Elimination of inhomogeneous broadening for a ground-state hyperfine transition in an optical trap

Jai Min Choi and D. Cho
Department of Physics, Korea University, Seoul 136-713, Korea
E-mail: cho@korea.ac.kr

Abstract. We propose a way to eliminate the inhomogeneous broadening for a ground-state hyperfine transition of an alkali metal atom in an optical trap by using a properly polarized trapping field. The ac Stark shift contribution from the vector polarizability has opposite sign for a pair of ground hyperfine levels. It can be used to eliminate the inhomogeneous broadening from the difference in the scalar polarizabilities due to the hyperfine splitting. The size of the vector term is determined by the polarization state of the trapping field, and by controlling the polarization tightly one can achieve a very narrow linewidth. We estimate required tolerance in the polarization control to achieve 1-Hz linewidth for a specific case of a cesium atom. This proposal has significant implications for an electric dipole moment measurement using cesium atoms and quantum information processing using an optical lattice.

1. Introduction
One of the most important motivations of the early cooling and trapping research was to hold atoms longer so that more precise spectroscopic measurements could be made. When atoms are held in a trap, however, the trapping fields inevitably perturb internal states of an atom in addition to confining its center-of-mass motion. For example, in an optical trap, which is considered to be the least perturbing among various atom traps, opposite ac Stark shifts of a pair of states lead to a shift in resonance frequency and inhomogeneous broadening of the transition. One generally has to turn off the trapping fields and perform a spectroscopic measurement on expanding and falling atoms [1]. In the case of a fountain clock, atoms are tossed up from a magneto-optical trap to further increase the interrogation time [2]. In some special cases of an optical trap, it has been demonstrated that the differential ac Stark shift can be eliminated by exploiting a three-level configuration of an atom and carefully choosing the wavelength of the trapping light [3]. The idea was successfully employed to observe a perturbation-free strontium intercombination line for optical frequency metrology [4] and in a cavity QED experiment to study atom-photon interaction at the fundamental level [5].

Besides optical frequency metrology and cavity QED experiments, optically trapped atoms provide a tantalizing possibility for measurement of a permanent electric dipole moment (edm) of an atom [6]. The presence of a permanent edm of an elementary particle implies time reversal symmetry violation, and there has been a long and intense search for an edm [7]. The search for a neutron edm is the best known example. For an electron edm, the cesium atom is one of the best systems to study because an electron edm can lead to a large atomic edm in heavy atoms like cesium due to the so-called $Z^3$ enhancement, where $Z$ is the atomic number [8]. By
using large detuning and trap volume in a ultrahigh vacuum, one can hold many atoms for a long time to greatly increase precision. In addition, optically trapped atoms are stationary and the $\vec{v} \times \vec{B}$ systematic effect, which has been a limiting effect in beam-based atomic edm measurements [9], is not present. In spite of these promises, there is no published result on an edm measurement using optically trapped atoms. In order to measure an edm from cesium atoms one has to measure a small frequency shift of a transition between ground-state hyperfine levels. Due to the hyperfine splitting, however, each hyperfine level experiences a different ac Stark shift in an optical trap. The intensity gradient inherent in the trap leads to a large inhomogeneous broadening, wiping out the advantage gained by the long observation time. The three-level scheme, which was successfully applied to an optical transition, cannot be applied in the 9.2-GHz hyperfine transition.

One way to overcome this problem is to use a blue-detuned optical trap [10] where atoms are confined in a field-free region by the surrounding light field. However, a blue-detuned optical trap can hold much less atoms and due to diffraction there still remains an inhomogeneous broadening. In this paper we propose an alternative way to eliminate the differential ac Stark shift in a usual red-detuned optical trap by using the vector polarizability of a cesium atom. Although in most applications of an optical dipole force the scalar polarizability plays a dominant role, when the light is elliptically polarized, the circular component can give rise to a first-rank tensor interaction. The vector part of an ac Stark shift takes the form of a Zeeman shift, and one can associate a fictitious magnetic field with the shift [11]. In particular, the pair of ground hyperfine levels of an alkali metal atom have Landé $g$ factors of opposite sign, and with a proper polarization the differential ac Stark shift from the scalar part can be cancelled by the tensor part. In the following sections, we will describe a theory of tensor polarizabilities for an alkali metal atom in a ground state and its application for perturbation-free microwave spectroscopy in an optical trap. A few applications of the idea in addition to an edm measurement will be discussed at the end of the paper.

