequency-to-voltage converter (FVC). The cooling laser signals are controlled with acousto-optic modulators (AOMs) driven with amplified RF signals from voltage controlled oscillators (VCOS).

The clock line laser at 578 nm is produced by use of a third frequency doubling cavity, where the master laser is an extended cavity diode laser (LD1001 Time-Base) and Hänisch-Couillaud frequency stabilization is used with the doubling cavity. The yellow light frequency is tuned with an additional AOM (AOM-1 in Fig. 1) whose RF is set with a synthesizer (Agilent E4428c) that is referenced to the H-maser (with laser current, respectively. The sign of the RF drive frequency of the AOM in the path of the 578 nm light. The event sequence is presented in Fig. 3. Each cycle has a duration of 0.5 s, the majority of which is used to load atoms drifts at a rate of 20.3 mHz·s$^{-1}$. The free spectral range of the cavity is 1496.5210(1) MHz. This is found by measuring the frequency separation between consecutive modes of the cavity with the frequency comb and the 1157 nm beat signal. A more accurate determination is then found using the $^{171}$Yb clock transition frequency. By locking to the 173167th mode of the cavity and offsetting the 578 nm light by ~284.5 MHz with AOM-1 the $^1S_0 - ^3P_1$ transition frequency can be reached. The $^1S_0 - ^3P_1$ transition for $^{171}$Yb lies approximately midway between cavity modes, which makes frequency offsetting with AOMs difficult. To overcome this obstacle one could use offset sideband locking with the ultrastable cavity. Instead, our approach is to lock the 1157 nm laser to the frequency comb, which also gives flexibility in reaching the $^1S_0 - ^3P_1$ transition (rather than using a sequence of AOMs). There are some constraints because the comb is also used to stabilize the 556 nm light frequency, and beat signals can only lie at frequencies set by the bandpass filters; here, 21 MHz and 30 MHz for the 1112 nm and 1157 nm beats, respectively.

The setup for the probe laser frequency stabilization by use of a FVC (Analog Devices AD652) is shown in Fig. 1. After filtering and amplifying the beat signal from the avalanche photodiode it is divided in frequency by 128 with a prescaler (Fujitsu MB506). A comparator (LM360) regularizes the waveform making it suitable for the CMOS compatible FVC. The AD652 is a synchronous VFC, implying that its transfer function is governed by an external clock, in this case an OCXO at 10 MHz, divided by 4. A laser servo based on this scheme has been shown to produce a fractional frequency instability of $\sim 3 \times 10^{-14}$ for $0.2 < \tau < 10^3$ s, where $\tau$ is the integration time. Apart from the flexibility provided by locking to the comb, the FVC-comb lock is also extremely robust.

\[ v = 2(n f_R - f_O \pm f_B) + f_{aom} \]

where $f_R$ and $f_O$ are the frequency comb’s mode spacing and offset frequency, respectively ($f_O = 20$ MHz), $f_B$ is the beat frequency, $n$ is the mode number of the comb, and $f_{aom}$ is the RF drive frequency of the AOM in the path of the 578 nm light. The sign of $f_B$ may depend on the isotope. The comb’s repetition rate is controlled by mixing its fourth harmonic with a ~1.00 GHz signal that is the sum of ~20 MHz and 980.0 MHz signals provided by a direct digital synthesizer (DDS) and a dielectric resonator oscillator (DRO), respectively. Hence $f_R = (f_{DDS} + f_{DRO})/4$. Both the DRO and DDS are locked to a 10 MHz signal from the H-maser. The DDS frequency is imposed by that of the 556 nm cooling light.

