Magnetic Field-Induced Spectroscopy of Forbidden Optical Transitions with Application to Lattice-Based Optical Atomic Clocks

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We develop a method of spectroscopy that uses a weak static magnetic field to enable direct optical excitation of forbidden electric-dipole transitions that are otherwise prohibitively weak. The power of this scheme is demonstrated using the important application of optical atomic clocks based on neutral atoms confined to an optical lattice. The simple experimental implementation of this method—a single clock laser combined with a dc magnetic field—relaxes stringent requirements in current lattice-based clocks (e.g., magnetic field shielding and light polarization), and could therefore expedite the realization of the extraordinary performance level predicted for these clocks. We estimate that a clock using alkaline-earth-like atoms such as Yb could achieve a fractional frequency uncertainty of well below 10^{-17} for the metrologically preferred even isotopes.

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The long coherence times provided by "forbidden" excitation to long-lived atomic states are critical in several important applications including quantum computing and optical atomic clocks [1,2]. However, the search for narrower lines can lead to transitions between states with undesirably complex structure, such as levels with relatively large angular momentum. An important example of an ultranarrow transition is the $^1S_0 \rightarrow {}^3P_0$ spin- and angular momentum-forbidden clock transition in alkaline-earth-like atoms. Two of these atoms, Sr and Yb, are being pursued as strong candidates for lattice-based optical atomic clocks [3–8]. The long interaction times, high signal-to-noise ratio, and vanishing Doppler-related uncertainties provided by tight confinement of an atomic ensemble to individual optical lattice sites should lead to clocks with exceptional short-term stability and high accuracy in this rapidly developing field.

Current experimental work on optical lattice clocks [9] focuses on the $^1S_0 \rightarrow {}^3P_0$ transition in the odd isotopes of Sr and Yb [3,5,6,8], which is weakly allowed due to hyperfine mixing. In contrast to their even counterparts that have zero nuclear spin, however, the odd isotopes have an uncomfortably large sensitivity to magnetic fields (MHz/T) and lattice light polarization. They also have multiple ground state sublevels that considerably complicate spectroscopic line shapes. The method described in this Letter uses a small magnetic field (~ 1 mT) to mix a small, controllable fraction of the nearby $^3P_1$ state into the $^3P_0$ state, thereby allowing single-photon excitation of the $^1S_0 \rightarrow {}^3P_0$ clock transition for the even isotopes. In this way we can replace the odd isotopes with the even in lattice-based atomic clocks, which should greatly accelerate their progress. Estimates for Yb using experimentally realistic linewidths (1 Hz) show that induced frequency shifts can be controlled at a level that could enable a fractional frequency uncertainty of below 10^{-17} for a lattice clock. In contrast to multiphoton methods proposed for the even isotopes [10,11], this method of direct excitation requires only a single probe laser and no multiphoton frequency-mixing schemes, so it can be readily implemented in existing lattice clock experiments. Our method is equally effective for all alkaline-earth-like clock candidates (Yb, Sr, Ca, and Mg).

To excite the strongly forbidden $|1\rangle \rightarrow |2\rangle$ clock transition at frequency ω_{21} as shown in Fig. 1, we apply a static magnetic field $\mathbf{B}$ that couples the states $|2\rangle$ and $|3\rangle$. These states are split by a frequency Δ_{32} and have a coupling matrix element $\Omega_B = \langle 2|\hat{\mu}\cdot\mathbf{B}|3\rangle/\hbar$, where $\hat{\mu}$ is the magnetic-dipole operator. According to first-order perturbation theory with the condition $|\Omega_B/\Delta_{32}| \ll 1$, the state $|2\rangle$ acquires a small admixture of the state $|3\rangle$ due to the presence of the static magnetic field:

