Optical Lattice Polarization Effects on Hyperpolarizability of Atomic Clock Transitions

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The light-induced frequency shift due to hyperpolarizability (i.e., terms of second-order in intensity) is studied for a forbidden optical transition, $J = 0 \rightarrow J = 0$. A simple universal dependence on the field ellipticity is obtained. This result allows minimization of the second-order light shift with respect to the field polarization for optical lattices operating at a magic wavelength (at which the first-order shift vanishes). We show the possibility for the existence of a magic elliptical polarization, for which the second-order frequency shift vanishes. The optimal polarization of the lattice field can be either linear, circular, or magic elliptical. The obtained results could improve the accuracy of lattice-based atomic clocks.

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In recent years significant attention has been devoted to optical lattice atomic clocks [1,2], in part because the prospects for a fractional frequency uncertainty of such a clock could achieve a level 10^{-17}–10^{-18}. Apart from obvious practical applications, such 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 [3]. The crucial ingredient, for achieving such high metrological performance, is the existence of the magic wavelength λ_m, at which the first-order (in intensity) light shift of the clock transition $^1S_0 \rightarrow {}^3P_0$ cancels for alkaline-earth-like atoms (such as Mg, Ca, Sr, Yb, Zn, Cd). To date, in several experiments cold atoms were trapped in optical lattices at the magic wavelength and the clock transition was observed [2,4–6]. From the metrological viewpoint even isotopes (with zero nuclear spin) are more attractive. To excite strictly forbidden clock transitions in even isotopes the method of magnetic field-induced spectroscopy was proposed [7] and used [5].

Obviously, the achievement of such an extraordinary accuracy in frequency standards is a challenging goal. On the way to this goal it will be necessary to use new approaches and to solve step-by-step the critical physical problems [2]. For example, since at the magic wavelength λ_m the first-order shift vanishes, one of the main factors that limits the accuracy of these optical clocks is the second-order shift due to the atomic hyperpolarizability. Indeed, for the formation of optical lattices with the potential depth of order of MHz [2,4–6] it is necessary to use laser beams with the intensity at a level of a few tens of kW/cm^2. According to our numerical estimates for different alkaline-earth-like atoms [8,9] and first experimental observations for Sr [6], the second-order shift can be at a level of 1–10 Hz in such high-intensity fields. In this case to get planned accuracy we need strictly to control the spatially nonuniform optical lattice fields at a level of 10^{-3}–10^{-5} under conditions of strong focusing, reflections, and interference of light beams. Here apart from long-term stabilization of the laser radiation (power, transverse distribution of intensity) we need precision long-term stability of the whole optical system. A significant reduction of a lattice field intensity is not an effective solution of the problem, because in this case both the number and lifetime of the trapped atoms will be reduced also.

Thus, the hyperpolarizability effect on atoms in optical lattices is an important physical problem, which needs to be carefully studied (the papers [1,6,8] have begun such investigations). In this context the search for alternative methods of minimization of the second-order shifts is especially relevant.

In the present Letter we study the dependence of the second-order shift on the elliptical field polarization for the optically forbidden $J = 0 \rightarrow J = 0$ transition. We show how to minimize this shift with respect to the ellipticity of the lattice field polarization. It turns out that under certain conditions there exists a magic ellipticity at which the second-order light shift vanishes.

Consider an atom in a monochromatic elliptically polarized light field with frequency ω:

$$\mathbf{E}(t) = \mathrm{Re}\{E\mathbf{e}e^{-i\omega t}\}, \tag{1}$$

where E is a scalar field amplitude, $\mathbf{e}$ is a complex unit polarization vector, $(\mathbf{e} \cdot \mathbf{e}^*) = 1$. If the quantization axis

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Oz is orthogonal to the polarization ellipse, we have the following expansions in Cartesian $\{\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z\}$ and spherical $\{\mathbf{e}_0 = \mathbf{e}_z, \mathbf{e}_{\pm 1} = \mp(\mathbf{e}_x \pm i\mathbf{e}_y)/\sqrt{2}\}$ bases:

$$\begin{aligned}\mathbf{e} &= \cos(\varepsilon)\mathbf{e}_x + i\sin(\varepsilon)\mathbf{e}_y \\ &= -\sin(\varepsilon - \pi/4)\mathbf{e}_{-1} - \cos(\varepsilon - \pi/4)\mathbf{e}_{+1}. \end{aligned} \quad (2)$$

