# Frequency Shifts in an Optical Lattice Clock Due to Magnetic-Dipole and Electric-Quadrupole Transitions

A. V. Taichenachev* and V. I. Yudin*
*Institute of Laser Physics SB RAS, Novosibirsk 630090, Russia*
*Novosibirsk State University, Novosibirsk 630090, Russia*
*Novosibirsk State Technical University, Novosibirsk 630092, Russia*

V. D. Ovsiannikov
*Physics Department, Voronezh State University, Voronezh 394006, Russia*

V. G. Pal'chikov
*Institute of Metrology for Time and Space at National Research Institute for Physical-Technical and Radiotechnical Measurements, Mendeleevo, Moscow Region, 141579 Russia*

C. W. Oates
*National Institute of Standards and Technology, Boulder, Colorado 80305, USA*
(Received 10 March 2008; revised manuscript received 22 August 2008; published 4 November 2008)

We report a hitherto undiscovered frequency shift for forbidden $J = 0 \rightarrow J = 0$ clock transitions excited in atoms confined to an optical lattice. These shifts result from magnetic-dipole and electric-quadrupole transitions, which have a spatial dependence in an optical lattice that differs from that of the stronger electric-dipole transitions. In combination with the residual translational motion of atoms in an optical lattice, this spatial mismatch leads to a frequency shift via differential energy level spacing in the lattice wells for ground state and excited state atoms. We estimate that this effect could lead to fractional frequency shifts as large as $10^{-16}$, which might prevent lattice-based optical clocks from reaching their predicted performance levels. Moreover, these effects could shift the magic wavelength in lattice clocks in three dimensions by as much as 100 MHz, depending on the lattice configuration.

DOI: 10.1103/PhysRevLett.101.193601 PACS numbers: 42.50.Gy, 42.62.Fi, 42.62.Eh

The last few years have been marked by theoretical [1] and experimental [2–6] breakthroughs in the field of fundamental laser frequency metrology that has demonstrated the feasibility of exciting strongly forbidden optical transitions in a large number of neutral atoms that are confined to an optical lattice. Tight confinement of the atoms to the Lamb-Dicke regime has enabled spectroscopy of atomic transitions with Hertz level linewidths [4,6] and raises the prospect of neutral atom-based optical frequency standards with a fractional frequency uncertainty at a level below $10^{-17}$ [3].

Critical to reaching this level of performance is the suppression of the shifts due to the lattice light itself. Indeed, the red-detuned optical lattices used in these experiments rely on these Stark shifts to confine the atoms. These shifts result primarily from electric dipole ($E1$) transitions, have a linear dependence on the lattice intensity, and can be as large as 1 MHz ($10^{-9}$ fractionally). Thus, common-mode rejection of the Stark shifts at the $10^{-8}$ level or below is required. This challenge is accomplished largely by tuning the wavelength of the lattice to a value (the so-called magic wavelength) that produces equal shifts for the ground and excited states of the largely forbidden (i.e., extremely narrow) clock transition [1]. This approach has been extremely effective, and clock performance with a $1.5 \times 10^{-16}$ fractional uncertainty has been demonstrated [3]. However, to reach such levels and below, it is necessary to consider the effects of the lattice light in more detail. Higher order effects due to two photon transitions (with a quadratic dependence on intensity) have been carefully evaluated in several works and have been shown to impose no serious barriers to reaching fractional uncertainties below $10^{-17}$ [5,7,8].

