Light shifts and their spherical-tensor properties

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https://bartmcguyer.com/notes/note-9-LightShifts.pdf

TL;DR: A brief introduction to some special topics about light shifts for atoms and molecules and why their spherical-tensor properties are useful.

This note's intended to help experimental physicists familiar with light shifts learn more about their spherical-tensor properties, which enable you to take advantage of the mathematical machinery from the quantum theory of angular momentum. This was very useful in my previous work to engineer magic-wavelength optical lattices[1,2] and exploit mixed quantization in spectroscopy.[3] Additionally, it provides a short introduction to some special topics about light shifts.

CONTENTS

I. LIGHT SHIFTS

If you use light to study particles like atoms or molecules, it's important to understand the ways that the light can change (or shift) each particle's energy. Such light shifts (also known as dynamic or AC Stark shifts) are often used to confine and control particles in optical dipole traps and lattices. However, light shifts can also perturb measurements or produce errors, for example, in atomic clocks. For an introduction to light shifts, I recommend Refs. 4–8.

To proceed, let's assume that the particles are neutral and nonpolar, that there is enough light to treat it semiclassically, and that the light only induces virtual transitions. The origin of the light shift is then the electric-dipole (E1) interaction

$$V_{\mathrm{E1}} = -\mathbf{d} \cdot \mathbf{E}(t) \tag{1}$$

of the light's electric field $\mathbf{E}(t)$ and the particle's instantaneous electric-dipole moment $\mathbf{d}$. This interaction is nonzero only between quantum states of neutral particles with different parities, so only leads to second- and higher-order perturbations. Note that this interaction may also affect the hyperfine and Zeeman structure of particles (see below).[9,10]

While higher-order perturbations do exist and are important (e.g., to atomic clocks[11]), the "light shift" almost always refers to the second-order energy perturbation from (1). Conventionally, this shift is expressed using an electric-dipole polarizability α as the work

$$W = -\frac{1}{2}\operatorname{Re}(\alpha)\left\langle|\mathbf{E}(t)|^2\right\rangle \tag{2}$$

for a single particle, where the brackets denote a time average and the factor of $1/2$ comes from inducing the dipole moment. The complex-valued polarizability α captures how the shift depends on the properties of the particle and of the light, and is well known for most combinations of particles and wavelengths of experimental interest.

There are some rather important properties of light shifts that aren't obvious from the simple form of (2). We can expose many of them by rewriting the light shift in a spherical-tensor form, even without actually calculating the shift. In the next section, we'll do just that to illustrate the light shift's dependence on the electric field's geometry, the light's polarization, and the particle's quantum state (angular momentum), as well as its ability to couple and mix different states with "off-diagonal" shifts. Afterwards, we'll touch on some additional special topics, such as why extrapolating to zero intensity ($W \propto |\mathbf{E}(t)|^2 \longrightarrow 0$) doesn't always remove light shifts, despite the appearance of (2).

II. SPHERICAL-TENSOR FORM FOR LIGHT SHIFTS

While the light shift (2) is a second-order perturbation from the interaction (1), it's convenient to model it as a first-order expectation of an effective potential using a polarizability operator (or polarizability tensor) $\boldsymbol{\alpha}$.[9,12] In this approach, the light shift is represented by the first-order expectation of a potential that is conventionally written in a dyadic form as

$$V_{\text{eff}} = -\frac{1}{2}\left\langle\mathbf{E}(t)\cdot\boldsymbol{\alpha}\cdot\mathbf{E}^*(t)\right\rangle, \tag{3}$$

where again brackets denote a time average. The same potential can often be used for several states of interest, such as the ground-state manifold of an atom or molecule. Approximations like (3) are common in molecular physics, for example, where they are sometimes called effective Hamiltonians. They are typically derived using a Van Vleck, contact, or related transformation.[9,10,12,13] Importantly, they reproduce not only energy shifts but also mixing effects to second order in the applied field strength. For more on this, I recommend the treatment of Van Vleck transformations in Ref. 13 (p. 315).

