Field Compensation of Field Frequency Shifts in the Ramsey Spectroscopy of Clock Optical Transitions

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We develop the method of Ramsey spectroscopy with the use of an additional field compensating the frequency shifts of clock optical transitions. This method in combination with the method of magnetically induced spectroscopy [11] allows the practically complete compensation (down to the fractional level below $10^{-17}$ with respect to the unperturbed clock frequency) of field shifts of various origin for even isotopes of alkaline-earth-like atoms confined to an optical lattice at magic wavelength (lattice-based atomic clocks). Apart from this, the new method can be used in the two-photon Doppler-free Ramsey spectroscopy of other forbidden optical transitions (for example, $1S_0 \rightarrow 1S_0$ and $1S_0 \rightarrow D_2$) and other atoms. This opens perspectives for development of principally new variants of primary optical frequency standards based on free atoms. As a whole, the method of the shift-compensating field is distinguished by simplicity and high efficiency, and it can be applied for any transitions in any atoms and ions.

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The last few years were marked by great achievements in high-resolution spectroscopy and fundamental laser metrology. First of all we should mention optical frequency standards on a single ion in an rf (Paul) trap [1, 2], and atomic clocks based on a large number of neutral atoms confined to an optical lattice on a magic wavelength (so called lattice-based atomic clocks) [3, 4]. These titanic efforts are directed to the creation of optical frequency standards with presently unattainable frequency uncertainty and accuracy at the level $10^{-17}$-$10^{-18}$. While the progress in ion standards is a natural consequence of longstanding (more than 20 years) careful work, the experimental activity on lattice-based clocks started only two-three years ago, when simultaneously in several laboratories super-narrow resonances have been obtained on the strongly forbidden optical transition $1S_0 \rightarrow 3P_0$ of Sr [5, 6, 7, 8] and Yb [9]. Even for so short period the very impressive results were achieved [10], when the metrological characteristics of the lattice-based clock is already better than for cesium fountain primary standards. As a whole, this new direction (lattice-based atomic clocks) is now on a stage of rapid development, which stimulates a generation of interesting physical ideas, including a new spectroscope (with respect to the field amplitude) Stark shifts of the levels of clock transition are equal to each other. This leads to the practically complete compensation of the frequency shift for the clock transition. However, there are additional frequency shifts caused by lattice field, which, despite of their relative smallness, can have a principal significance for the definition of metrological characteristics of atomic clocks. First of all, it is the field shift caused by the hyperpolarizability, which has been investigated experimentally in [7, 14] and theoretically in [13]. Besides, recently we have discovered the other shift originating from the quantization of translational motion of atoms in a lattice with account for contributions due to the magneto-dipole and quadrupole transitions [10].

However, apart from these frequency shifts caused by the lattice field, there exist shifts due to the presence of additional fields. For example, in the case of odd isotopes of alkaline-earth-like atoms a static magnetic field is used to spectrally split the Zeeman structure of levels originating from nonzero nuclear spin. Here both the linear and quadratic Zeeman shifts of the clock transition frequency take place. In the case of even isotopes (zero nuclear spin) a magnetic field is used in the method of magnetically induced spectroscopy [11], leading to the quadratic shift only. Besides, for even isotopes the AC Stark shift caused by the clock probe laser has an essential (from metrological viewpoint) significance (see numerical estimates in [11]). Namely, the possibility to eliminate such shifts (i.e., shifts caused by the additional fields) is one of the main subjects of the present paper.

Thus, as a concrete example we will consider the method of magnetically induced spectroscopy [11] of
strongly forbidden transitions $^1S_0 \rightarrow ^3P_0$ in even isotopes of alkaline-earth-like atoms (see in Fig. 1) confined to an optical lattice on the magic wavelength. For the sake of definiteness we will consider the frequency of clock transition $\omega_0$ (i.e. in the presence only of lattice field, including the BBR shift and collisional shift) as unperturbed. Hence we have the following frequency shift (see in [11]):

