Functional Form of the Imaginary Part of the Atomic Polarizability

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Abstract. The dynamic atomic polarizability describes the response of the atom to incoming electromagnetic radiation. The functional form of the imaginary part of the polarizability for small driving frequencies ω has been a matter of long-standing discussion, with both a linear dependence and an ω^3 dependence being presented as candidate formulas. The imaginary part of the polarizability enters the expressions of a number of fundamental physical processes which involve the thermal dissipation of energy, such as blackbody friction, or non-contact friction. Here, we solve the long-standing problem by calculating the imaginary part of the polarizability in both the length ("$\boldsymbol{d} \cdot \boldsymbol{E}$") as well as the velocity-gauge ("$\boldsymbol{p} \cdot \boldsymbol{A}$") form of the dipole interaction, verify the gauge invariance, and find general expressions applicable to atomic theory; the ω^3 form is obtained in both gauges. The seagull term in the velocity gauge is found to be crucial in establishing gauge invariance.
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1 Introduction

The "susceptibility" of an atom toward the generation of an atomic dipole moment is described by the dynamic atomic (dipole) polarizability. Alternatively, the atomic polarizability describes photon absorption from a field of photons and subsequent emission of a photon into the same or a vacuum mode of different wave vector but the same frequency. The external oscillating field can be a laser field (and this is the intuitive picture we shall use in this paper). In this case, the real part of the polarizability describes the ac Stark shift [1]. The atomic polarizability has both a real as well as an imaginary part. Physically, this can be illustrated by considering the relative permittivity $\epsilon_r(\omega)$ of a dilute gas and its relation to the dynamic dipole polarizability $\alpha(\omega)$ of the gas atoms,

$$\epsilon_r(\omega) = 1 + \frac{N_V}{\epsilon_0} \alpha(\omega) \,, \tag{1}$$

where ϵ_0 is the vacuum permittivity and N_V is the volume density of gas atoms [2]. The Kramers–Kronig relations dictate that the real part of $\epsilon(\omega)$ should be an even function of the driving frequency ω, while the imaginary part of $\epsilon(\omega)$ cannot vanish and should be an odd function of ω. These considerations are valid upon an interpretation of the dielectric constant in terms of the retarded Green function G_R which describes the relation of the dielectric displacement $\boldsymbol{D}(\boldsymbol{r}, t)$ to the electric field $\boldsymbol{E}(\boldsymbol{r}, t)$,

$$\boldsymbol{D}(\boldsymbol{r}, t) = \epsilon_0 \, \boldsymbol{E}(\boldsymbol{r}, t) + \epsilon_0 \int_0^\infty \mathrm{d}\tau \, G_R(\tau) \, \boldsymbol{E}(\boldsymbol{r}, t - \tau) \,. \tag{2}$$

The Fourier transform is

$$G_R(\omega) = \frac{N_V}{\epsilon_0} \alpha(\omega) \,. \tag{3}$$

The symmetry properties of the imaginary part of the polarizability have been discussed at various places in the literature [3–7]. The conventions used here and also the result of our calculation of the non-resonant contribution to the imaginary part, to be reported in the following, are consistent with the interpretation as a retarded Green function and, notably, with the conclusions of Ref. [4] [see Eq. (2) and the text following Eq. (31) of Ref. [4]].

The imaginary part of the polarizability enters the description of processes such as the black-body friction [8,9], where an atom is decelerated by interaction with a thermal bath of black-body photons. The blackbody (BB) friction force $F_{\mathrm{BB}} = -\eta_{\mathrm{BB}} \, v$ is linear in the velocity,

$$\eta_{\mathrm{BB}} = \frac{\beta \hbar^2}{12\pi^2 \, \epsilon_0 \, c^5} \int_0^\infty \frac{\mathrm{d}\omega \, \omega^5 \, \mathrm{Im}[\alpha(\omega)]}{\sinh^2(\frac{1}{2}\beta \hbar \omega)} \,. \tag{4}$$