$$|2'\rangle = |2\rangle + \frac{\Omega_B}{\Delta_{32}}|3\rangle. \quad (1)$$

The result is that the transition $|1\rangle \rightarrow |2'\rangle$ becomes partially allowed (e.g., for spontaneous emission [12,13]). An optical field with amplitude $\mathbf{E}$ and frequency ω [i.e., $\tilde{\mathbf{E}}(t) = (\mathbf{E}e^{-i\omega t} + \text{c.c.})/2$] acts via the $|1\rangle \rightarrow |3\rangle$ electric-dipole transition (see Fig. 1). We assume that this transition is at least weakly allowed (e.g., an intercombination transition) and has a decay rate γ. The corresponding coupling matrix element is the Rabi frequency, $\Omega_L = \langle 3|\hat{\mathbf{d}}\cdot\mathbf{E}|1\rangle/\hbar$, where $\hat{\mathbf{d}}$ is the electric-dipole operator [14]. Because of the slight admixture of state $|3\rangle$ into the bare state $|2\rangle$, a resonance will be observed on the forbidden transition ($|1\rangle \rightarrow |2\rangle$) when $\omega \approx \omega_{21}$. (We note that static electric fields can be used in an analogous way to enable direct transitions

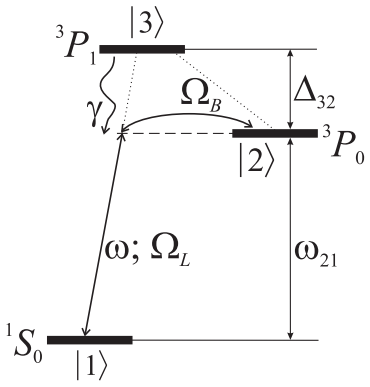

FIG. 1. Magnetic field-induced excitation of a strongly forbidden transition in a generic three-level atomic system. A small magnetic field ($\sim$ mT) mixes excited states $|2\rangle$ and $|3\rangle$, allowing single-photon excitation (for $\omega = \omega_{21}$) of the otherwise forbidden transition between states $|1\rangle$ and $|2\rangle$. This approach can work for a number of interesting alkaline-earth-like elements (Yb, Sr, Ca, and Mg), for which the relevant states are labeled $^{2S+1}L_J$.

between states of like parity [15].) Using Eq. (1), we can write the effective Rabi frequency for excitation of the clock transition,

$$\Omega_{12} = \langle 2'|\hat{\mathbf{d}} \cdot \mathbf{E}|1\rangle/\hbar = \frac{\Omega_L \Omega_B}{\Delta_{32}}. \quad (2)$$

Remarkably, we find that a reasonable Rabi frequency ($\Omega_{12}/2\pi \sim 1$ Hz) results from very modest field values (e.g., $B \sim 2$ mT and light intensity ~ 10 mW/cm^2) for alkaline-earth-like atomic parameters.

Because of the small admixture of $|3\rangle$, state $|2'\rangle$ has a finite lifetime, which leads to broadening of the forbidden transition. Including power broadening due to the laser light, the total broadening can be estimated in the framework of perturbation theory as

$$\gamma_{12} \sim \gamma \frac{\Omega_L^2/4 + \Omega_B^2}{\Delta_{32}^2}. \quad (3)$$

This broadening effectively comes from the fraction of population transferred by the two fields to the $|3\rangle$ state, which decays with a rate γ. This broadening is inversely proportional to the square of the large frequency splitting Δ_{32}, so it typically will be less than 1 μHz. Instead, the spectroscopic linewidth will be determined by other experimental factors such as lattice decoherence effects or intentional power broadening due to the effective Rabi frequency (Ω_{12}).