In the present Letter, we evaluate the contributions due to magnetic-dipole ($M1$) and electric-quadrupole ($E2$) transitions and demonstrate a previously unknown frequency shift. Such contributions were first considered in this context in Ref. [1], where it was concluded that they would affect only the value of the magic wavelength, and even then in only a negligible way due to their much weaker line strengths (e.g., $10^{-7}$ times that of $E1$ transitions for Sr atoms [1]). While this is indeed the case for traveling light waves, for the standing waves that are characteristic of an optical lattice, the situation is different. In this case, there is an inhomogeneous spatial distribution of the electric and magnetic fields that modifies the spacing of the energy levels in the potential wells formed by lattice light. This leads to a shift associated with the quantization of atomic translational motion in an optical lattice for a forbidden optical transition $J = 0 \rightarrow J = 0$ (for instance, $^1S_0 \rightarrow {}^3P_0$ in alkaline-earth-like atoms). This shift is proportional to the square root of the lattice field intensity (in

 

the Lamb-Dicke regime), and it does not vanish at the magic wavelength $\lambda_m$, at which the first-order (in intensity) light shift cancels. Estimates show that this shift has considerable significance for lattice-based atomic clocks as we strive for fractional uncertainties at the $10^{-16}$ level and below.

Consider an atom confined to an optical lattice that is produced by a one-dimensional elliptically polarized standing wave (with the frequency $\omega$). The electric field vector has the form

$$\mathbf{E}(\mathbf{r}, t) = E_0 \mathbf{e} \cos(\mathbf{k} \cdot \mathbf{r}) e^{-i\omega t} + \text{c.c.}, \quad (1)$$

where $E_0$ is the scalar amplitude, $\mathbf{k}$ is the wave vector ($k = |\mathbf{k}| = \omega/c$), and $\mathbf{e}$ is the complex unit polarization vector ($\mathbf{e} \cdot \mathbf{e}^*) = 1$. The condition $(\mathbf{e} \cdot \mathbf{k}) = 0$ is satisfied due to the transverse nature of the electromagnetic field.

First we consider the frequency shift of a transition $J_g = 0 \rightarrow J_e = 0$ in a potential produced only by the contributions of $E1$ transitions $J_j = 0 \rightarrow J = 1$ ($j = g, e$). For the standing-wave field (1) the light shift (potential) of a $j$th level is spatially modulated and has the following form (the negative sign results from the red detuning):

$$U_j^E(\mathbf{r}) = -\mathcal{W}_j \cos^2(kz), \qquad \mathcal{W}_j > 0 \qquad (j = g, e). \quad (2)$$

Here, for convenience, we choose the $z$ axis to lie along the wave vector $\mathbf{k}$. The potential amplitude $\mathcal{W}_j$ depends on the frequency $\omega$ and is proportional to the field intensity $I$ at the lattice antinode, which can be written as $I = c|E_0|^2/2\pi$.

Now we quantize the translational motion of atoms. Here we assume the atoms are localized around the field antinodes $kz = l\pi$ ($l = 0, \pm 1, \pm 2 \ldots$) to within much less than the lattice wavelength (the Lamb-Dicke regime). In this case we can describe the atomic motion with a harmonic oscillator approximation around the point $z = 0$. For the condition $|kz| \ll 1$ we can use the approximation $\cos^2(kz) \approx 1 - k^2 z^2$, which allows us to rewrite the shift (2) as a constant plus a harmonic oscillator potential:

$$U_j^E(\mathbf{r}) \approx -\mathcal{W}_j + \frac{M(2\pi\Omega_j)^2 z^2}{2}, \qquad (j = g, e), \quad (3)$$

where $M$ is the atomic mass, and the oscillator frequency $\Omega_j$ for the $j$th level has the form

$$\Omega_j = \frac{1}{2\pi}\sqrt{\frac{2\mathcal{W}_j k^2}{M}}, \qquad (j = g, e). \quad (4)$$

For the potential (3), standard quantum theory for the harmonic oscillator yields energies of the upper and lower levels that contain a vibrational structure:

$$\mathcal{E}_j(n) = \mathcal{E}_j^{(0)} - \mathcal{W}_j + h\Omega_j(n + 1/2), \qquad (j = g, e), \quad (5)$$

where $h = 2\pi\hbar$, $\mathcal{E}_j^{(0)}$ is the energy of the unperturbed $j$th state in a free space, and $n = 0, 1, 2, \ldots$ is the vibrational quantum number (see Fig. 1).