$$\Delta = \kappa I_p + \beta |B|^2,$$

induced by the probe laser field (with the intensity $I_p$) and by the static magnetic field $B$. Under the creation of frequency standards (especially of primary standards) an evident question arises: what to do with these shifts? We see two variants of answer to this question. The first variant consists in the precision experimental measurement of the coefficients $\kappa$ and $\beta$, and the high-degree control of the values $I_p$ and $|B|$. Then, using results of spectroscopic measurements, one can calculate the frequency of unperturbed transition $\omega_0$ at $I_p=0$ and $B=0$. For example, if we use the magnetic field of order of $|B| \sim 10^{-3}$ T ($\sim 10$ G), then the quadratic shift $\beta |B|^2$ is of order of 10 Hz. In this case in order to have a possibility to achieve the absolute accuracy at the level of 1 mHz and lower (the fractional error less than $10^{-17}$) it is necessary to know the coefficient $\beta$ and to control the value of magnetic field $|B|$ with the accuracy $10^{-4}$ (for the magnetic field it means value of order 0.1 $\mu$T). Obviously, the achievement of such a high accuracy of measurement of the coefficient $\beta$ will require a persistent efforts during many years. Probably we can not expect the same accuracy (i.e. of the order of 0.01%) of theoretical calculations for many-electron atoms due to the diamagnetic contribution. Similar estimations can be made for the shift caused by the probe laser field ($\kappa I_p$).

The other possible variant of solution of the problem of the field shift ([11]) is connected with the use of the Ramsey spectroscopy with separated (in time) pulses (see in Fig. 2). As applied to the method of magnetic field-induced spectroscopy, it is assumed here that during the pulse (with the Rabi frequency $V$ and duration $\tau$) atoms are driven by the probe laser field with the frequency $\omega$ in the presence of static magnetic field $B$, and during the free evolution (of the duration $T$) both the magnetic and probe fields are simultaneously turned off. Thus, the given version of the Ramsey spectroscopy is distinguished by the feature, consisting in the appearance of the frequency shift $\Delta$ only during the pulse action. If in the initial moment $t=0$ atoms were in the lower level ($^1S_0$), then after the action of two pulses the population of atoms in the excited state ($^3P_0$) is determined by the following expression:

$$n_e = \frac{V^2}{\Omega^2} \times \left[ \cos \left( \frac{T\delta}{2} \right) \sin(2\Omega\tau) - \frac{\delta - \Delta}{\Omega} \sin \left( \frac{T\delta}{2} \right) \sin^2(\Omega\tau) \right]^2,$$

where $\delta=(\omega-\omega_0)$ is the detuning of the probe field frequency from the frequency of unperturbed transition (i.e. during free evolution between Ramsey pulses), and $\Omega=\sqrt{V^2+(\delta-\Delta)^2/4}$ is the generalized Rabi frequency.

The formula (2) describes typical Ramsey fringes, where the central resonance (as a function of $\delta$) is a reference point for our purposes. The presence of additional frequency shift $\Delta$ during the pulse action leads to the shift of position of the central resonance $\delta \omega_0$ with respect to the frequency of unperturbed transition $\omega_0$. If $|\Delta/V| \ll 1$, the amplitude of the central resonance is maximal ($\approx 1$) for $\tau V = (2l+1)\pi/4$ (where $l=0,1,2,...$). For $l=0$ the shift of the central resonance top $\delta \omega_0$ (in units of $s^{-1}$) with a good accuracy can be written as:

$$\delta \omega_0 \approx \xi \frac{1}{T} \frac{\Delta}{V}, \quad (3)$$

where for $1 \leq (VT) < \infty$ the dimensionless coefficient $\xi$ monotonically increases in the interval $0.5 \leq \xi < 1$.