Here the ellipticity angle ε can take values $-\pi/4 \le \varepsilon \le \pi/4$. Obviously [see in Fig. 1(a)], $|\tan(\varepsilon)|$ is equal to the ratio of the minor axis to the major axis and the sign of ε determines the helicity. Note that $\varepsilon = 0$ corresponds to a linear polarization, while $\varepsilon = \pm\pi/4$ correspond to circular polarizations. The atom-field interaction will be considered in the dipole approximation $-(\hat{\mathbf{d}}\mathbf{E})$.

Each energy level of the forbidden $J_g = 0 \to J_e = 0$ transition is shifted by amount $\Delta\mathcal{E}_j$ ($j = e, g$) in response to the field (1). These shifts can be expanded in series in even powers of the field amplitude E:

$$\Delta\mathcal{E}_j/\hbar = \alpha_j I + \beta_j I^2 + \ldots \qquad (j = e, g), \quad (3)$$

where $I = c|E|^2/8\pi$. The first term ($\propto I$) describes a first-order Stark shift, which for levels with $J = 0$ does not depend on the field polarization $\mathbf{e}$; i.e., the polarizabilities α_j are completely determined by the field frequency ω alone. The second term ($\propto I^2$) in (3) describes energy level shifts due to the hyperpolarizability (the general expression of the hyperpolarizability for levels with arbitrary angular momentum is presented in [10]). Coefficients β_j depend both on the frequency ω and on the ellipticity ε [11]. From Eq. (3) it follows that the frequency of forbidden transition ω_0 is shifted by the external field (1) by an amount

$$\begin{aligned}&\Delta\omega_0 \equiv (\Delta\mathcal{E}_e - \Delta\mathcal{E}_g)/\hbar = \tilde{\alpha}(\omega)I + \tilde{\beta}(\omega, \varepsilon)I^2 + \ldots \\ &\tilde{\alpha}(\omega) = \alpha_e - \alpha_g, \qquad \tilde{\beta}(\omega, \varepsilon) = \beta_e - \beta_g, \qquad \ldots.\end{aligned} \quad (4)$$

At the magic frequency $\omega_m = 2\pi c/\lambda_m$ the first-order Stark shift vanishes, i.e., $\tilde{\alpha}(\omega_m) = 0$.

The dependence of the coefficient $\tilde{\beta}(\omega, \varepsilon)$ on ellipticity can be used to minimize the influence of the second term

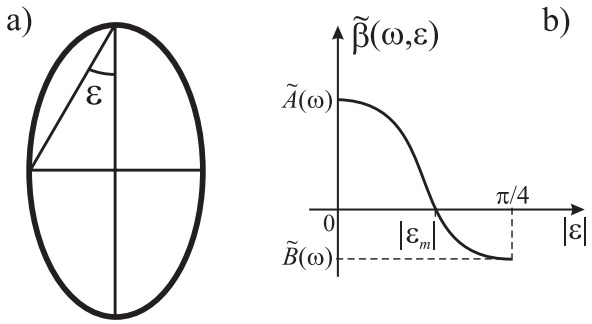

FIG. 1. (a) Definition of the elliptical polarization parameter ε [see Eq. (2)]. (b) Illustration of the existence of a magic elliptical polarization ε_m [see Eq. (11)], when the coefficients $\tilde{A}(\omega)$ and $\tilde{B}(\omega)$ have opposite signs.