2. Theory

2.1. ac Stark shift in a spherical tensor form

When an atom in the state $|\Psi_0\rangle$ is irradiated by a laser field whose oscillating electric field is $\vec{E}(t) = \vec{E}_0 e^{-i\omega t} + \vec{E}_e^* e^{i\omega t}$, it experiences an ac Stark shift,

$$U_{AC}(\Psi_0) = \sum_{n \neq 0} \frac{\langle \Psi_0 | d \cdot \vec{E}_n | \Psi_n \rangle \langle \Psi_n | \vec{d} \cdot \vec{E} | \Psi_0 \rangle}{\omega_0 - \omega_n + \omega} + \sum_{n \neq 0} \frac{\langle \Psi_0 | \vec{d} \cdot \vec{E}_n | \Psi_n \rangle \langle \Psi_n | \vec{d} \cdot \vec{E}^* | \Psi_0 \rangle}{\omega_0 - \omega_n - \omega}, \quad (1)$$

where atomic units are used. $\omega_n$ is the unperturbed energy eigenvalue of the state $|\Psi_n\rangle$ and $\vec{d}$ is the electric dipole moment operator. The second term, which is off resonant, is much smaller than the first in most cases and in this paper we will consider only the first term [12]. It can be written as an expectation value $\langle \Psi_0 | \Omega(\vec{E}, \omega) | \Psi_0 \rangle$ with the operator $\Omega$ given by

$$\Omega(\vec{E}, \omega) = \sum_{i, j} \mathcal{E}_i^* d_i (\omega_0 - H_0 + \omega)^{-1} d_j \mathcal{E}_j, \quad (2)$$

where $H_0$ is the unperturbed atomic Hamiltonian. $\mathcal{E}_i$, $\mathcal{E}_j$ and $d_i$, $d_j$ for $i, j = x, y, z$ are the Cartesian components of $\vec{E}$ and $\vec{d}$, respectively. The operator can be rewritten in a spherical tensor form,

$$\Omega(\vec{E}, \omega) = \sum_{L=0}^{2} (-1)^L \sum_{m=-L}^{L} (-1)^m \mathcal{D}^{(L)}_m(\omega) \mathcal{F}^{(L)}_m, \quad (3)$$
where $D^{(L)}$ and $F^{(L)}$ with $L = 0, 1, 2$ are the scalar, vector and the second-rank spherical tensor operators:

$$D^{(L)}_m(\omega) = (-1)^{m+1} \sqrt{2L+1} \sum_{\mu=-1}^{1} \left( \begin{array}{c} 1 \\ \mu \\ m-\mu \\ -m \end{array} \right) d_\mu(\omega_0 - H_0 + \omega)^{-1}d_{m-\mu}. \quad (4)$$

Here $d_0 = z$ and $d_{\pm1} = \mp(x \pm iy)/\sqrt{2}$. $F^{(L)}_m$ is similarly defined in terms of $\mathcal{E}_i^*$ and $\mathcal{E}_j$ [13]:

$$F^{(0)}_0 = \frac{1}{\sqrt{3}}|\mathcal{E}|^2$$
$$F^{(1)}_m = -\frac{i}{\sqrt{2}}(\mathcal{E}^* \times \mathcal{E})_m$$
$$F^{(2)}_0 = \frac{1}{\sqrt{6}}(|\mathcal{E}|^2 - 3|\mathcal{E}_0|^2)$$
$$F^{(2)}_{\pm1} = -\frac{1}{\sqrt{2}}(\mathcal{E}_0^* \mathcal{E}_{\pm1} + \mathcal{E}_{\pm1}^* \mathcal{E}_0)$$
$$F^{(2)}_{\pm2} = -(\mathcal{E}_{\pm1}^* \mathcal{E}_{\pm1}),$$

where $\mathcal{E}_0 = \mathcal{E}_z$ and $\mathcal{E}_{\pm1} = \mp(\mathcal{E}_x \pm i\mathcal{E}_y)/\sqrt{2}$.