Of considerable importance to clock applications are the quadratic shifts of the transition frequency $|1\rangle \rightarrow |2\rangle$ that result from the applied fields. For a three-state system, the optical Stark shift is

$$\Delta_L = \frac{\Omega_L^2}{4\Delta_{32}}, \quad (4)$$

while the second-order Zeeman shift is

$$\Delta_B = -\frac{\Omega_B^2}{\Delta_{32}}. \quad (5)$$

The Rabi frequency in Eq. (2) can then be rewritten in terms of the induced field shifts, $\Omega_{12} = 2\sqrt{|\Delta_L \Delta_B|}$. In the example below, we will see that reasonable excitation rates lead to shifts of a few hertz or less, which can be controlled at a much finer level. Moreover, the same induced Rabi frequency can be realized with different combinations of Δ_L and Δ_B, which allows a high degree of experimental flexibility. For example, if the magnetic field is more easily controlled than the optical field, it could be preferable from the metrological viewpoint to work under the condition $\Delta_B > \Delta_L$.

This flexibility is in stark contrast to the case of the odd isotopes. Because of their intrinsic nuclear magnetic moments, these atoms have fixed effective magnetic fields ($\sim$1 T), which are much larger than the applied fields we propose to use ($\sim$1 mT). The nuclear magnetic field mixes a much larger fraction ($\sim$1000 times) of $|3\rangle$ into $|2\rangle$ [see Eq. (1)], producing a natural linewidth of $\sim$15 mHz for the forbidden transition in Yb. As a result, relatively little probe laser intensity (a few μW/cm^2) is required to excite the forbidden transition. In exchange, however, one finds a much larger magnetic sensitivity with a first-order dependence on the B field of several MHz/T. For reasonable experimental parameters, the light shifts are negligible (sub-mHz), but the magnetic shifts are large, requiring field control (or isolation) at the 100 pT (microgauss) level. One of the strengths of our method is that we can tune the size of the magnetic field so that the uncertainties resulting from the induced shifts are of similar size. As a result, the magnetic field shielding requirements are relaxed by nearly a factor of 1000, whereas the light shifts remain at a manageable level (see example below), greatly simplifying the experimental apparatus.

Let us now expand our discussion based on a three-level atom to account for the level structure of a real atom (refer again to Fig. 1), using the example of even isotopes of alkaline-earth-like atoms (e.g., Mg, Ca, Sr, and Yb). Our objective is to excite the forbidden (but tantalizing) $^1S_0 \rightarrow {}^3P_0$ optical clock transition, using the intercombination transition $^1S_0 \rightarrow {}^3P_1$ to supply the required electric-dipole interaction. The magnetic-dipole coupling with the static field $\mathbf{B}$ is implemented via the $^3P_1 \rightarrow {}^3P_0$ transition. We note that the lowest-lying $^1P_1$ state contributes to the induced Rabi frequency in the same way as the $^3P_1$ state, but at a reduced level due to its approximately 10 times larger detuning.

Taking into account the vector nature of the applied fields and the Zeeman degeneracy of the level $^3P_1$, the expression for the Rabi frequency in Eq. (2) takes the specific form

$$\Omega_{12} = \frac{\langle\|d\|\rangle\langle\|\mu\|\rangle(\mathbf{E} \cdot \mathbf{B})}{\hbar^2 \Delta_{32}}. \quad (6)$$

Here $\langle\|d\|\rangle$ is the reduced matrix element of the electric-dipole moment on the $^1S_0 \rightarrow {}^3P_1$ transition, and $\langle\|\mu\|\rangle$ is the reduced matrix element of the magnetic-dipole mo-

ment on the $^3P_1 \rightarrow {}^3P_0$ transition. For even isotopes of the alkaline-earth-like elements, $\langle \|\mu\| \rangle = \sqrt{2/3}\mu_B$, where μ_B is the Bohr magneton. Values for the fine-structure splittings (Δ_{32}) and electric-dipole matrix elements, however, are strongly atom-dependent (see Table I). To evaluate the applicability of this method for various atoms, it is useful to rewrite the expressions for the Rabi frequency (Ω_{12}) and the induced shifts ($\Delta_{L,B}$) in terms of the applied fields. Combining all the constant terms into a single value, α, the expression in Eq. (6) can be rewritten as