Here, $\beta = 1/(k_B \, T)$ is the Boltzmann factor, c is the speed of light, and $\hbar$ is the quantum unit of action (reduced Planck constant), while the integration is carried out over the entire blackbody spectrum of angular frequencies ω. This force is due to dissipative processes; the atom absorbs an incoming blue-shifted photon from the front, and emits photons in all directions. The friction force at a distance $\mathcal{Z}$ above a surface composed of a dielectric material, due to

the dragging of the mirror charges inside the electric with frequency-dependent dielectric constant $\epsilon(\omega)$, is calculated as $F_{\mathrm{QF}} = -\eta_{\mathrm{QF}}\, v$. According to Ref. [10], the result for the non-contact quantum friction (QF) coefficient in the relation is given by a Green–Kubo formula,

$$\eta_{\mathrm{QF}} = \frac{3\beta\hbar^2}{32\pi^2\epsilon_0\mathcal{Z}^5} \int\limits_0^\infty \frac{\mathrm{d}\omega\,\mathrm{Im}[\alpha(\omega)]}{\sinh^2(\frac{1}{2}\beta\hbar\omega)} \mathrm{Im}\left[\frac{\epsilon(\omega)-1}{\epsilon(\omega)+1}\right]\,. \quad (5)$$

The formulas (4) and (5) raise the question what could be deemed to be the most consistent physical picture behind the imaginary part of the polarizability. The imaginary part of the polarizability involves a spontaneous photon emission process, and this spontaneous emission can only be understood if one quantizes the electromagnetic field. From a quantum theory point of view, the reference state in the calculation of the leading-order polarizability is given by the atom in the reference ground state and n_L photons in the laser field. The virtual states for the calculation of the polarizability involve the atom in a virtual excited state, and $n_L \pm 1$ photons in the laser field. The dipole coupling leads to the creation or annihilation of a virtual laser photon.

Let us try to calculate the imaginary part of the polarizability on the basis of an energy shift calculation. In many cases, the imaginary part of an energy shift describes a decay process, with the spontaneous emission of radiation quanta. If one includes the energy of the emitted quanta into the energy balance, one sees that the final quantum state of the decay process actually has the same same energy as the initial state. We consider as an example the spontaneous emission of a photon by an atom. The initial quantum state has the atom in an excited state, and zero photons in the radiation field. In the final state of the process, the atom is in the ground state, and we have one photon in the radiation field, with an energy that exactly compensates the energy loss of the atom. The imaginary part of the self-energy of an excited state describes the decay width of the excited reference state, against one-photon decay [11]. This consideration has recently been generalized to the two-photon self-energy [12], which helped clarify the role of cascade contributions which need to be separated from the coherent two-photon correction to the decay rate [13].

How can this intuitive picture be applied to the imaginary part of the polarizability? From a quantum theory point of view [1], the initial state/reference state in the calculation of the polarizability involves the atom in the ground state, and n_L photons in the radiation field. The laser photons are not in resonance with any atomic transition, but rather, in a typical calculation, of very low frequency. One may ask what is the energetically degenerate state to which the reference state could "decay". In the case of the ground-state atomic polarizability, one may additionally point out that the atom already is in the ground state and cannot go energetically lower. How could the "decay rate" be formulated under these conditions? The answer is that a quantum state with the atom in the ground state, $n_L - 1$ laser photons of energy ω_L,

and one photon of wave vector $\boldsymbol{k}$ and polarization λ, with energy $\omega_{\boldsymbol{k}\lambda} = \omega_L$ (but not the same polarization or propagation direction) is energetically degenerate with respect to the reference state (with the atom still in the ground state, and n_L laser photons). The reference state in our calculation will thus be the product state of atom in the reference state (in general the ground state) $|\phi\rangle$, the laser field in the state with n_L photons, and zero photons in other modes of the quantized electromagnetic field. We denote this state as

$$|\phi_0\rangle = |\phi, n_L, 0\rangle\,. \quad (6)$$

The calculation of the energy shift then proceeds in analogy to the self-energy calculation: One inserts radiative loops into the diagrams that describe the ac Stark shift and calculates the decay to the state

$$|\phi_f\rangle = |\phi, n_L - 1, 1_{\boldsymbol{k}\,\lambda}\rangle\,. \quad (7)$$

The notation adopted here involves the occupation numbers of the laser mode (subscript L), and the mode with wave vector $\boldsymbol{k}$ and polarization λ.