The scalar term in Eq. (3) is proportional to $|\mathcal{E}|^2$. The vector term leads to an ac Stark shift proportional to $\langle \Psi_0|D^{(1)}|\Psi_0 \rangle \cdot (\mathcal{E}^* \times \mathcal{E})$. The second-rank term is nonvanishing only if total angular momentum of the state $\Psi_0$ is at least 1. Alkali metal atom has a nuclear spin in addition to the electron spin. The consequent total angular momentum in cesium ground state, for example, is 3 or 4, and there is a contribution to the ac Stark shift from the second-rank term. However, when the detuning of the applied field from the $D$ transitions is much larger than the hyperfine splitting, as in a far-off-resonance optical trap, the second-rank term can be neglected. Under this approximation, the ac Stark shift of the ground hyperfine level $|nS_{1/2}, F, m_F\rangle$, where $F$ is the total angular momentum and $m_F$ is the magnetic quantum number, can be written as

$$U_{AC} = \alpha|\mathcal{E}_0|^2 - i\beta\langle S_{1/2}, F, m_F|\vec{d}|S_{1/2}, F, m_F \rangle \cdot (\mathcal{E}^* \times \mathcal{E}), \quad (5)$$

where $D^{(1)}$ in the vector term is replaced with the Pauli spin operator using the Wigner-Eckart theorem. $\alpha$ and $\beta$ are the scalar and the vector polarizabilities, respectively. The polarizabilities can be obtained by comparing Eq. (1) with Eq. (5) neglecting the hyperfine structure:

$$\alpha = \frac{1}{2} \sum_{n'} \frac{|\langle n'P_{1/2}\|d\|nS_{1/2}\rangle|^2}{\Delta_1} + \frac{1}{2} \sum_{n'} \frac{|\langle n'P_{3/2}\|d\|nS_{1/2}\rangle|^2}{\Delta_3} \quad (6)$$

and

$$\beta = \frac{1}{2} \sum_{n'} \frac{|\langle n'P_{1/2}\|d\|nS_{1/2}\rangle|^2}{\Delta_1} - \frac{1}{4} \sum_{n'} \frac{|\langle n'P_{3/2}\|d\|nS_{1/2}\rangle|^2}{\Delta_3}, \quad (7)$$

where $\langle n'P_{J}\|d\|nS_{1/2}\rangle$ is the reduced matrix element of the dipole operator $\vec{d}$ and $\Delta_{2J}$ is the detuning $\omega - (\omega_{n'P_{J}} - \omega_{nS_{1/2}})$. Detailed derivation of the formulae in the spherical tensor formalism is given in Ref. [11].

2.2. Differential ac Stark shift

This expression in Eq. (5) can be further simplified to

$$U_{AC} = \alpha|\mathcal{E}|^2 - \beta g_{Fm_F\eta}|\mathcal{E}|^2, \quad (8)$$
where \( g_F \) is the Landé \( g \) factor for the given \( F \) and \( \eta = |\hat{\epsilon} \times \hat{\epsilon}^*| \) is a measure of ellipticity. \( \hat{\epsilon} \) is the Jones vector representing the polarization state of \( \vec{E} \).

When the trapping field is linearly polarized, \( \eta = 0 \) and the ac Stark shift takes a simple form of \( U_{AC} = \alpha |\vec{E}|^2 \). Because of the ground hyperfine splitting, the \( F = 3 \) and \( F = 4 \) levels of the \( 6S_{1/2} \) state have different scalar polarizabilities and the difference in the ac Stark shifts is \((\alpha_{F=4} - \alpha_{F=3})|\vec{E}|^2 \). It can be approximately written as

\[
\delta U_{AC} \approx U_{AC} \frac{\delta_0}{\Delta},
\]

where \( \delta_0 \) is the ground hyperfine splitting and \( \Delta = (\Delta_1 + \Delta_3)/2 \). When the well depth is 0.1 mK and the trapping field is provided by a Nd:YAG laser with 1064 nm wavelength, the differential ac Stark shift corresponds to \( \delta f = 310 \) Hz. It means that the hyperfine transition frequency for the atoms at the bottom of the optical trap is shifted by \( \delta f \). We expect that inhomogeneous broadening due to the intensity gradient of the trapping field may have the same order of magnitude as \( \delta f \), although the exact linewidth should depend on temperature of the trapped atoms via the Maxwell-Boltzmann distribution. The photon scattering rate at the given trap parameters is on the order of 1 s\(^{-1}\), and the trap lifetime at ultrahigh vacuum condition can be much longer than 1 s. This implies that the unperturbed linewidth can be of order 1 Hz or smaller, and the inhomogeneous broadening is the dominant factor in determining the linewidth.