For the $^{171}$Yb clock transition the 1157 nm laser can be stabilized in one of two ways (summarized in Fig. 1): (i) with use of an ultrastable cavity and Pound-Drever-Hall lock, or (ii) by locking to the frequency comb using the FVC technique. The ultrastable cavity (Stable Laser Systems) and laser frequency stabilization have been described previously. The coefficient of thermal expansion is zero at 29.7(2) °C, and the second order thermal expansion coefficient is $6.6 \times 10^{-10}$ K$^{-2}$ (for $v = 259.1$ THz). The laser when locked to the cavity...
FIG. 3. Event sequence for the light fields. The 578 nm probe remains off every second cycle. The duration \( \Delta t_p = 40 \text{ ms} \). The saturation intensities are \( I^{(399)}_S = 595 \text{ W m}^{-2} \) and \( I^{(556)}_S = 1.4 \text{ W m}^{-2} \) for the \( 1S_0 - 1P_1 \) and \( 1S_0 - 3P_1 \) transitions, respectively. The frequency detuning of the cooling lasers is also varied to optimize signal level.

into the MOT. The 399 nm light is ramped down over 20 ms during which time the second stage cooling with 556 nm light takes effect. The 556 nm intensity is also ramped down to reduce the temperature of the atoms and minimize the ac Stark shift. The magnetic quadrupole field is held fixed with a z-axis gradient of 0.42 T m\(^{-1}\). The yellow 578 nm light pulse has a period of 40 ms to interact with the atoms, after which the 399 nm is switched back on and detection of the ground state atoms is made (and MOT loading recommences). Unless otherwise stated, the maximum intensity of the 578 nm light was 5 kW m\(^{-2}\) with a corresponding Rabi frequency, \( \Omega_2 \), of \( \sim 4 \text{ kHz} \) (representing \( \sim 1/50^{th} \) of the transition linewidth).

A 1 Hz modulation in the population transfer arises by applying the 578 nm light every second cycle. Fluorescence at 399 nm is filtered both spatially and spectrally before detecting the 578 nm light every second cycle. The frequencies subtracted from the line centers are \( 9 \text{ kHz} \) and \( 6 \text{ kHz} \), which is the same modulation shifted in frequency through aliasing.

A full line spectrum of the \( 1S_0 - 3P_1 \) transition is produced by stepping the frequency of AOM-1 (with a fixed frequency for each time record) and extracting the SNR from the FFTs. Figures S5(a) and S5(b) show the \( 1S_0 - 3P_0 \) line spectra for the \( ^{171}\text{Yb} \) and \( ^{173}\text{Yb} \), respectively. Each data point is determined from a separate FFT. Figure S5(a) is shown with a logarithmic scale to illustrate the sensitivity in the wings of the profile. The absolute frequency is determined from Eq. 1. The frequencies subtracted from the line centers are \( \nu_{171} = 518295836590.9 \text{ kHz} \) and \( \nu_{173} = 518294576847.6 \text{ kHz} \).

The inset of Fig. S5(a) shows the spectrum by use of a conventional frequency sweep for \( ^{171}\text{Yb} \) with a similar number of atoms. In this case the 578 nm pulse is applied every cycle of the event sequence, unlike in Fig. 3. Here the total sampling time was 140 s with the BCA set to 3-sample averaging. Note, the noise here is not white frequency noise.

To compare the detection sensitivity of the two methods we introduce a sensitivity index \( d' = (\mu_s - \mu_n)/\sigma_n \), where \( \mu_i \) is
the mean of the signal level, $\mu_s$ is the mean of the noise level, and $\sigma_s$ is the standard deviation of the noise. Here $\mu_s$ corresponds to the on-resonance signal. For the FFT approach the units are magnitude-squared, therefore, $d_{\text{fft}} = \sqrt{(SNR_{\text{fft, or}}^2)}$. In the case of the conventional sweep we obtain $d_{\text{swp}} = 3.3$, hence $d_{\text{fft, or}}^2 = d_{\text{swp}}^2 = 23$; or in power units, the gain in sensitivity is $\sim 27$ dB. Hence the means of detecting the transition over similar time scales is very much enhanced through the FFT approach.

