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PRECISE THEORY OF THE LIGHT SHIFT IN OPTICAL FREQUENCY STANDARDS

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Abstract - **A general method is presented for calculating the higher-order terms of series in powers of the ac laser field for the Stark-state wavefunctions and corresponding frequency shifts of alkaline-earth atoms. It is based on the Green's function method for summation over all intermediate states. Numerical calculations of ac multipole polarizabilities and dipole hyperpolarizabilities for the light shift of $5s^2\ {}^1S_0$ - $5s5p\ {}^3P_0$ (M_J=0) transition frequency in ^{87}Sr atoms have been performed in connection with precise measurements on cooled and trapped alkaline-earth atoms and the design of precise optical frequency standards.**

Keywords - **Light shift, ac Stark effect, multipole susceptibilities**

I. INTRODUCTION

Alkaline- and alkaline-earth atoms are widely used in a number of important applications including atomic clocks, laser cooling of atoms at temperatures close to the recoil limit, parity-nonconserving experiments, the search for a permanent electron dipole moment, atomic interferometry, etc. Presently, there is also important interest in properties of divalent atoms in conjunction with low-temperature Bose-Einstein condensation experiments [1].

As a prime candidate for an optical frequency standards in neutral atoms, laser cooling of alkaline earth atoms have been studied in Mg, Ca, and Sr (see, for example, [2-7]). It is important to note, that optical frequency standards can be developed based on narrow transitions that allow exceptionnally high Q-factors, that are orders of magnitude higher than in Cs-fountains. On the other side, the configuration of two outer electrons introduces a unique complexity in their energy structure, specifically, the spin-singlet ground state and the spin-forbidden triplet excited states [2]. An "optical lattice clock" in Sr atoms has been proposed in [2,6-7], which utilizes millions of neutral atoms separately confined in an optical lattice that is designed to adjust the dipole polarizabilities for the probed electronic states in order to cancel light field perturbations on the measured spectrum. In striking contrast with conventional approaches toward frequency standards, the proposed scheme interrogates atoms while they are strongly perturbed by an external field. To further reduce potential uncertainties originating from collisional frequency shifts as well as tensor light shifts, in the paper [2,7] has been proposed to employ the $^1S_0(F=9/2)$-$^3P_0(F=9/2)$ transition of the ^{87}Sr isotope confined in a three-dimensional optical lattice.

This paper is motivated by emerging experiments on cooling and trapping of alkaline-earth atoms, in particular, on sideband cooling and spectroscopy neutral atoms in the atomic trap [2,6-7]. Here we report our theoretical results for the light shift in Sr atoms as a function of the trapping laser wavelength in the vicinity of the crossing point for the both $^3P_0$ and $^1S_0$ -levels.

II. THEORY AND NUMERICAL RESULTS

In order to provide theoretical support for measurements employing a "light shift cancellation technique", we have used general theory of [7-12] for calculating the light shift for the of $5s^2\ {}^1S_0$ - $5s5p\ {}^3P_0$ transition in ^{87}Sr atoms, including consecutive estimates for the higher-order field contributions (described by ac hyperpolarizabilities), on the basis of the Green's function method in the Fues' model potential approximation [9] for describing one-electron states in atoms.

The first result of numerical calculations for the second order field contributions presented in Fig.1 (the Stark shifts as determined by ac polarizabilities $\alpha(\omega)$ for $5s^2\ {}^1S_0$ and $5s5p\ {}^3P_0$ states) has already been described in [2].