$$\Omega_{12} = \alpha\sqrt{I}|\mathbf{B}|\cos\theta, \tag{7}$$

where α is a measure of the induced Rabi frequency per unit of each of the fields, I is the light field intensity, and θ is the angle between linearly polarized $\mathbf{E}$ and $\mathbf{B}$ fields. The quadratic Zeeman shift can be written as $\Delta_B = \beta\mathbf{B}^2$, while the light shift on the transition frequency ω_{21} can be expressed as $\Delta_L = \kappa I$, where β and κ are the respective shift coefficients. To estimate the light shift (i.e., the coefficient κ), it is necessary to take into account the contributions of all states connected by dipole transitions to working levels $^1S_0$ and $^3P_0$. In particular, the main contributions to the shifts on $^1S_0$ and $^3P_0$ are the nearest respective $^1P_1$ and $^3S_1$ states. Using these new expressions for Δ_B and Δ_L, Eq. (7) can be written as

$$\Omega_{12} = \xi\sqrt{|\Delta_L\Delta_B|}\cos\theta, \tag{8}$$

where $\xi \equiv \alpha/\sqrt{\beta\kappa}$. The factor ξ can be considered a dimensionless quality factor for the clock transition in this scheme, since it relates the strength of the excitation to the magnitudes of the induced field shifts. A larger value for ξ implies that for a given spectroscopic linewidth, the induced shifts will be smaller. Note that $\xi = 2$ when only the light shift on the ground state due to level $^3P_1$ is considered, as in a simple three-level case.

Table I summarizes the relevant parameters for four different alkaline-earth-like atoms that have been considered as excellent clock candidates [9,16–18]. Despite the wide range of intercombination transition line strengths (given in Table I as natural decay rate γ) and fine-structure splittings (Δ_{32}), we find similar values for α and ξ. Therefore the scheme proposed here may be applied to any of these atoms. This opens the door to new clock possibilities, especially for Mg and Ca, for which multiphoton schemes designed to use the even isotopes require experimentally inconvenient lasers (e.g., in the visible and IR range not covered by laser diodes). We do emphasize, however, that the level shifts ($\Delta_{L,B}$) will generally be different for the same light intensities and magnetic field magnitudes. Thus, different atoms will generally require different combinations of field values to achieve comparable induced Rabi frequencies (Ω_{12}) while keeping field shifts manageable. The values for κ in Table I have an uncertainty of a few mHz/(mW/cm^2) due to uncertainties in transition rates to higher lying states, leading to corresponding uncertainties in ξ. The values for β were calculated using Eq. (5); corrections due to neglected effects such as diamagnetism are estimated to be a few percent of the stated values.

Using the values for Yb, let us estimate the frequency uncertainty induced by the fields for our method, using realistic field magnitudes and assumptions about their control. A magnetic field $|\mathbf{B}| = 1$ mT (10 G) with uncertainty 10^{-7} T (1 mG) leads to a quadratic Zeeman shift of 6.2 Hz with an uncertainty of 1.2 mHz. With a probe laser intensity $I = 8$ mW/cm^2, we estimate the light shift to be 120 mHz, a value that can be verified experimentally. If we assume that the intensity can be controlled on the level of 1%, then the resulting light shift uncertainty is 1.2 mHz. In this case the combined frequency uncertainty due to these two effects is estimated to be 1.7 mHz, corresponding to a fractional frequency uncertainty of 3×10^{-18} on the forbidden transition frequency $\omega_{12}/2\pi = 518$ THz.