($\propto I^2$). According to electric dipole selection rules, the second-order shifts for levels with $J = 0$ originate from all possible transitions $J = 0 \to J' = 1 \to J'' = 0, 1, 2$ (quadratic on the field amplitude with frequencies $0, \pm 2\omega$), i.e., via virtual levels with $J' = 1$. It can be rigorously proven that each of the three generalized channels gives only two contributions with different polarization dependencies in β_j:

$$\beta_j = \sum_{J''=0,1,2} [R^{(j)}_{J''}(\omega)\mathcal{P}_{J''}(\mathbf{e}) + S^{(j)}_{J''}(\omega)\mathcal{Q}_{J''}(\mathbf{e})], \quad (5)$$

where $R^{(j)}_{J''}(\omega)$ and $S^{(j)}_{J''}(\omega)$ depend on the frequency only. All polarization dependencies are contained in the factors

$$\begin{aligned}\mathcal{P}_{J''}(\mathbf{e}) &= (\{\mathbf{e}^* \otimes \mathbf{e}^*\}_{J''} \cdot \{\mathbf{e} \otimes \mathbf{e}\}_{J''}), \\ \mathcal{Q}_{J''}(\mathbf{e}) &= (\{\mathbf{e} \otimes \mathbf{e}^*\}_{J''} \cdot \{\mathbf{e}^* \otimes \mathbf{e}\}_{J''}),\end{aligned} \quad (6)$$

which are the scalar products of tensors composed of the unit polarization vectors $\mathbf{e}$ and $\mathbf{e}^*$ and can be presented explicitly in terms of the scalar $(\mathbf{e} \cdot \mathbf{e})$ and vector $[\mathbf{e} \times \mathbf{e}^*]$ products as [12]:

$$\begin{aligned}\mathcal{P}_0(\mathbf{e}) &= |(\mathbf{e} \cdot \mathbf{e})|^2/3; \qquad \mathcal{Q}_0(\mathbf{e}) = 1/3; \\ \mathcal{P}_1(\mathbf{e}) &= |[\mathbf{e} \times \mathbf{e}]|^2/2 \equiv 0; \qquad \mathcal{Q}_1(\mathbf{e}) = |[\mathbf{e} \times \mathbf{e}^*]|^2/2; \\ \mathcal{P}_2(\mathbf{e}) &= 1 - |(\mathbf{e} \cdot \mathbf{e})|^2/3; \qquad \mathcal{Q}_2(\mathbf{e}) = 1/6 + |(\mathbf{e} \cdot \mathbf{e})|^2/2.\end{aligned} \quad (7)$$

Using expansion (2), we find:

$$|(\mathbf{e} \cdot \mathbf{e})|^2 = \cos^2(2\varepsilon), \qquad |[\mathbf{e} \times \mathbf{e}^*]|^2 = \sin^2(2\varepsilon). \quad (8)$$

Equations (5), (7), and (8) allow us to present the polarization dependence of the coefficients β_j in Eq. (3) as $\beta_j = A_j(\omega)\cos^2(2\varepsilon) + B_j(\omega)\sin^2(2\varepsilon)$. As a result, we can write the universal expression for the polarization dependence of the second-order light shift in the following simple form:

$$\tilde{\beta}(\omega, \varepsilon) = \tilde{A}(\omega)\cos^2(2\varepsilon) + \tilde{B}(\omega)\sin^2(2\varepsilon). \quad (9)$$

Thus, to reconstruct a complete polarization dependence of the coefficient $\tilde{\beta}(\omega, \varepsilon)$, we need to know (by calculation or experiment) its value only in two points, for example, for linear ($\varepsilon = 0$) and circular ($\varepsilon = \pm\pi/4$) polarizations:

$$\tilde{A}(\omega) = \tilde{\beta}(\omega, 0), \qquad \tilde{B}(\omega) = \tilde{\beta}(\omega, \pm\pi/4). \quad (10)$$