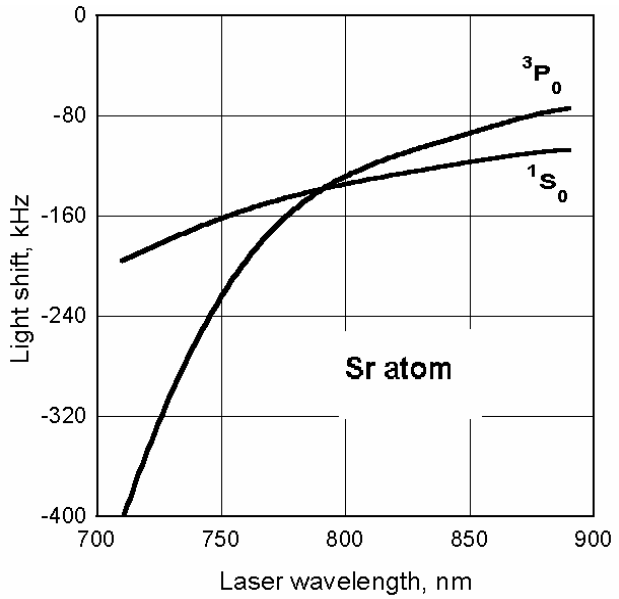

Fig1. Light shift as a function of the trapping laser wavelength at intensity 100 mW/π (17μm)^2. in the vicinity of the crossing point. A linearly polarized beam is used.

The model potential calculations have reproduced these data to within 3 percent accuracy both for the absolute value of the ac level shift and the frequency ω corresponding to equal values of the upper and lower level shifts. So we shall focus here on the calculation of the higher-order corrections to the transition frequency, described by the hyperpolarizability γ(ω) (the atomic units are used)

$$\nu = \nu^{(0)} - \frac{1}{4}\Delta\alpha(\vec{e},\omega)F^2 - \frac{1}{64}\Delta\gamma(\vec{e},\omega)F^4 - ..., \qquad (1)$$

and by the higher multipole corrections to the polarizability (magnetic dipole M1 and electric quadrupole E2 terms in addition to the electric dipole polarizability α (E1):

$$\alpha(\vec{e},\omega) = \alpha_{E1}(\vec{e},\omega) + \alpha_{M1}(\vec{e},\omega) + \alpha_{E2}(\vec{e},\omega). \qquad (2)$$

$\nu^{(0)}$ is the frequency of transition between unperturbed atomic states, $\Delta\alpha(\vec{e},\omega)$ ($\Delta\gamma(\vec{e},\omega)$) is the difference between the ac polarizabilities (hyperpolarizabilities) of the upper and lower levels, which in general case depend both on the light wave frequency ω and on the unit polarization vector $\vec{e}$. To calculate the E2 and M1 contributions to the polarizability (2) the magnetic dipole and electric quadrupole atom-field interaction should be taken into account together with the electric dipole term [7]

$$\begin{aligned} &\mathrm{H}(\vec{r},t) = V(\vec{r})e^{-i\omega t} + V^{+}(\vec{r})e^{i\omega t}; \\ &V = V_{E1} + V_{M1} + V_{E2}. \end{aligned} \qquad (3)$$

The magnetic dipole polarizability for the ground state equals zero, while for the excited state 5s5p $^3P_0$ its value is proportional to the fine-structure constant α=1/137 squared, to the splitting of the triplet states $E_{10} = E_{5p^3P_1} - E_{5p^3P_0}$ and to the square of the wave functions overlapping integral. Considerable values this quantity may have only in the closest vicinity of resonance, $\omega - E_{10} \approx \alpha^2$. The quadrupole polarizabilities of the both levels are of the order of $(\alpha\omega)^2$ and may also become considerable only in the closest vicinity of resonance on a quadrupole-allowed transition. Numerical estimates for the frequency ω_0 where $\Delta\alpha_{E1}(\omega_0) = 0$ gives $\alpha_{M1} \approx \alpha_{E2} \approx \alpha_{E1} \times 10^{-7}$ for the both levels.

The hyperpolarizability γ is calculated, starting from a formal expression for the second and fourth--order quasi-energies in terms of the field-free wave functions |0> and the reduced quasi-energy Green's functions G [9]

$$\Delta E^{(4)} = -\frac{F^4}{64}\gamma(\vec{e},\omega) = - << 0 \mid HGHGHGH \mid 0 >> + << 0 \mid HGH \mid 0 >><< 0 \mid HG^2H \mid 0 >> . \qquad (4)$$

Only the dipole term of equation (3) is taken into account in the interaction Hamiltonian. The double brackets in (4) mean the integration over the spatial variables and averaging over time.