The remainder of this paper is organized as follows. In Sec. 2, we discuss the derivation of the imaginary part of the polarizability in the length gauge, where the interaction of the atom with the laser field, and also the interaction of the atom with the quantized radiation field, are modeled on the basis of the dipole interaction $-e\,\boldsymbol{r}\cdot\boldsymbol{E}$. Gauge invariance (see Sec. 3) is used as a method to verify our results. In the velocity gauge, we use the "dipole" coupling $-e\,\boldsymbol{p}\cdot\boldsymbol{A}/m$ and the "seagull term" $e^2\boldsymbol{A}^2/(2m)$. Here, $\boldsymbol{A}$ is the vector potential, while $\boldsymbol{E} = -\partial_t\boldsymbol{A}$ is the electric field. We employ the dipole approximation throughout the paper and work in SI mksA units (with the exception of a few illustrative unit conversions in Sec. 4).

2 Length Gauge

We start with the length-gauge calculation. The laser is z polarized, and we consider the electric dipole interaction which is the dominant interaction for an atom. The electric field operators for the laser field (subscript L) and for the quantized field interaction (subscript I) read as follows,

$$\boldsymbol{E}_L = \hat{e}_z \sqrt{\frac{\hbar\omega_L}{2\epsilon_0\mathcal{V}_L}}\left(a_L + a_L^+\right) = \hat{e}_z\, E_L\,, \quad (8a)$$

$$\boldsymbol{E}_I = \sum_{\boldsymbol{k}\,\lambda} \sqrt{\frac{\hbar\omega_{\boldsymbol{k}\,\lambda}}{2\epsilon_0\mathcal{V}}}\, \hat{\epsilon}_{\boldsymbol{k}\,\lambda}\left(a_{\boldsymbol{k}\,\lambda} + a_{\boldsymbol{k}\,\lambda}^+\right)\,, \quad (8b)$$

$$H_L = -e\,z\,E_L\,, \qquad H_I = -e\,\boldsymbol{r}\cdot\boldsymbol{E}_I\,. \quad (8c)$$

We employ the dipole approximation, setting the spatial phase factors $\exp(\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r})$ equal to unity. The use of an explicit normalization volume $\mathcal{V}_L$ for the laser mode (and $\mathcal{V}$ for the modes of the quantized field) allows for a consistent normalization of the laser intensity [see Eq. (11) below]. The sum over the modes of the vacuum in H_I explicitly excludes the laser mode, in the sense of the requirement

$\boldsymbol{k}\,\lambda \neq L$. However, the absence of the one mode does not enter the matching condition

$$\sum_{\boldsymbol{k}} = \mathcal{V} \int \frac{\mathrm{d}^3 k}{(2\pi)^3}\,, \tag{9}$$

because the one highly occupied laser mode does not feature prominently in the sum over all modes, and the sum over polarizations can be carried out using the relation

$$\sum_{\lambda} \hat{\epsilon}_{\boldsymbol{k}\,\lambda}^i\, \hat{\epsilon}_{\boldsymbol{k}\,\lambda}^j = \delta^{ij} - \frac{k^i\, k^j}{\boldsymbol{k}^2}\,. \tag{10}$$