It is worth noting, that the terms $R_{J''}(\omega)\mathcal{P}_{J''}(\mathbf{e})$ in Eq. (5) may demonstrate, in particular, the contributions of two-photon resonances to states $J'' = 0, 2$. However, the scalar product $\mathcal{P}_0(\mathbf{e})$ will vanish for circular polarization, so the two-photon resonances to an excited state $J'' = 0$ will appear only for the noncircular polarization, $\varepsilon \ne \pm\pi/4$ [see Eqs. (7) and (8)]. Two-photon resonances to $J'' = 1$ states do not appear, as the scalar product $\mathcal{P}_1(\mathbf{e}) \equiv 0$ [see Eq. (7)]. The terms $S_{J''}(\omega)\mathcal{Q}_{J''}(\mathbf{e})$ may have large "resonance" values when the fine-structure splitting (for ex-

ample, between the metastable $^3P_0$ and $^3P_{1,2}$ sublevels of the ^3P_J triplet) is small. The matrix elements with $J'' = 1$ contribute only to the term $S_1(\omega)\mathcal{Q}_1(\mathbf{e})$ of Eq. (5), which disappears for linear polarization ($\varepsilon = 0$), since $\mathcal{Q}_1(\mathbf{e}) = 0$ for $\mathbf{e} = \mathbf{e}^*$. Thus, in the vicinity of resonance one can anticipate a strong dependence of the hyperpolarizability both on the frequency and polarization of the field.

Equation (9) allows us to optimize the light shift (4) with respect to the ellipticity parameter ε. This optimization consists of determining the optimal ellipticity ε_{opt}, which minimizes the absolute value $|\tilde{\beta}(\omega, \varepsilon)|$, i.e., $|\tilde{\beta}(\omega, \varepsilon_{\text{opt}})| = \min\{|\tilde{\beta}(\omega, \varepsilon)|\}$. Such a minimization is very important for optical frequency standards based on optical lattices at magic frequency, where the first-order Stark shifts cancel out [$\tilde{\alpha}(\omega_m) = 0$] and the higher-order shift $\propto I^2$ becomes one of the main factors limiting the accuracy of the future optical frequency standards.

As is seen from (9), if the coefficients $\tilde{A}(\omega)$ and $\tilde{B}(\omega)$ have the same sign, then the optimal polarization is either linear ($\varepsilon_{\text{opt}} = 0$) or circular ($\varepsilon_{\text{opt}} = \pm\pi/4$). Apart from this, (9) allows for a very intriguing possibility, when the coefficients $\tilde{A}(\omega)$ and $\tilde{B}(\omega)$ have opposite signs. In this case a magic elliptical polarization ε_m always exists [see in Fig. 1(b)], for which the second-order light shift vanishes:

$$\tilde{\beta}(\omega, \varepsilon_m) = 0 \Rightarrow \tan(2\varepsilon_m) = \pm\sqrt{-\tilde{A}(\omega)/\tilde{B}(\omega)} \quad (11)$$

and, consequently, $\varepsilon_{\text{opt}} = \varepsilon_m$. Obviously, the most interesting case arises for the magic ellipticity at the magic frequency ω_m, i.e., when $\tilde{\beta}(\omega_m, \varepsilon_m) = 0$. One of possible candidates for such a remarkable coincidence is Yb.

According to the experimental results [5], the magic wavelength λ_m for the forbidden transition $(6s^2)^1S_0 \rightarrow (6s6p)^3P_0$ in Yb equals approximately to 759.35 nm. Comparing this value with the energy spectrum, one finds that this wavelength nearly meets the two-photon resonance conditions that occur at 759.71 nm [$(6s6p)^3P_0 \rightarrow (6s8p)^3P_0$ resonance] and 754.23 nm [$(6s6p)^3P_0 \rightarrow (6s8p)^3P_2$ resonance]. Therefore, the main contribution to the second-order shift $\propto I^2$ is due to the shifts of metastable level $J_e = 0$ [i.e., $(6s6p)^3P_0$], originating from interactions via levels indicated in Fig. 2 (i.e., resonant contributions). Thus,

$$\tilde{\beta}(\omega, \varepsilon) \approx \beta_e^{(1)} + \beta_e^{(2)} + \beta_e^{(3)} + \beta_e^{(4)}, \quad (12)$$