To proceed, we won't need to know much more about $\boldsymbol{\alpha}$. However, for clarity, we can connect (2) and (3) as follows. For a particle with eigenstates $|j\rangle$ and energies E_j, the light shift $W = \langle\psi|V_{\text{eff}}|\psi\rangle$ of a chosen state $|\psi\rangle$ from monochromatic light with angular frequency ω can usually be modeled by $\boldsymbol{\alpha} = (2/\hbar)\sum_{j\neq\mu}\mathbf{p}|j\rangle\langle j|\mathbf{p}\,\omega_j/(\omega_j^2-\omega^2)$ with transition

frequencies $\omega_j = (E_j - E_k)/\hbar$, in the rotating-wave (or secular) approximation. This gives the typical result $W = -(1/\hbar)\sum_{j \neq \psi}\langle\psi|\mathbf{E}\cdot\mathbf{p}|j\rangle\langle j|\mathbf{p}\cdot\mathbf{E}^*|\psi\rangle\omega_j/(\omega_j^2 - \omega^2)$.[12]

Finally, we can rewrite (3) in spherical-tensor form using Eq. (5.52) of Ref. 14 as

$$V_{\text{eff}} = -\frac{1}{4}\sum_{k=0}^{2}(-1)^k\, T^k(\boldsymbol{\alpha}) \cdot T^k(\mathbf{E}, \mathbf{E}^*)$$

$$= -\frac{1}{4}\sum_{k=0}^{2}\sum_{p=0}^{k}(-1)^{k-p}\, T_p^k(\boldsymbol{\alpha})\, T_{-p}^k(\mathbf{E}, \mathbf{E}^*), \tag{4}$$

assuming monochromatic light with the time-independent field amplitude $\mathbf{E}$ and its complex conjugate $\mathbf{E}^*$. Here and subsequently, the spherical-tensor notation follows Ref. 13. For convenience, the second line expands the tensor contractions to show the tensor elements T_p^k. The effects of the different contractions with $k = 0$, 1, and 2 are usually called "scalar," "vector," and "tensor" light shifts, respectively. Sometimes, the tensor rank k is referred to as an angular momentum. If this seems odd for potentials and perturbations (versus particles), note that it's just a mathematical property from the rotational invariance of (1).

The spherical-tensor form (4) is useful because it separates the dependence on the particle from the dependence on the field. That is, the tensor $T^k(\boldsymbol{\alpha})$ only operates on the particle and the tensor $T^k(\mathbf{E}, \mathbf{E}^*)$ only depends on the field. Therefore, the particle and field portions can be evaluated independently. However, note that the light's frequency ω enters parametrically through $\boldsymbol{\alpha} = \boldsymbol{\alpha}(\omega)$. To help evaluate expressions like (4), I recommend the summary of spherical-tensor algebra at the end of Ch. 5 in Ref. 13 (p. 171).

The form (4) directly illustrates the dependence on the light's geometry and polarization through the tensor $T^k(\mathbf{E}, \mathbf{E}^*)$. For reference, explicit formulas for the elements $T_p^k(\mathbf{E}, \mathbf{F})$ in terms of Cartesian components, spherical components, and vector expressions are

$$T_0^0(\mathbf{E}, \mathbf{F}) = -\frac{1}{\sqrt{3}}(E_x F_x + E_y F_y + E_z F_z) = -\frac{1}{\sqrt{3}}\mathbf{E}\cdot\mathbf{F} \tag{5}$$

$$T_0^1(\mathbf{E}, \mathbf{F}) = \frac{i}{\sqrt{2}}(E_x F_y - E_y F_x) = \frac{i}{\sqrt{2}}(E_1 F_{-1} - E_{-1} F_1) = \frac{i}{\sqrt{2}}[\mathbf{E}\times\mathbf{F}]_z \tag{6}$$