For Yb atoms, as it follows from the calculations in [11], the coefficients $\kappa$ and $\beta$ have opposite signs, that al-
In this case, even with account for possible variations of \( \omega \) with the frequency \( \nu \), the transition connected with one of the working levels can be compensated by the frequency shift (1). Here any suitable compensating field is syn-chronized with the Ramsey pulse (see in Fig.2) of the probe field (with the clock transition frequency \( \omega_c \)). In this case instead of the expression (1) for the parameters \( \Delta \) in eq. (2) we have to use the other:

\[
\Delta' = \kappa I_p + \beta |B|^2 + \eta(\omega_{sh}) I_{sh},
\]

where \( I_{sh} \) is the intensity of the compensating field, and the coefficient \( \eta(\omega_{sh}) \) governs the dependence of additional shift on the frequency \( \omega_{sh} \). In particular, near the transition \( ^1S_0 \rightarrow ^3P \) the following relationship takes place:

\[
\eta(\omega_{sh}) \approx (\omega_{sh} - \omega_1)^{-1},
\]

where \( \omega_1 \) is the frequency of the transition \( ^1S_0 \rightarrow ^3P \) (see in Fig.1). The key idea of our method is that the condition \( \Delta' \approx 0 \) can be satisfied by the proper choice of the frequency \( \omega_{sh} \) and intensity \( I_{sh} \). In this case, even with account for possible variations of the values \( I_p, B, \omega_{sh} \), and \( I_{sh} \), the regime \( |\Delta'/V| \ll 1 \) is realized, leading to the high efficiency of the Ramsey spectroscopy.

Let us describe the procedure of spectroscopic measurements. Initially, we fix the values \( I_p \) and \( B \), which determine the Rabi frequency \( \nu \propto |B| \sqrt{T_p} \) (see in [11]). Then, we fix the compensating field frequency \( \omega_{sh} \), for which there certainly exists a point \( I_{sh}^{(0)} \) (under scanning of the intensity \( I_{sh} \)), where the condition \( \Delta' \approx 0 \) is satisfied. After that measurements of the resonance position \( \delta \omega_0 \) versus the intensity \( I_{sh} \) are made for several values of the free evolution time \( T \) and/or pulse duration \( \tau \). The point \( I_{sh}^{(0)} \) is determined as a crossing point of these curves (see in Fig.3). And, finally, fixing the intensity \( I_{sh}=I_{sh}^{(0)} \) and optimal pulse duration \( (\tau V = \pi/4) \), the probe filed frequency is stabilized on the unperturbed frequency \( \omega_0 \). Another way is also possible – vice versa, to fix the intensity \( I_{sh} \) and to scan the frequency \( \omega_{sh} \) at several different values \( T \) and/or \( \tau \). The crossing of these curves determines the frequency \( \omega_{sh}^{(0)} \) for which \( \Delta' = 0 \).

The radical advantage of our method in comparison with the usual method of single-pulse spectroscopy (currently adopted for the lattice-based clocks) consists in that at the output we get immediately the probe filed with the frequency \( \omega_0 \). Moreover, we do not need in the precision measurements (or high-accuracy theoretical calculations) of the coefficients \( \kappa \) and \( \beta \) in [11]. Thus, the main inaccuracy of the frequency measurements will be determined only by the collisional shift, BBR shift, and shift due to the lattice field. All these prove that the proposed method has essentially better perspectives as related to the measurement of the unperturbed frequency of clock transition. Note also that the possibility of practical realization of the method of shift-compensating field in the Ramsey spectroscopy does not rise any doubts in view of the experimental realization of the method of magnetic field-induced spectroscopy for different atoms [8, 12].

Taking Sr as an example, let us estimate the fractional frequency uncertainty of our method, using the coefficients calculated in [11]. Let \( I_p = 100 \, \text{mW/cm}^2 \) and \( |B|=0.3 \, \text{mT} \), that leads to the values \( V \approx 0.3 \, \text{Hz} \) and \( \tau \approx 0.42 \, s \) \((\tau V = \pi/4)\). The corresponding shift \( \Delta \approx 3.9 \, \text{Hz} \) should be compensated by the shift-compensating field \( I_{sh} \). We will assume possible fluctuations of the intensities \( I_p \) and \( I_{sh} \) at level of \(<0.5\%\), and the frequency stabilization at level of \(<0.5 \, \text{MHz} \) for the detuning \( (\omega_{sh} - \omega_1) > 100 \, \text{MHz} \). In this case for \( T=1.5 \, s \) \((VT \approx 2.8)\) from the formula (3) we find the estimate of absolute frequency fluctuations \( \delta \omega_0 \approx 5 \, \text{mHz} \), what corresponds to the fractional frequency uncertainty \( <10^{-17} \).