After the time averaging and angular integration, applying standard angular momentum theory, the dipole dynamic polarizability and hyperpolarizability tensors may in general be resolved into 3 and 5 irreducible parts correspondingly, α_p, p=0,1,2 and γ_q, q=0,1,2,3,4, of which only scalar terms α_0 and γ_0 contribute in a state with the total momentum J=0 [9]. α_p and γ_q are determined by linear combinations of frequency-dependent radial matrix elements of the second and fourth order correspondingly. This dependence appears in the energy of stationary radial Green's functions.

So, the terms of the order of F^4 become more significant in the laser field than in a dc field of the same strength F, due to a more complicated polarization and frequency dependence of the hyperpolarizability in comparison with the polarizability. It is to note first that the scalar parts of $\gamma_0(\vec{e},\omega)$ are different for the linear and circular type of polarization, $\gamma^{l}_0(\omega) \neq \gamma^{c}_0(\omega)$, even for atoms in S-states, while all the terms of the polarizability (2) are independent of $\vec{e}$ for a state with J=0. Secondly, the number and type of singularities for hyperpolarizability exceeds that for the polarizability and the contribution of these singularities also depends on the polarization of the laser wave. E.g., for a linearly polarized radiation, there are two-photon singularities of $\gamma^{l}_0(\omega)$ on the (J=0)-states, while for the circular polarization such singularities cannot appear and the hyperpolarizability $\gamma^{c}_0(\omega)$ remains smooth. So, the hyperpolarizability of the ground state with two equivalent electrons $5s^2\ ^1S_0$ in Sr may be presented as

$$\begin{aligned} \gamma^{l}(\omega) &= \gamma^{c}(\omega) + \frac{8}{9}\left[\sigma_{101}(\omega,2\omega,\omega) + \frac{3}{5}\Sigma_{121} - \frac{2}{5}\sigma_{121}(\omega,2\omega,\omega)\right] \\ \gamma^{c}(\omega) &= \frac{8}{9}\left[\Sigma_{101} + \frac{1}{5}\Sigma_{121} + \frac{6}{5}\sigma_{121}(\omega,2\omega,\omega)\right] - 2\alpha_0(\omega)S_{-3}(\omega) \end{aligned} \qquad (5)$$

where

$$\alpha_0(\omega) = \frac{2}{3} < 0 \mid r\left(g^{\omega_1} + g^{-\omega_1}\right)r \mid 0 > \qquad (6)$$

is the polarizability,

$$S_{-3}(\omega)=\frac{2}{3}<0|r\left(g^{\omega_1}g^{\omega_1}+g^{-\omega_1}g^{-\omega_1}\right)r|0> \quad (7)$$

is the so-called frequency-dependent oscillator strengths moment; $\left(g^{\omega_1}\right)$ is the radial Green's function in the subspace of the jumping electron's states with angular momentum l; the following notations for the radial matrix elements and their combinations were used above:

$$\sum_{l_1l_2l_3}=\sigma_{l_1l_2l_3}(\omega,0,\omega)+\sigma_{l_1l_2l_3}(\omega,0,-\omega);$$

$$\sigma_{l_1l_2l_3}(\omega_1,\omega_2,\omega_3)=R_{l_1l_2l_3}(\omega_1,\omega_2,\omega_3)+R_{l_1l_2l_3}(-\omega_1,-\omega_2,-\omega_3),$$

$$R_{l_1l_2l_3}(\omega_1,\omega_2,\omega_3)=<0|rg_{l_1}^{\omega_1}rg_{l_2}^{\omega_2}rg_{l_3}^{\omega_3}r|0>. \quad (8)$$