$$T_{\pm 1}^1(\mathbf{E}, \mathbf{F}) = \frac{1}{2}[\pm i(E_y F_z - E_z F_y) - (E_z F_x - E_x F_z)] = \pm\frac{1}{\sqrt{2}}(E_{\pm 1} F_0 - E_0 F_{\pm 1})$$

$$= \frac{1}{\sqrt{2}}(\pm i[\mathbf{E}\times\mathbf{F}]_x - [\mathbf{E}\times\mathbf{F}]_y) \tag{7}$$

$$T_0^2(\mathbf{E}, \mathbf{F}) = \frac{1}{\sqrt{6}}(2E_z F_z - E_x F_x - E_y F_y) = \frac{1}{\sqrt{6}}(E_1 F_{-1} + 2E_0 F_0 + E_{-1} F_1)$$

$$= \frac{1}{\sqrt{6}}\mathbf{E}\cdot(3\hat{z}\hat{z} - \mathbb{1})\cdot\mathbf{F} \tag{8}$$

$$T_{\pm 1}^2(\mathbf{E}, \mathbf{F}) = \frac{1}{2}[\mp(E_x F_z + E_z F_x) + i(E_y F_z + E_z F_y)] = \frac{1}{\sqrt{2}}(E_0 F_{\pm 1} + E_{\pm 1} F_0) \tag{9}$$

$$T_{\pm 2}^2(\mathbf{E}, \mathbf{F}) = \frac{1}{2}[E_x F_x - E_y F_y \pm i(E_x F_y + E_y F_x)] = E_{\pm 1} F_{\pm 1} \tag{10}$$

where the vector spherical components $E_0 = E_z$ and $E_{\pm 1} = i(E_y \mp E_x)/\sqrt{2}$ (Ref. 13 pp. 161–162, with minor fixes for T_p^1; Ref. 15 pp. 64–65).

An immediate consequence is that the geometry and polarization of the light only affect the vector and tensor shifts, because $T_0^0(\mathbf{E}, \mathbf{E}^*) \propto |\mathbf{E}|^2$. Additionally, there are no vector shifts for linearly polarized light, because $T_p^1(\mathbf{E}, \mathbf{E}^*) = 0$ if $\mathbf{E} = \mathbf{E}^*$. If the geometry or polarization are variable, these elements will show how that variability affects the different terms in the light shift. For example, for linearly polarized light with a variable orientation such that $\mathbf{E} \propto \cos(\theta)\hat{z} + \sin(\theta)\hat{x}$, the only nonzero elements are $T_0^0(\mathbf{E}, \mathbf{E}^*) = -|\mathbf{E}|^2/\sqrt{3}$, $T_0^2(\mathbf{E}, \mathbf{E}^*) = [3\cos(\theta)^2 - 1]|\mathbf{E}|^2/\sqrt{6}$, $T_{\pm 1}^2(\mathbf{E}, \mathbf{E}^*) = \mp \sin(2\theta)|\mathbf{E}|^2/2$, and $T_{\pm 2}^2(\mathbf{E}, \mathbf{E}^*) = \sin(\theta)^2|\mathbf{E}|^2/2$.

For particles with energy eigenstates that have well-defined angular momentum, the form (4) directly illustrates which terms cause "diagonal" energy shifts ($p = 0$) and which cause "off-diagonal" state mixing ($p \neq 0$), assuming the tensors are rotated to have the same quantization axis as the particle. This is because p directly relates to the change in azimuthal angular momentum quantum number m through the Wigner-Eckart theorem: $\langle Jm|T_p^k|J'm'\rangle \propto \begin{pmatrix} J & k & J' \\ -m & p & m' \end{pmatrix}$. Another consequence of this theorem is that vector shifts exist only if the quantum number $J \geq 1/2$, and tensor shifts exist only if $J \geq 1$.