### 2.3. Elimination of the differential ac Stark shift

In an edm measurement, the signal-to-noise ratio is directly related to the linewidth. In order to achieve a linewidth limited by only the uncertainty principle, one needs to eliminate the inhomogeneous broadening. The \( \beta \) term in Eq. (8) has opposite sign for \( F = 3 \) and \( F = 4 \) and it can be used to cancel \( \delta U_{AC} \) due to \( \alpha_{F=4} \) = \( \alpha_{F=3} \), thereby eliminating the inhomogeneous broadening. Suppose an edm measurement is performed using a transition between the \( |6S_{1/2}, F = 3, m_F\rangle \) and \( |6S_{1/2}, F = 4, m_F\rangle \) states. If the trapping field is elliptically polarized with \( \eta \) as defined above, the difference in the ac Stark shifts of the two states is

\[
\delta U_{AC} = (\alpha_{F=4} - \alpha_{F=3})|\vec{E}|^2 + \frac{\beta}{4}(m_F + m_F')\eta|\vec{E}|^2,
\]

where we use \( g_{F=4} = -g_{F=3} = 1/4 \). It can be made vanish with proper choice of \( \eta \):

\[
\eta \approx \frac{4\alpha}{\beta(m_F + m_F')} \frac{\delta_0}{\Delta}.
\]

With the Nd:YAG laser \( \alpha = -1144 \) and \( \beta = -100 \) in atomic units. With \( m_F = m_F' = 3 \) as a benchmark case, \( \eta \approx 1.1 \times 10^{-3} \). It is a small number and it is obvious that either intended or unintended birefringence can easily introduce this much ellipticity. If one wants to eliminate \( \delta U_{AC} \) down to 1 Hz, \( \eta \) should be controlled at the level of \( 3.7 \times 10^{-6} \). One has to control the polarization of the trapping field very tightly and remove residual birefringence from a lens or a window of an apparatus very carefully.

### 2.4. Effect of the off-resonant terms

So far we have neglected the off-resonant terms in Eq. (1) in calculating the differential ac Stark shift. Their effects can be written as

\[
\delta U_{AC}^{\text{off-res}} \approx U_{AC} \frac{\Delta \delta_0}{(\omega_0 + \omega)^2},
\]

here \( \omega_0 \) is the average resonance frequency of the \( D1 \) and \( D2 \) transitions. Numerically it corresponds to only 1/100 of \( \delta U_{AC} \) due to the near resonant terms.
2.5. Effect of the upper hyperfine structures

If we neglected the fine structure and assumed $\Delta_1 = \Delta_3$, the vector polarizability in Eq. (7) would vanish because $\left| \langle n'P_{3/2} | d | nS_{1/2} \rangle \right|^2 = 2 \left| \langle n'P_{1/2} | d | nS_{1/2} \rangle \right|^2$. i.e. the fine structure gives rise to the vector polarizability. Similarly, the hyperfine structure gives rise to the second-rank tensor polarizability $\gamma$ as well as modifies $\alpha$ and $\beta$. Explicit expressions for the scalar, vector and second-rank tensor polarizabilities, which include the ground and upper hyperfine structures, are given in Ref. [14]. From the expressions, the extra differential ac Stark shift contributed by $\alpha$ and $\gamma$ due to the upper hyperfine structure is approximately given by

$$\delta U_{AC}^{\text{hyperfine}} \approx U_{AC} \xi \frac{\delta P}{\Delta},$$

where $\xi$ is from the Clebsch-Gordan coefficients and of the order $1/10$ and $\delta P$ is the upper hyperfine structure, which is smaller than 1 GHz. Numerically it also corresponds to only $1/100$ of $\delta U_{AC}$ in Eq. (9).