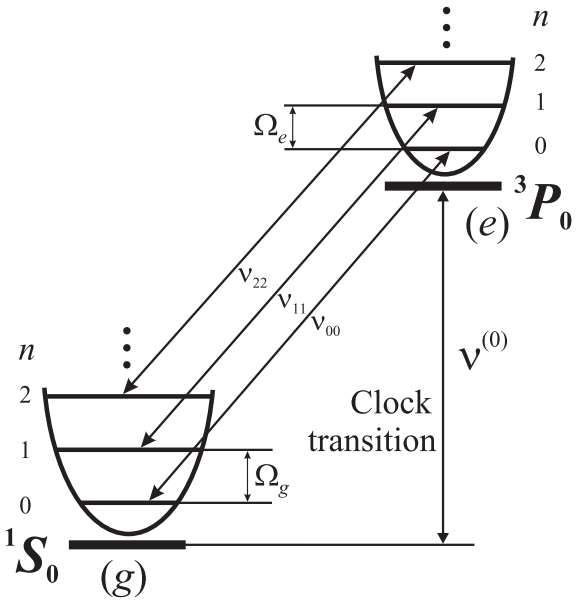


FIG. 1. An illustration of optical transitions between vibrational levels for atoms confined to an optical lattice.

Consider the frequency for an optical transition between vibrational levels with the same quantum numbers $n$ (the usual case for precision spectroscopy):

$$\nu_{nn} = \frac{\mathcal{E}_e(n) - \mathcal{E}_g(n)}{h} = \nu^{(0)} - (\mathcal{W}_e - \mathcal{W}_g)/h + (\Omega_e - \Omega_g)(n + 1/2), \quad (6)$$

where $\nu^{(0)} = (\mathcal{E}_e^{(0)} - \mathcal{E}_g^{(0)})/h$ is the frequency of the unperturbed $J_g = 0 \rightarrow J_e = 0$ transition. Based on the relationships between $I$, $\mathcal{W}$, and $\Omega$, the frequency shift can be written as

$$\Delta\nu_{nn} \equiv \nu_{nn} - \nu^{(0)} = \alpha(\omega) I + (n + 1/2)\beta(\omega)\sqrt{I}, \quad (7)$$

where the coefficients $\alpha(\omega)$ and $\beta(\omega)$ depend upon the given atomic element. They are defined as follows:

$$\alpha(\omega) I = -(\mathcal{W}_e - \mathcal{W}_g)/h, \qquad \beta(\omega)\sqrt{I} = \Omega_e - \Omega_g. \quad (8)$$

Thus, despite the fact that we started with a potential (2) that is proportional to $I$, the effects of the quantization of atomic motion lead to the appearance of an additional square-root dependence ($\propto \sqrt{I}$) for the frequency shift (7). Beyond the Lamb-Dicke regime the intensity dependence of the frequency shift is more complicated due to the anharmonicity of the potential.

For the case of potential (2), which is induced only by $E1$ transitions, $\alpha(\omega_m)$ and $\beta(\omega_m)$ are simultaneously equal to zero at the magic frequency $\lambda_m$, because $\mathcal{W}_e(\omega_m) = \mathcal{W}_g(\omega_m)$, and the shifts cancel in the usual way. However, as we now show, if we take into account contributions due

to $M1$ and $E2$ transitions, we can no longer null $\alpha(\omega_m)$ and $\beta(\omega_m)$ simultaneously, which has important implications for precision metrology.