For these values of the fields the induced Rabi frequency given in Eq. (7) with $\mathbf{E} \parallel \mathbf{B}$ is $\Omega_{12}/2\pi \approx 0.5$ Hz, a convenient value for optical atomic clock studies. If, as anticipated, this excitation rate is considerably faster than the dominant loss mechanisms (such as lattice decoherence), the excitation profile will be the standard Rabi spectrum [14]. In that case, maximum excitation (on resonance) occurs for a probe pulse with a duration of 1 s, which would yield a Fourier transform-limited feature with an approximate width of 0.9 Hz. Resolving such a narrow line would require a laser with an extremely narrow linewidth, but not one beyond present state-of-the-art capabilities [19]. Because of its lower sensitivity to field shifts in comparison with proposed multiphoton schemes for the even isotopes [10,11], this method enables a more experimentally accessible excitation rate while keeping the field shifts well below 10^{-17}. If we set our linewidth to the more experimentally challenging value of 10 mHz (as assumed in Refs. [10,11]), our field shift uncertainties contribute less than 1 part in 10^{19}. Of course, other potential shifts need to be considered when determining the total uncertainty for an absolute frequency measurement. For ex-

TABLE I. Atomic species comparison. All quantities are expressed in frequency units.

	γ [kHz]	Δ_{32} [THz]	α [Hz/(T$\sqrt{\text{mW/cm}^2}$)]	β [MHz/T^2]	κ [mHz/(mW/cm^2)]	ξ
Yb	182	21	186	−6.2	15	0.60
Sr	7	5.6	198	−23.3	−18	0.30
Ca	0.4	1.5	154	−83.5	−3.5	0.28
Mg	0.07	0.6	98	−217	−0.5	0.28

ample, a particular concern for a high-precision Yb clock is the shift due to blackbody radiation, which is expected to be approximately -1.25 Hz at room temperature with a temperature dependence of ~ 17 mHz/K [20]. The evaluation of such small shifts will benefit greatly from the high stability that could be achieved with a lattice-based system. With a sample of only 10^4 atoms, a single measurement with the required 1 s interaction time could yield an imprecision approaching 10^{-17}, assuming adequate laser prestabilization.

The necessary interaction time is provided by the nondissipative optical lattice, which holds the atoms against gravity. The lattice wavelength is chosen to yield equal Stark shifts for the upper and lower clock states ($|1\rangle$ and $|2\rangle$ in Fig. 1) [3,5,9]. An optical clock based on odd-isotope Yb atoms confined to such a shift-canceling lattice has been projected by Porsev *et al.* [16] to have a potential fractional frequency uncertainty of 10^{-17}. With the scheme proposed here we could potentially achieve an even lower uncertainty, since the even isotopes have less magnetic field sensitivity and no complications due to the multiplicity of the ground state (e.g., optical pumping and lattice polarization sensitivity). Furthermore, spectroscopy based on a single ground state will simplify the determination of the unperturbed transition frequency since only a single spectroscopic feature will be present (instead of a multipeaked Zeeman spectrum).

A particularly attractive aspect of this approach is that it could be implemented in current odd-isotope lattice clock experiments with minimal change to the apparatus. In contrast to the other schemes for the even isotopes [10,11], our method requires neither additional probe lasers nor associated nonlinear optics to perform requisite sum or difference frequency generation. Instead only a modest magnetic field needs to be generated, which can be done with a pair of current-carrying coils.

In summary, we have shown that strongly forbidden transitions can be accessed via single-photon excitation with the application of a relatively weak static magnetic field. With this method reasonable excitation rates can be achieved without inducing significant Stark shifts, Zeeman shifts, or line broadening. This technique should be useful in experimental fields that require atomic transitions with long coherence times such as atom interferometry, quantum computing, and optical atomic clocks. We have shown how this approach can significantly improve lattice-based optical clocks. This technique should enable new clock possibilities and greatly expedite the development of future time standards. Improved clocks will be critical for a variety of terrestrial and space-borne applications including improved tests of the basic laws of physics and searches for drifts in the fundamental constants [21].

After submission of this work, we used the technique proposed in this Letter to directly excite the clock transition at 578 nm in an even isotope of Yb confined to an optical lattice [22].

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