3. Discussions and summary

In this paper we made a proposal to eliminate the differential ac Stark shift between the ground hyperfine levels by using a properly polarized trapping field to exploit the vector polarizability. In realizing the proposal, one of the most challenging technical problems is a tight control of the polarization, at the level of a few times $10^{-6}$. The simplest way to control the polarization is using a waveplate. If the input field propagating along the $z$ axis is linearly polarized along the $x$ axis and a waveplate with retardation $r$ is oriented with its axis rotated by $\theta$ with respect to the $x$ axis, the output polarization has $\eta = \sin r \sin 2\theta$. When $r$ and $\theta$ are of similar magnitude, each angle is $\sim 2.3 \times 10^{-2}$ and each one should be controlled to $5.6 \times 10^{-5}$ in order to achieve 1-Hz linewidth. This appears to be a rather tight tolerance. One has to (i) start with a clean linear polarization, (ii) control $r$ and $\theta$ very tightly, and (iii) eliminate or control residual birefringence from a lens or a window to a high precision. The temperature of the waveplate and other optical surfaces should be stabilized to remove thermal drift of the birefringence, and there should be a way to measure the polarization change precisely. Eventually the measured linewidth of the hyperfine transition will be an ultimate measure of $\eta$. By monitoring both the polarization and the linewidth simultaneously while varying $\theta$, one may be able to achieve the 1-Hz linewidth.

We note that our proposed scheme to eliminate the differential ac Stark shift is not applicable to a clock transition between $m_F = 0$ and $m'_F = 0$ because the vector term vanishes for the states. For an atomic clock, fountain remains a most viable solution to achieve perturbation free operation. However, for an edm measurement, which requires application of a strong electric field throughout the interrogation time, fountain arrangement is not practical. Optical trap can be easily incorporated with field plates. In addition, the proposed scheme has a potential to achieve smaller linewidth than can be obtained with a fountain.

If one can achieve 1-Hz linewidth for a ground hyperfine transition for an alkali metal atom in an optical trap, there are many other interesting experimental possibilities besides edm measurement. As an example, one can perform sideband resolved spectroscopy. The situation is analogous to an ion trap except that it is a microwave transition with many atoms. In particular, if one uses a standing wave configuration with many wells, it has an implication for many-qubit quantum information processing. For this type of experiment, one can use $m_F = 0$ and $m'_F = 1$, and the sensitivity of the linewidth on $\eta$ is reduced by a factor of 6 compared with our benchmark case.

Acknowledgments

This work was supported by the Korea Science and Engineering Foundation (F01-2005-10214) and by the Seoul R&BD program.
References
[1] Wilpers G, Binnewies T, Degenhardt C, Sterr U, Helmcke J and Riehle F 2002 Phys. Rev. Lett. 89 230801
[2] Gibble K and Chu S 1993 Phys. Rev. Lett. 70 1771
[3] Kim J Y, Lee J S, Han J H and Cho D 2003 J. Korean Phys. Soc. 42 483
[4] Takamoto M, Hong F L, Higashi R and Katori H 2005 Nature 435 321
[5] Mckeever J, Boca A, Boozer A D and Kimble H J 2003 Nature 425 268
[6] Bijlsma M, Verhaar B J and Heinzen D J 1994 Phys. Rev. A 49 R4285
[7] Commins E D 1999 Adv. At. Mol. Opt. Phys. 40 1
[8] Johnson W R, Guo D S, Idrees M and Sapirstein J 1986 Phys. Rev. A 34 1043
[9] Regan B C, Commins E D, Schmidt C J and DeMille D 2002 Phys. Rev. Lett. 88 071805
[10] Kuga T, Torii Y, Shiokawa N, Hirano T, Shimizu Y and Sasada H 1997 Phys. Rev. Lett. 78 4713
[11] Cho D 1997 J. Korean Phys. Soc. 30 373
[12] In an optical trap with extremely large detuning, the second term may not be neglected.
[13] A. Khadjavi, A. Lurio, and W. Happer, Phys. Rev. 167, 128 (1968).
[14] Kim J Y and Cho D 2000 J. Korean Phys. Soc. 37 744