In accordance with Maxwell's equations for a standing wave, the magnetic field vector $\mathbf{B}$ has a $\sin(\mathbf{k}\cdot\mathbf{r})$ spatial dependence rather than the $\cos(\mathbf{k}\cdot\mathbf{r})$ dependence of the corresponding electric field (1):

$$\mathbf{B}(\mathbf{r},t) = B_0\mathbf{e}_B \sin(\mathbf{k}\cdot\mathbf{r})e^{-i\omega t} + \text{c.c.}, \tag{9}$$

where $B_0 = iE_0$ is the scalar amplitude of the magnetic field, and $\mathbf{e}_B = [\mathbf{k}\times\mathbf{e}]/k$ is the unit polarization vector of the magnetic field. As a result, the contribution to the potential of $j$th level ( $\propto |\mathbf{B}|^2$) due to $M1$ transitions $J_j = 0 \rightarrow J = 1$ has a spatial dependence different from that of the potential (2):

$$U_j^B(\mathbf{r}) = \mathcal{B}_j \sin^2(kz), \qquad (j = g, e), \tag{10}$$

where the potential amplitude $\mathcal{B}_j(\omega)$ is proportional to the intensity $I$, but has a frequency dependence that differs from that of $\mathcal{W}_j$. It can be shown that the contribution due to $E2$ transitions $J_j = 0 \rightarrow J = 2$ also has a $\sin^2(kz)$ spatial dependence (for a 1D standing wave):

$$U_j^Q(\mathbf{r}) = \mathcal{Q}_j \sin^2(kz), \qquad (j = g, e). \tag{11}$$

Hence, for the $j$th level the total potential proportional to the intensity $I$ has the form

$$\begin{aligned} U_j(\mathbf{r}) &= U_j^E(\mathbf{r}) + U_j^B(\mathbf{r}) + U_j^Q(\mathbf{r}) \\ &= -\mathcal{W}_j \cos^2(kz) + \{\mathcal{B}_j + \mathcal{Q}_j\}\sin^2(kz), \qquad (j = g, e). \end{aligned} \tag{12}$$

In contrast to the traveling wave case, where the spatial dependence for the $E1$, $M1$, and $E2$ transitions is the same, here we find that the $M1$ and $E2$ potential wells are spatially shifted relative to the $E1$ potential wells. Thus, for complete cancellation of the Stark shifts, we would need to find a lattice frequency that simultaneously nulls the difference in contributions between the $E1$ ground and excited states (i.e., the usual magic wavelength) and this difference for the sum of the $M1$ and $E2$ contributions. Since such a value is prohibitively unlikely, the concept of the ideal magic frequency is really only valid for a single traveling wave. In this case, however, the confining optical lattice potential is absent and therefore it is not useful for lattice-based atomic clocks. For the standing-wave case, we can estimate the size of this effect by realizing that the contribution due to $E1$ transitions dominates, so the other contributions can be considered as very small perturbations.

Expanding the expression (12) in powers of $(kz)$ and using the harmonic approximation [i.e., $\cos^2(kz) \approx 1 - k^2z^2$ and $\sin^2(kz) \approx k^2z^2$], we obtain an expression for the potential analogous to (3):

$$U_j(\mathbf{r}) \approx -\mathcal{W}_j + \frac{M(2\pi\tilde{\Omega}_j)^2 z^2}{2}, \qquad (j = g, e). \tag{13}$$

However, we now have to use a modified expression for the vibrational frequency $\tilde{\Omega}_j$ that accounts for the $M1$ and $E2$ contributions:

$$\tilde{\Omega}_j = \frac{1}{2\pi}\sqrt{\frac{2\{\mathcal{W}_j + \mathcal{B}_j + \mathcal{Q}_j\}k^2}{M}}, \qquad (j = g, e). \tag{14}$$

From Eqs. (13) and (14) it follows that for a 1D standing wave the $M1$ and $E2$ transitions affect only the coefficient $\beta(\omega)$ in the formula for the shifts (7), while the coefficient $\alpha(\omega)$ is governed solely by the $E1$ transitions as before.