The quantity $\mathcal{V}_L$ is the normalization volume for the laser field, whereas $\mathcal{V}$ is the corresponding term for the quantized field mode. The laser field intensity is

$$I_L = \frac{n_L \hbar \omega_L\, c}{\mathcal{V}_L}\,. \tag{11}$$

The unperturbed Hamiltonian is given as follows,

$$H_0 = H_A + H_{EM}\,, \tag{12a}$$

$$H_A = \sum_m E_m\, |\phi_m\rangle \langle \phi_m|\,, \tag{12b}$$

$$H_{EM} = \sum_{\boldsymbol{k}\,\lambda \neq L} \hbar \omega_{\boldsymbol{k}\,\lambda} a_{\boldsymbol{k}\,\lambda}^+\, a_{\boldsymbol{k}\,\lambda} + \hbar \omega_L\, a_L^+\, a_L\,. \tag{12c}$$

Here, the subscript A stands for atom, while the subscript EM denotes the (quantized) electromagnetic field. The unperturbed state is given by

$$|\phi_0\rangle = |\phi, n_L, 0\rangle\,, \qquad H_A\, |\phi\rangle = E\, |\phi\rangle\,. \tag{13}$$

denoting the atom in the reference state $|\phi\rangle$ (notably, the ground state), n_L photons in the laser mode, and the vacuum state for the remaining modes of the electromagnetic field. We should add that the Fock state of the laser field is just a calculational device for our derivation of the imaginary part of the polarizability. (Of course, in a Fock state, the occupation number is precisely n_L, which is in contrast a coherent state which is a superposition of Fock states.)

The atomic Hamiltonian (Schrödinger Hamiltonian) is denoted as H_A, and we do all calculations for hydrogen, here, whereas for many-electron atoms, in the nonrelativistic approximation, one would replace $z \to \sum z_a$, where a denotes the ath electron. All results below will be reported for atomic hydrogen with a single electron coordinate, but the results hold more generally, with the only necessary modification for a many-electron atom being the addition of the coordinates of the other electrons. The reference state fulfills the relations

$$H_0\, |\phi_0\rangle = E_0\, |\phi_0\rangle\,, \qquad E_0 = E + n_L \hbar \omega_L\,, \tag{14}$$

where n_L is the number of laser photons.

In the quantized formalism [1], the second-order energy shift, which we use for normalization, can be calculated as

$$\delta E^{(2)} = -\left\langle H_L\, \frac{1}{(H_0 - E_0)'}\, H_L \right\rangle\,, \tag{15}$$

where the prime denotes the reduced Green function. (Replacement of H_L by H_I leads to the low-energy part of the Lamb shift, see Ref. [14]. Here, the Lamb shift is absorbed into a renormalized reference state energy E.) In the matrix elements used in this paper, the reference state is the full quantized-field reference state $|\phi_0\rangle$ unless indicated differently. After tracing out the photon degrees of freedom, one obtains

$$\delta E^{(2)} = -e^2 \frac{n_L \hbar \omega_L}{2\epsilon_0 \mathcal{V}_L} \left(\left\langle \phi \left| z \left(\frac{1}{H_A - E + \hbar \omega_L} \right) z \right. \right. \right.$$
$$\left. \left. \left. + z \left(\frac{1}{H_A - E - \hbar \omega_L} \right) z \right| \phi \right\rangle \right)\,, \tag{16}$$

which is a matrix element that involves only the atomic Green function, and is evaluated on the atomic reference state $|\phi\rangle$. With the identification (11) of the laser intensity, the second-order ac Stark shift can be expressed in terms of the dipole polarizability $\alpha(\omega)$, which we write as

$$\delta E^{(2)} = -\frac{I_L}{2\epsilon_0 c}\, \alpha(\omega_L)\,, \tag{17a}$$

$$\alpha(\omega) = \frac{e^2}{3} \sum_{\pm} \left\langle \phi \left| x^i \left(\frac{1}{H_A - E \pm \hbar \omega_L} \right) x^i \right| \phi \right\rangle\,, \tag{17b}$$

for a radially symmetric atomic reference state $|\phi\rangle$ which, in general, is the ground state. The x^i are the Cartesian components of the electron coordinate (the summation convention is used for i), while the sum over the two terms with both signs $\pm \omega$ will be encountered several times in the following; it is denoted by the symbol $\sum_{\pm}$ here.