In the vicinity of one-photon resonance with the frequency detuning $|\Delta|<<\omega$ the product of the second-order and third-order matrix elements in (4) (corresponding to the term $\alpha_0 S_{-3}$ in (5)) dominates (the third-order poles of order Δ^{-3}). The fourth-order matrix element in (4) has the second-order poles (Δ^{-2}). As is seen from equations (5), (7), the two-photon (2ω) resonance singularity on the J=0 states (in the radial matrix elements q appears only for $\gamma^{I}(\omega)$ and on the J=2 states (in $R_{121}(\omega,2\omega,\omega)$ both for $\gamma^{I}(\omega)$ and $\gamma^{c}(\omega)$.

The numerical results for $\gamma^{I}{}_{5s^2\,{}^1S_0}(\omega_0)$ and $\gamma^{(o)}{}_{5s5p\,{}^3P_0}(\omega_0)$ at the crossing point ω_0 (Fig.1) are 6.3×10^6 a.u and 2.7×10^8 a.u., respectively. Thus, as it seen from these data and Eq.(1) the fourth-order ac Stark contribution ($\Delta E_{5s^2\,{}^1S_0}(\omega_0)\approx-10^{-4}$ Hz and $\Delta E_{5s5p\ {}^3P_0}(\omega_0)\approx-10^{-4}$ Hz) is not significant for the employed trapping laser intensity of 100 mW/π $(17\mu m)^2$. The theory and the method for the numerical calculation of hyperpolarizability tensor for arbitrary alkaline-earth atoms, demands a separate examination and will be discussed in a forthcoming paper.

III. Conclusions

In connection with the laser cooling spectroscopy measurements which have been carried out and planned [2,6-7], we have investigated the light shift on the clock transition $5s^2\ {}^1S_0$ - $5s5p\ {}^3P_0$ in ^{87}Sr atoms. For that purpose we have used the general theory of [7-11] for consecutive estimates for the higher-order laser field contributions (described by ac hyperpolarizabilities). We have applied the analytic Sturm-series representation of the Green's function for the Fues model potential, corresponding to calculation of the infinite sums over the total atomic spectrum including continuum. It provides a reliable control for the accuracy of calculation with the finite sums of discrete states. Such estimated indicate good prospects for a ^{87}Sr optical clock on the 0-0 transition (below 1 Hz accuracy).

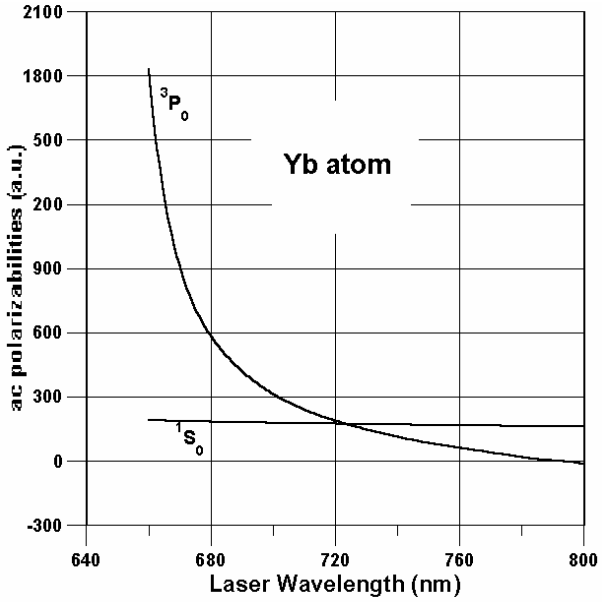

Fig. 2. Difference of polarizabilities (a.u.) as a function of the trapping laser wavelength at intensity 100 mW/π $(17\mu m)^2$. in the vicinity of the crossing point. A linearly polarized beam is used.

The present study may also be important for further experimental investigation on the Yb atoms (Fig.2) and, in particular, on other alkaline-earth Mg and Ca atoms, where the experimental accuracy can be improved considerably [3,5].

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