where terms $\beta_e^{(1,2)}$ are related to the resonance two-photon transitions $(6s6p)^3P_0 \rightarrow (6s8p)^3P_{0,2}$ at the doubled magic frequency $2\omega_m$, and $\beta_e^{(3,4)}$ originate from the interaction of the level $(6s6p)^3P_0$ with the other levels $(6s6p)^3P_{1,2}$ of the same fine-structure manifold. The polarization dependencies for $\beta_e^{(1,2)}$ are determined by $\mathcal{P}_{0,2}(\mathbf{e})$, whereas for $\beta_e^{(3,4)}$ are determined by $\mathcal{Q}_{1,2}(\mathbf{e})$. Note that the resonance two-photon transition $J = 0 \rightarrow J = 1$ is forbidden in the dipole approximation [13] [see also $\mathcal{P}_1(\mathbf{e}) \equiv 0$ in Eq. (7)]; therefore, in (12) the contribution from the transition $(6s6p)^3P_0 \rightarrow (6s8p)^3P_1$ (see Fig. 2) is neglected.

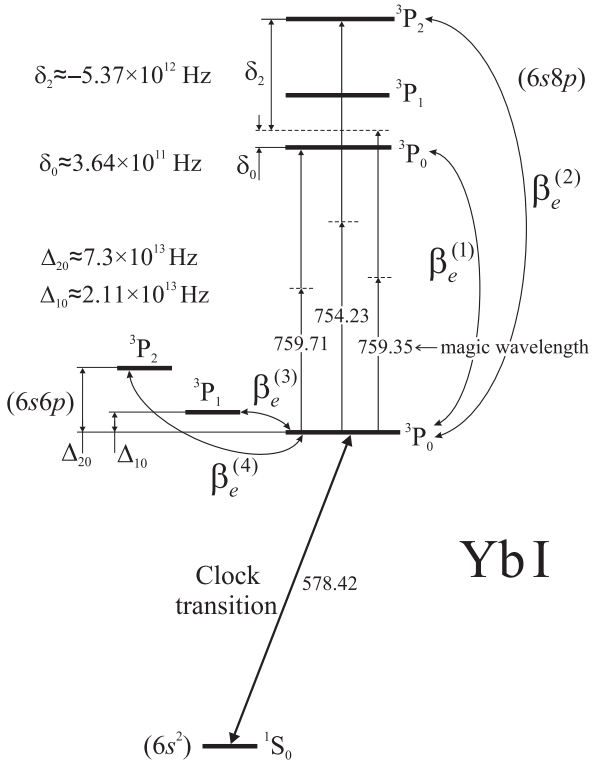

FIG. 2. Yb energy levels responsible for the main contributions $\beta_e^{(1,2,3,4)}$ to the second-order shift of the forbidden transition $(6s^2)^1S_0 \rightarrow (6s6p)^3P_0$ (all wavelengths are given in nm).

Using (7) and (8), the terms in (12) can be written as:

$$\beta_e^{(1)}(\omega, \varepsilon) = \frac{b_1(\omega)}{\delta_0}\frac{\cos^2(2\varepsilon)}{3} \geq 0 \quad (\delta_0 > 0) \quad (13)$$

$$\beta_e^{(2)}(\omega, \varepsilon) = \frac{b_2(\omega)}{\delta_2}\frac{3 - \cos^2(2\varepsilon)}{3} < 0 \quad (\delta_2 < 0) \quad (14)$$

$$\beta_e^{(3)}(\omega, \varepsilon) = -\frac{b_3(\omega)}{\Delta_{10}}\frac{\sin^2(2\varepsilon)}{2} \leq 0 \quad (15)$$

$$\beta_e^{(4)}(\omega, \varepsilon) = -\frac{b_4(\omega)}{\Delta_{20}}\frac{1 + 3\cos^2(2\varepsilon)}{6} < 0, \quad (16)$$

where the coefficients $b_{1,2,3,4}$ are assumed to be positive, δ_0 and δ_2 are the two-photon detunings from the transitions $(6s6p)^3P_0 \rightarrow (6s8p)^3P_{0,2}$, and Δ_{10} and Δ_{20} are the fine-structure splittings of the $(6s6p)^3P_{0,1,2}$ state (see Fig. 2). From Eq. (13), the term $\beta_e^{(1)}$ is positive (because $\delta_0 > 0$), while all the other terms $\beta_e^{(2,3,4)}$ are negative.