The spectra in Figs. 5(a) and Fig. 5(b) have their own SNR. In this case we do not use the noise floor of the FFT to set the noise level. More appropriate is the rms of the residuals to the line shape fits. Here the SNR is 38 and 16 for $^{171}$Yb and $^{173}$Yb, respectively. The conventional scan for $^{173}$Yb produced no evident transition over the 140 s of sampling, due to the lower number of atoms trapped. The total sample time for Figs. 5(a) and Fig. 5(b) was 1500 s and 1300 s, respectively, so while there is a significant gain in SNR, the total sampling time increased by $\sim 10$ fold. From the Lorentzian line shape fits (to suit the wings) the FWHM for traces (a) and (b) are $210$ kHz and $290$ kHz, respectively. The width for $^{173}$Yb is consistent with the temperature of the atoms determined by imaging of ballistic expansion, $T \sim 50 \mu K$. For $^{173}$Yb there may be a linewidth contribution from laser noise, since the FVC lock to the comb was used rather than the ultrastable cavity.

Figure 6(a) shows the on-resonance 1 Hz SNR versus the number of atoms contributing to the signal for $^{171}$Yb with $\Omega_R = 4$ kHz and $10$ kHz (varied through $578$ nm intensity). The number of atoms is determined by use of $N_a = V/(hGf\gamma_p)$, where $V$ is the dc voltage from the PMT (background subtracted), $h$ is Planck’s constant, $f$ is the frequency of the $399$ nm light, $G$ is the gain of the PMT (V/W), $f$ is the collected fraction of fluorescence, and $\gamma_p$ is the photon scattering rate. The scattering rate follows from: $\gamma_p = s_0\gamma/(1 + s_0 + (2\delta/\gamma)^2)/2$, where $s_0$ is the $399$ nm intensity normalized by the saturation intensity, $I_s^{(399)}$, $\delta$ is the frequency detuning and $\gamma$ is the natural linewidth of the $^1S_0 - ^1P_1$ transition. The atom number was varied by changing the current through the Zeeman slower coils. With $\sim 10^3$ atoms a Fourier signal of $\gtrsim 10$ dB SNR remains apparent when $\Omega_R = 10$ kHz. The sensitivity increases with $578$ nm intensity. Note, our fluorescence collection efficiency is only $0.55 \%$.

FIG. 6. (a) SNR of the $1$ Hz FFT signal, on resonance, versus atom number for two values of $\Omega_R$. (b) Frequency of the $^{171}$Yb $^1S_0 - ^3P_0$ transition versus $556$ nm light intensity. Inset: spectrum of the $^{171}$Yb clock line after optimization. GSF, ground state fraction.

Once an atomic line is found, the modulated BCA output can aid optimization, such as improving the probe beam overlap with the atom cloud. An example of the $^{171}$Yb clock line recorded with the conventional scan is shown in the inset of Fig. 6(b) after such an optimization (and $T_{\text{even}} = 410 ^\circ$C). The green light intensity during the $578$ nm pulse was $I/I_s^{(556)} = 140$. By repeating with different levels of $556$ nm light the ac-Stark shift becomes evident, as seen in Fig. 6(b). The line center for zero light intensity is within $9$ kHz of previous reports of the transition frequency. The gradient of the line fit for the ac-Stark shift is $0.19(3)$ kHz W$^{-1}$ m$^{-2}$, where the uncertainty is dominated by that of the light intensity.

In summary, we have demonstrated a sensitive method of detecting weak optical transitions in cold atoms that rely on cyclic routines. In terms of detecting the presence of a transition, the sensitivity index is increased 20-fold compared to conventional line scanning for similar measurement times. One can detect (and resolve) a clock transition in an optical lattice with $\sim 10^3$ atoms, but to our knowledge the detection of such a transition with $10^3$ atoms in a MOT has not been previously demonstrated. With regard to line spectra, the increased SNR comes at the expense of a 10-fold increase in sampling time, which places a restriction on $\Delta f/\Delta f$ (mentioned in the opening). An upper limit for $\Delta f/\Delta f$ may be $\sim 20$ for practical purposes, but it depends on factors such as cycle time and how well search procedures can be automated.

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**DATA AVAILABILITY**

The data that support the findings of this study are available from the corresponding author upon request.

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