Note that when off-diagonal mixing is significant, it can influence the particle's quantization. However, if the particle's quantization is not affected by the mixing, or approximately so, then the $p \neq 0$ terms can usually be ignored. Returning to the example of linearly polarized light with $\mathbf{E} \propto \cos(\theta)\hat{z} + \sin(\theta)\hat{x}$, the elements $T_0^0(\mathbf{E}, \mathbf{E}^*)$ and $T_0^2(\mathbf{E}, \mathbf{E}^*)$ will produce diagonal light shifts, and the elements $T_{\pm 1}^2(\mathbf{E}, \mathbf{E}^*)$ and $T_{\pm 2}^2(\mathbf{E}, \mathbf{E}^*)$ will produce off-diagonal mixing. Assuming the particle's quantization is fixed, only the first two elements with $p = 0$ will be significant, and the angle θ can be used to tune the light shift.

With some effort, one can clean up the results for the typical case of linearly polarized light and a particle in an angular-momentum state $|Jm\rangle$ with $J \geq 1$ so that the light shift is given by the form (2) with the effective polarizability

$$\alpha = \alpha(J, m, \omega, \theta) = \alpha_0(J, \omega) + \alpha_2(J, \omega) \left(\frac{3\cos^2\theta - 1}{2}\right)\left(\frac{3m^2 - J(J+1)}{J(2J-1)}\right). \tag{11}$$

This follows from the Wigner-Eckart theorem and repackaging its reduced matrix elements into scalar α_0 and tensor α_2 polarizabilities so that α matches the standard "J representation" form[16] when $\theta = 0$. Again, (11) assumes the quantization axis is fixed, for example, by an applied magnetic field, which leads to the control of the tensor shift through the angle θ of tilt between the linear polarization orientation and the quantization axis. For other light polarizations, a similar effective polarizability can be constructed that will include vector shifts.[17] Please note that care is needed in using formulas such as (11) for atoms with hyperfine structure.[10,17]

Before we continue, please note that there are many different conventions and notations for light shifts, particle polarizabilities, light polarizations, and spherical tensors. Therefore, please double check any formulas you use, including those in this note. In full disclosure, I'm not confident I've sanitized this note of all typo's, especially for complex-valued $\mathbf{E}$. Please feel free to share corrections by emailing contact-me (at) bartmcguyer (dot) com.

For much more on this subject, I recommend Ch. 19 of the textbook Ref. 17. For a detailed derivation of light shifts for a complex alkali-metal atom (Cs), I recommend the

paper Ref. 10. For calculating polarizabilities for alkaline-earth atoms like Ca, Mg, and Sr, I recommend Ref. 18. And last but not least, for spectroscopic data to input into such calculations, I recommend the NIST ASD database.[19]

A. "Magic" wavelengths, angles, and traps

Light shifts often produce undesirable effects when they aren't identical for certain states of a particle, say ground and excited states or different m sublevels. Fortunately, it's often possible to arrange an experiment's conditions to null, or minimize, the differential light shifts between certain states. Such conditions are traditionally called "magic."

For example, consider a measurement of a transition linewidth for a particle held in an optical trap made from linearly polarized light. If the trap's light shift is not identical for the states connected by the transition, then the trap will broaden the measured linewidth. To remove this broadening, the simplest approach is to turn off the trap, making the total light shifts from the trap zero for both states. While this is a common approach to remove shifts from non-trapping light, for trapping light it's undesirable because it also releases the particles and limits the measurement time, introducing its own broadening. Instead, we could make a "magic" trap with identical light shifts for the states connected by the transition. Without introducing new light, we can do this by making the effective polarizabilities of each state, both of the form (11), identical using a "magic" (zero-shift or tune-out) wavelength and also a "magic" angle θ (if $J \geq 1$) for the trapping light.

This is particularly straightforward if the states of interest are the different sublevels m of the same manifold with $J \geq 1$, say in the particle's ground state. In this case, they share the same effective polarizability (11), whose value depends on $|m|$ so is magic already for sublevels with the same $|m|$. The polarizability can be tuned to be identical for all m by nulling the tensor shift, which occurs at the well-known[4] magic angles of $\theta = \pm \arccos\left(1/\sqrt{3}\right) \approx \pm 54.7°$. These zero-tensor-shift magic angles have a long history (i.e., ^{201}Hg, p. 99 of Ref. 9) and continue to find applications (e.g., trapping polar KRb[20]).