As it follows from (13) and (15), in the case of linear polarization ($\varepsilon = 0$) the term $\beta_e^{(1)}$ becomes maximal, and $\beta_e^{(3)} = 0$. Because of the strong resonance conditions $|\delta_2/\delta_0| \approx 15$ and $\Delta_{20}/\delta_0 \approx 200$, we expect that the term

$\beta_e^{(1)}$ will dominate over $\beta_e^{(2)}$ and $\beta_e^{(4)}$. This directly leads to a positive value for $\tilde{\beta}(\omega_m, 0)$, i.e., $\tilde{A}(\omega_m) > 0$ in accordance with (10). For circular polarization ($\varepsilon = \pm\pi/4$) we have $\beta_e^{(1)} = 0$ and $\beta_e^{(2,3,4)} < 0$, which leads to a negative $\tilde{\beta}(\omega_m, \pm\pi/4)$, i.e., $\tilde{B}(\omega_m) < 0$ according to (10). Thus, the coefficients $\tilde{A}(\omega_m)$ and $\tilde{B}(\omega_m)$ may have opposite signs, thus providing a sufficient condition for the existence of a magic elliptical polarization ε_m (11) at the magic frequency ω_m for Yb.

It should be stressed that the qualitative analysis above does not guarantee the existence of a magic ellipticity for Yb, because we did not take into account contributions to hyperpolarizability from the inner-shell electrons, nor numerous off-resonant contributions in $\tilde{\beta}(\omega_m, \varepsilon)$ of the jumping electron. So, while there is a good chance of a magic ellipticity for Yb, the ultimate answer will be given by an experiment. Nevertheless, the analysis shows that the presence of the near-resonance two-photon transitions can lead to an intriguing situation. In this context, it is worth noting that for Sr atoms there also is a near-resonant two-photon transition $(5s5p)^3P_0 \rightarrow (5s4f)^3F_2$ [6]. In [6] the second-order shifts have been investigated only for a linearly polarized field. From the experimental results it follows that $\tilde{A}(\omega_m) > 0$. The value and sign of second-order shift in circularly polarized field (i.e., the coefficient $\tilde{B}(\omega_m)$) are still unknown. Moreover, for circular polarization the negative contribution due to the interaction with the level $(5s5p)^3P_1$ (analogue of the term $\beta_e^{(3)}$ in Fig. 2) becomes maximal [see Eq. (15)], while for linear polarization it equals to zero. Consequently, the question of the optimal polarization remains open and the possibility of a magic ellipticity for Sr still takes play.

Concluding, for forbidden optical transition $J = 0 \rightarrow J = 0$ (for example, $^1S_0 \rightarrow {}^3P_0$ clock transition in even isotopes of alkaline-earth-like atoms) we have investigated the polarization dependence of the higher-order frequency shifts $\propto I^2$, originating from the atomic hyperpolarizability. This dependence has a simple universal form (9) and we have described the method for minimizing the second-order shift for optical lattices at the magic frequency ω_m. To this end, the higher-order shifts for linear and circular field polarization should be measured and compared. If these shifts are the same sign, then the optimal polarization (either circular or linear) will correspond to minimal absolute value of the shift. If the signs of the measured shifts are different, then a magic ellipticity ε_m will exist, where the second-order shift vanishes. The magic ellipticity can be estimated from (11), and determined more accurately from experiments.

It should be stressed that the existence of a magic ellipticity allows a practically ideal one-dimensional standing wave optical lattices for the frequency standards, because in this case it is not necessary to control strictly the lattice field intensity. Consequently, one can use high-intensity fields to create deep potential lattices with high efficiency of trapping and with longer capture time. Note also that in deep potential lattices cold atoms are localized on length scales much less than the field wavelength, i.e., the strong Lamb-Dicke regime is realized.

These results can be extended to 2D and 3D lattices in the field with spatially nonuniform polarization [14]. Here the lattice field configuration should be chosen in such a way that at the potential energy minimum the local field polarization coincides with the optimal value (either linear, circular, or magic) for the given element. In this case for lower vibrational levels the second-order shifts will be minimal, assuming the Lamb-Dicke regime.

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