The polarizability of an atom can be written as the sum of "positive-frequency" and "negative-frequency" components $\alpha_+(\omega_L)$ and $\alpha_-(\omega_L)$, according to the formulas

$$\alpha(\omega_L) = \alpha_+(\omega_L) + \alpha_-(\omega_L)\,, \tag{18a}$$

$$\alpha_+(\omega_L) = \frac{e^2}{3} \left\langle \phi \left| x^i \left(\frac{1}{H_A - E + \hbar \omega_L + \mathrm{i}\epsilon} \right) x^i \right| \phi \right\rangle\,, \tag{18b}$$

$$\alpha_-(\omega_L) = \frac{e^2}{3} \left\langle \phi \left| x^i \left(\frac{1}{H_A - E - \hbar \omega_L - \mathrm{i}\epsilon} \right) x^i \right| \phi \right\rangle\,, \tag{18c}$$

where the infinitesimal imaginary parts are introduced in accordance with the paradigm that the poles of the polarizability, as a function of ω_L, have to be located in the lower half of the complex plane. This is consistent with the fact that the polarizability corresponds to a causal (retarded) Green function. We take note of the identity

$$\frac{1}{x - \mathrm{i}\epsilon} = (\mathrm{P.V.}) \frac{1}{x} + \mathrm{i}\,\pi\, \delta(x)\,. \tag{18d}$$

We the help of this relation, one easily shows that the resonant, tree-level imaginary part of the polarizability

$\mathrm{Im}\,[\alpha_R(\omega_L)]$ is an odd function of the driving frequency ω_L,

$$
\begin{aligned}
\mathrm{Im}\,[\alpha_R(\omega_L)] = \frac{\pi\,e^2}{3}\sum_{n,i}\Big(&\left|\langle\phi\,|x^i|\,\phi_n\rangle\right|^2\,\delta(E_n - E - \hbar\omega_L)\\
&- \sum_{n,i}\left|\langle\phi\,|x^i|\,\phi_n\rangle\right|^2\,\delta(E_n - E + \hbar\omega_L)\Big)\\
= \frac{\pi}{2}\sum_n &\frac{f_{n0}\,\delta(E_n - E - \hbar\omega_L)}{E_n - E} - (\omega_L \leftrightarrow -\omega_L)\,,
\end{aligned}
$$
(18e)

where $E_n - E$ is the resonant frequency for the excitation of the ground state to the excited virtual state $|\phi_n\rangle$ of energy E_n, and we have implicitly defined the dipole oscillator strength f_{n0} according to Ref. [15]. The second-order contribution to the polarizability $\mathrm{Im}\,[\alpha_R(\omega_L)]$ is in itself gauge invariant, and it constitutes a sum of resonant peaks, which are relevant as the laser frequency is tuned across an atomic resonance. Note that the contribution $\mathrm{Im}\,[\alpha_R(\omega_L)]$ corresponds to the cutting of the following Feynman diagram,
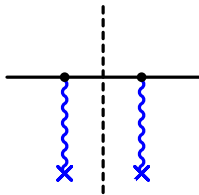
according to the Cutkosky rules [16]. Wiggly lines denote photons (here, the interactions with the photons of the laser field).

However, there is an additional contribution to the imaginary part of the polarizability, created by the insertion of a virtual photon into the second-order diagram, as shown in Fig. 1. The terms with paired interactions with the laser and the radiation field, in the fourth order, are given by

$$
\begin{aligned}
\delta E^{(4)} = &- \langle H_I\,G'(E_0)\,H_L\,G'(E_0)\,H_L\,G'(E_0)\,H_I\rangle\\
&- \langle H_L\,G'(E_0)\,H_I\,G'(E_0)\,H_I\,G'(E_0)\,H_L\rangle\\
&- 2\,\langle H_I\,G'(E_0)\,H_L\,G'(E_0)\,H_I\,G'(E_0)\,H_L\rangle\,,
\end{aligned}
$$
(19)