Note that a given magic wavelength likely isn't suitable for all experiments. For example, optical traps generally need magic wavelengths far from any particle transitions (resonances), to keep particle lifetimes and temperatures as long and cold as possible in the trap. Finding such far-off-resonance magic wavelengths tends to require precise theoretical calculations. However, traditional optical-pumping experiments almost exclusively use resonant light, so need magic wavelengths very close to a transition to keep pumping rates high. Fortunately, magic wavelengths tend to exist near transitions because the differential light shift for an ideal two-level system has a dispersive shape that crosses zero at resonance. For real atoms with hyperfine structure, such as those used in Rb vapor-cell atomic clocks, it's a little more complicated. For an introduction to this, I can offer Section 2.1.3 of my dissertation, Ref. 8. Interestingly, it took until 2016[21] to precisely measure the light shift of the ^{87}Rb 0–0 clock transition versus wavelength, which matched theoretical calculations from 1968.[22]

For an example of using (11) to experimentally find magic conditions for an optical lattice, I offer my previous work with ^{88}Sr$_2$ in Ref. 1. In particular, the Fig. S1 of its Supplementary Information plots data showing the wavelength and angle dependence of (11).

For three examples of feedback techniques to suppress the light shift in vapor-cell atomic clocks, I offer my previous work with Rb and Cs in Refs. 8 and 23 and recommend Ref. 24.

B. Mixing from off-diagonal light shifts

It seems to be less appreciated that light shifts also induce state mixing in addition to energy shifts, just like static electric and magnetic fields do. After the discovery of light shifts, though, such mixing seems to have been readily explored, for example, to induce resonances from fictitious fields,[25] to alter Zeeman shifts,[25] and to change ground-state structure.[26] Perhaps this is because mixing effects are typically less significant than energy shifts. Nevertheless, it's important to be aware of this mixing because it can affect state quantization, energies, spectroscopy, transition strengths, and experimental procedures, such as nulling stray magnetic fields (see below). This mixing can also be exploited in experiments, and may offer opportunities to, say, create novel optical traps or fine-tune the magnetic sensitivity of clock states similar to radio-frequency dressing techniques.

For an example of an experiment in which such mixing was significant and beneficial, I offer my previous work with $^{88}Sr_2$ spectroscopy in Ref. 3. This work studied transition strengths between states with different quantizations: ground states with mixed quantization from a competition of tensor light shifts and weak Zeeman shifts, and excited states quantized by strong Zeeman shifts. This differing quantization simplified the molecular spectroscopy by allowing the measurement of transitions between all possible ground and excited sublevels by detecting only a few ground sublevels. For example, Fig. 4 of Ref. 3 shows how this mixed quantization created the illusion of highly forbidden $|\Delta m| = 4$ single-photon transitions. Such mixed quantization also affected the measurement of transition strengths, requiring a correction derived in the Supplementary Material.

One common experimental procedure where this mixing is likely to be significant is the nulling of applied magnetic fields, because near zero total magnetic field such mixing can cause dramatic changes to quantization. For an example of this, I offer Fig. 3.13 in Section 3.4.1.2 of the dissertation Ref. 27. This phenomenon can also perturb measurements that extrapolate to zero total magnetic field, say to determine a zero-field binding energy.

C. Importance of simultaneous perturbations

Last but far from least, note that accurate calculations of light shifts and magic conditions require treating all competing perturbations simultaneously.[17] The effective potential (4) does not account for any such perturbations. Remember, the actual interaction is (1) and the light shift is its second-order perturbation, modeled for convenience as a first-order expectation of (4). For particle states with hyperfine or other significant interactions, all such interactions should be treated on equal footing, simultaneously with (1). Not doing this has led to incorrect predictions for magic conditions.[10,28] Therefore, make sure to search for the most in-depth literature on light shifts relevant to your experiment.