The magic frequency of lattice field $\omega_m$ can still be defined from the condition that nulls the linear shift ($\propto I$) in (7) [i.e., $\alpha(\omega_m) = 0$]. In this case $\mathcal{W}_e(\omega_m) = \mathcal{W}_g(\omega_m) = \mathcal{W}$. However, the remaining part ($\propto\sqrt{I}$) in Eq. (7) now differs from zero:

$$\Delta\nu_{nn} = (n + 1/2)\beta(\omega_m)\sqrt{I} \neq 0. \tag{15}$$

Expanding the expression (14) in the small parameter $|(\mathcal{B}_j + \mathcal{Q}_j)/\mathcal{W}| \ll 1$ and leaving only the first-order term, we obtain

$$\beta(\omega_m)\sqrt{I} = (\tilde{\Omega}_e - \tilde{\Omega}_g) \approx \Omega^{(0)}\xi. \tag{16}$$

Here the frequency $\Omega^{(0)}$ is equal to

$$\Omega^{(0)} = \frac{1}{2\pi}\sqrt{\frac{2\mathcal{W}k^2}{M}}, \tag{17}$$

and its value coincides with the vibrational frequency, which is nearly (but not exactly) the same (at $\omega_m$) for the upper and lower levels of the clock transition $J_g = 0 \rightarrow J_e = 0$. The dimensionless small coefficient $\xi$ in (16) is defined as

$$\xi = \xi_{M1} + \xi_{E2} = \frac{\mathcal{B}_e - \mathcal{B}_g}{2\mathcal{W}} + \frac{\mathcal{Q}_e - \mathcal{Q}_g}{2\mathcal{W}}, \qquad |\xi_{M1}, \xi_{E2}| \ll 1, \tag{18}$$

and it does not depend on the intensity $I$ and polarization $\mathbf{e}$ (for the odd isotopes, which have nonzero nuclear spin, there is a weak polarization dependence that is negligibly small). The terms $\xi_{M1}$ and $\xi_{E2}$ in (18) are governed by $M1$ transitions and $E2$ transitions, respectively.

We now estimate the metrological significance of the square-root-dependent shift (16). Based on general considerations, we expect the coefficient $\xi$ to have a value in the range $10^{-7}$–$10^{-6}$ for elements currently being used in optical lattice clock development. For typical experimental lattice intensities the vibrational frequency $\Omega^{(0)}$ is $\sim$50 kHz. Then the shift $\Delta\nu_{nn}$ could be as large as 50 mHz ($10^{-16}$, fractionally), large enough to be of considerable concern for standards with projected uncertainties below $10^{-17}$. Of even more concern is that this shift appears to be unavoidable and cannot be substantially

reduced by decreasing the field intensity, due to its weak square-root dependence, $\sqrt{I}$. This is in contrast to the case of the hyperpolarizability shift, which is proportional to $I^2$. This shift $\Delta\nu_{nn}$ may also need to be taken into consideration for high precision measurements of the magic frequency $\omega_m$.

The results obtained above can be generalized to the case of an arbitrary field configuration (including 2D and 3D optical lattices), when the electric field vector has the general form

$$\mathbf{E}(\mathbf{r},t)=\mathbf{E}(\mathbf{r})e^{-i\omega t}+\text{c.c.},\quad \mathbf{E}(\mathbf{r})=\sum_a \mathbf{E}_a e^{i(\mathbf{k}_a\mathbf{r})}, \tag{19}$$

where $\mathbf{E}_a$ is the vector amplitude of the $a$th running wave with the wave vector $\mathbf{k}_a$ ($|\mathbf{k}_a|=k=\omega/c$). The spatial dependence of potential induced by $E1$ transitions is governed by the expression

$$U_j^E(\mathbf{r})=w_j(\omega)|\mathbf{E}(\mathbf{r})|^2,\qquad (j=g,e), \tag{20}$$

where the frequency dependence $w_j(\omega)$ depends on the particular atomic element.