The magnetic field control in our method can be substantially (in one-two order) relaxed in comparison with the method of single-pulse magnetically induced spectroscopy [11]. For example, if in [11] in order to achieve the fractional frequency uncertainty \( 10^{-17} \) for Yb it is necessary to control the magnetic field of 1 mT at level of \( \approx 0.1 \, \mu\text{T} \), then in the Ramsey method for Yb atoms it is sufficient to control the magnetic field of 1 mT (during the pulse action) at level of 5-10 \( \mu\text{T} \) (including the residual field).
Let us compare now our method with the other version of the Ramsey spectroscopy proposed in [13] for even isotopes of alkaline-earth atoms in lattice-based clocks. As related to the fractional frequency uncertainty, the both methods are approximately equivalent to each other. However, in our method we always have a possibility to achieve the maximal amplitude (100% contrast) of the central Ramsey resonance. Therefore, as a whole, the calculated metrological characteristics of our method are better than for the method in [13]. As related to technical side of the problem, then in both methods formally two lasers are used (in our method they are the probe and shifting fields). However, in our methods they are completely independent laser sources, while in the method of [13] the two lasers (with substantially different frequencies) require the strict phase locking to each other to record the narrow resonance of the \( \Lambda \) type. Thus, the experimental realization of our method is significantly easier than in [13] (especially in the case of Yb atoms, for which, as it was mentioned above, it is not necessary to use the additional shifting field). It should be also stressed that our method is ideologically transparent and simple for theoretical analysis.

Apart from this, the proposed by us method of additional shift-compensating field can be generalized to the case of classical two-photon spectroscopy (in atoms and ions) [17], when the probe field frequency equals to the half of the clock transition frequency. For example, the dipole-forbidden transitions \( ^1S_0 \rightarrow ^1S_0 \) or \( ^1S_0 \rightarrow ^1D_2 \) can be used, as is shown in Fig.1. Here it is assumed that the higher levels \( ^1S_0 \) and \( ^1D_2 \) are long-lived (as, for example, \( ^1S_0 \rightarrow ^1S_0 \) for He, and \( ^1S_0 \rightarrow ^1D_2 \) for Ca, Ba). It is possible to work with atoms confined to an optical lattice at the magic wave length (for the corresponding clock transition) as well as with free atoms. Here for free atoms it is necessary to use a field of two counterpropagating plane waves with opposite circular polarizations (so called \( \sigma_+ - \sigma_- \) configuration), which allows to avoid the recoil effect and linear Doppler shift for moving atoms [18, 19]. Moreover, if we will use such a field for the two-photon spectroscopy of atoms confined to an 1D optical lattice, then its direction (the line of wavevectors) with respect to the spatial orientation of the lattice can be arbitrary (in particular, orthogonal). This in its turn may be substantial technical simplification of a setup.

It should be stressed that nowadays the classical two-photon spectroscopy is not seriously considered as a possible candidate for the primary frequency standards namely due to the significant shift \( \Delta \propto I \). Indeed, in the case of usual single-pulse spectroscopy this shift does not allow one to use the two-photon resonance as a reference point (for the primary frequency standards) due to fluctuations of the probe field intensity \( I \). But in the case of the Ramsey spectroscopy, apart from the large shift, here the condition \( |\Delta / V| > 1 \) can be fulfilled (here \( V \) is the two-photon Rabi frequency), which is not acceptable for the effective Ramsey spectroscopy. Thus, our method of additional shift-compensating field in the Ramsey spectroscopy opens new perspectives for the search of other variants of primary frequency standards. These variants can be considered as an extension (from the viewpoint of choice of atoms and/or clock transitions) or as a principal alternative (in the case of free atoms) to the modern direction of lattice-based clocks, in which the recoil effect and Doppler shift are suppressed.

Let us add that the method of shift-compensating laser field is quite universal, i.e. it is suitable for any transitions in any atoms and ions. However, it can not be excluded that in some cases the sign of the field frequency shift \( \Delta \) (for the magnetically induced spectroscopy or for the two-photon spectroscopy) will be so that for its compensation we can use a static electric (or even static magnetic) field, what can be a technical simplification.

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