For an introductory example, I offer my previous work with hyperfine shifts of alkali-

metal atoms in Ref. 29. This work calculates the change in ground hyperfine couplings from static electric fields in its Appendix (DC Stark shifts), which are important in atomic clocks. The leading effect is a third-order perturbation linear in hyperfine coupling and quadratic in (1). I offer this work because it's simpler than full-scale calculations of magic conditions.

III.   SPECIAL TOPICS

Light shifts are a surprisingly rich subject, providing a seemingly endless supply of special topics. This is probably unavoidable since they're one aspect of the vast subject of light-matter interactions. For experimentalists, though, this can make learning about them feel daunting, since the phrase "light shift" refers to different things in different contexts. To make this a little more clear, notice that this note only considered the most common type of light shifts. It did not at all address other important aspects, such as shifts for polar particles or ions, from real transitions, for quantized light (photons), or for multi-photon spectroscopy, nor the Autler-Townes effect (e.g., Mollow triplet), dressed states, or the connection with index of refraction and slow light. All of these aspects and more are worth exploring further.

While I've already included many examples from my previous work, please allow me to conclude this note with a few more to share the following special topics.

A.   Error from extrapolating to zero light intensity

A conventional approach to remove measurement errors due to light shifts is to extrapolate to zero light intensity ($|\mathbf{E}(t)|^2 \longrightarrow 0$). This follows from the form of (2), which shows that the light shift is proportional to the light intensity: $W \propto |\mathbf{E}(t)|^2$. However, this isn't guaranteed to always work for at least the following three reasons.

First, remember that the form (2) only includes second-order perturbations from (1), which are linear in intensity. In addition, there will be higher-order perturbations from (1) that are nonlinear with the light intensity, so do not linearly extrapolate to zero. Such nonlinear light shifts are important in very precise measurements and atomic clocks.[11]

Second, remember that the form (2) only connects the light shift at a given location with the intensity at that same location. If the electric field varies spatially, then the actual light shift for a particle depends on how it spatially samples the varying field. If this spatial sampling depends on the light intensity, as it usually does for particles in an optical trap, then extrapolating to zero intensity isn't guaranteed to fully remove the light shift. While such an error is generally small, it may be significant in precision measurements and optical lattice clocks. For more on this, I offer my work with $^{88}\mathrm{Sr}_2$ thermometry in Ref. 2.

Third, similarly, for spatially nonuniform non-trapping light, for example, in optical pumping experiments, the superposition of lineshapes from an ensemble may have a line center that varies nonlinearly with intensity.[30]

B. Measuring temperature with light shifts

If the light intensity varies spatially, then the light shift for a particle depends on how it samples the intensity, as mentioned above. As a result, for an ensemble of particles confined in an optical trap, the distribution of motional states maps into a distribution of light shifts. Quite often this is a nuisance because it broadens transitions and dephases ensembles. However, it also provides a direct way to measure temperature using light shifts. My previous work with $^{88}Sr_2$ in Ref. 2 demonstrated this by using a nearly magic optical lattice to imprint the temperature into the lineshape for a narrow transition. Unlike the conventional approach of using sideband areas, this approach only depends on frequencies.

C. Suppressing light shifts with quadrature error signals

My introduction to light shifts came from working to remove them in Rb vapor-cell atomic clocks.[31] Such clocks use a phase-sensitive, frequency-modulated feedback loop to lock a signal generator's frequency to a resonant "clock" frequency for an ensemble of Rb atoms. In a real clock, the Rb atoms will rarely share the same clock frequency because light shifts (and other shifts) differ spatially throughout the vapor cell. Surprisingly, this spread in clock frequencies produces an out-of-phase (quadrature) error signal that can be used to lock a laser to a magic wavelength.[23] The simplest explanation of the interference phenomenon producing this quadrature signal is available in Section 2.2.1 of my dissertation, Ref. 8. I couldn't resist including this topic because I suspect that similar interference phenomena may also be present in other applications that use feedback to lock to resonances.

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