The potential induced by $M1$ transitions has the form

$$\begin{aligned} U_j^B(\mathbf{r}) &= b_j(\omega)|\mathbf{B}(\mathbf{r})|^2,\qquad (j=g,e),\\ \mathbf{B}(\mathbf{r}) &= \sum_a e^{i(\mathbf{k}_a\mathbf{r})}[\mathbf{n}_{\mathbf{k}_a}\times\mathbf{E}_a],\quad (\mathbf{n}_{\mathbf{k}_a}=\mathbf{k}_a/k). \end{aligned} \tag{21}$$

The contribution due to $E2$ transitions can be presented in the form of the following scalar product [9]:

$$U_j^Q(\mathbf{r})=r_j(\omega)(\mathbf{Q}_2\cdot\mathbf{Q}_2^*),\qquad (j=g,e), \tag{22}$$

where the covariant components of irreducible tensor of the second rank $\mathbf{Q}_2$ are written as [9]

$$\mathbf{Q}_{2q}=\sum_a e^{i(\mathbf{k}_a\mathbf{r})}\{\mathbf{n}_{\mathbf{k}_a}\otimes\mathbf{E}_a\}_{2q},\quad (q=0,\pm1,\pm2). \tag{23}$$

In these more general (i.e., 2D–3D) cases all the spatial dependencies (21)–(23) differ from one another, so the $M1$ and $E2$ transitions can now also affect the linear term in (7), i.e., the coefficient $\alpha(\omega)$. This will occur, if at the minima points $\{\mathbf{r}_{\min}\}$ of the $E1$ potential $U_j^E(\mathbf{r})$ we have $U_j^B(\mathbf{r}_{\min})\neq 0$ and/or $U_j^Q(\mathbf{r}_{\min})\neq 0$ [in contrast to the case of the ideal 1D standing wave (1)]. Depending on the field configuration, we estimate that the shift in magic frequency could be as large as $|\xi|\omega_m$, or about 100 MHz. Moreover, in a similar way, even in the 1D case, if the counterpropagating waves are unbalanced (e.g., due to imperfect retroreflection), the $M1$ and $E2$ contributions can appreciably modify the value of the coefficient $\alpha(\omega)$ in (7), thereby shifting the magic frequency $\omega_m$.

From an experimental standpoint these results have several important implications. First, measurement of this effect will become important as the clocks are pushed to higher performance levels. However, this may be challenging as normal leveraging techniques (e.g., temporarily increasing the lattice intensity above its usual operational value in order to enhance the size of the shift) will be hampered by the presence of the hyperpolarizability shifts proportional to $I^2$. Second, in 1D experiments, the size of residual traveling waves must be considered when reporting magic wavelengths (the use of optical cavities could be used to suppress the traveling wave [5]). Third, in multidimensional lattices, the magic wavelength will be configuration dependent. Finally, since $\Delta\nu_{nn}$ is proportional to $(n+1/2)$, the frequency shift will increase with increasing temperature of the atoms in the lattice. Thus, it may well be advisable to cool atoms to the lowest vibrational level ($n=0$) (as demonstrated in Ref. [10]) before performing the precision spectroscopy.

In conclusion, we have found that when we take into account the effect of $M1$ and $E2$ transitions on the frequency of a strongly forbidden optical transition, the optical lattice appears not to be as benign as perhaps first thought. When the spatial inhomogeneity of the fields in a 1D or multidimensional lattice is considered, there arises a previously unconsidered frequency shift that results from residual atomic translational motion in the lattice. We find that this shift has a square-root dependence on the lattice intensity for atoms in the Lamb-Dicke regime, and it does not vanish at the magic wavelength. Our order-of-magnitude estimate for the size of this shift suggests that it might be significant for state-of-the-art optical lattice clocks. If so, this effect could have important consequences for the design and operation of future versions of such clocks.

A. V. T. and V. I. Yu. were supported by RFBR (07-02-01230, 07-02-01028, 08-02-01108), INTAS-SBRAS (06-1000013-9427), and Presidium of SB RAS. V. D. O. was supported by RFBR (07-02-00279), CRDF, and MinES RF (ANNEX-BP2M10).

*llf@laser.nsc.ru

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