where $G'(E_0) = [1/(H_0 - E_0)']$ is the reduced Green function. Written in this form, the terms correspond to the entries in Fig. 1, (a) and (b), and the last term to the sum of (c) and (d), while the photon loop involves a photon in the mode $\boldsymbol{k}\,\lambda$. The propagator denominators in Fig. 1(a) in the "outer" legs of the electron line read as $H - E + \omega_{\boldsymbol{k}\,\lambda}$ (twice) because the spontaneously emitted photon is part of the virtual state of atom+field. By contrast, the propagator denominators in the "outer" lines in Fig. 1(b) read as $H - E - \omega_L$ (twice) because the spontaneously emitted photon is not present. In all cases, one photon is "taken from" the laser field. For Figs. 1(c) and (d), we have a mixed configuration with two propagators, with denominators $(H - E + \omega)$ and $(H - E - \omega)$.
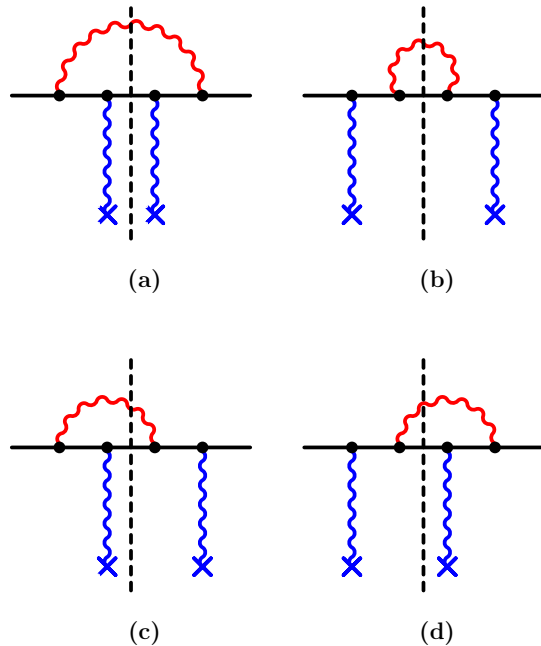
Fig. 1. (Color online.) Feynman diagrams contributing to the imaginary part of the polarizability in length gauge. Wiggly lines denote photons, straight lines denote the bound electron. A laser photon (interactions with the external laser field are denoted with a cross) is absorbed from the laser field, while the self-energy loop describes the self-interaction of the atomic electrons. When the virtual state denoted by the internal line becomes resonant with the reference state, a pole is generated in the integration over the degrees of freedom of the virtual photon (Cutkosky rules [16], indicated by the vertical dashed lines).

In the spirit of Ref. [17], the diagram (a) in Fig. 1 entails the following energy shift,

$$
\begin{aligned}
\delta E_a = -e^4\sum_{\boldsymbol{k}\lambda}&\frac{\hbar\omega_L}{2\epsilon_0\mathcal{V}_L}\frac{\hbar\omega_{\boldsymbol{k}\,\lambda}}{2\epsilon_0\mathcal{V}}\,\big\langle\phi_0\,\big|(\hat{\epsilon}_{\boldsymbol{k}\,\lambda}\cdot\boldsymbol{r})\,(a_{\boldsymbol{k}\,\lambda}^+ + a_{\boldsymbol{k}\,\lambda})\\
&\times\frac{1}{H_0 - E_0}\,z\,(a_L^+ + a_L)\,\frac{1}{H_0 - E_0}\,z\,(a_L^+ + a_L)\\
&\times\frac{1}{H_0 - E_0}\,(\hat{\epsilon}_{\boldsymbol{k}\,\lambda}\cdot\boldsymbol{r})\,(a_{\boldsymbol{k}\,\lambda}^+ + a_{\boldsymbol{k}\,\lambda})\,\big|\,\phi_0\big\rangle\,.
\end{aligned}
$$
(20)

We now use the matching condition (9) and calculate the sum over polarizations using Eq. (10). Isolating the terms which describe the absorption of a photon from the laser field and emission into the field mode $\boldsymbol{k}\,\lambda$, we obtain the following expression,

$$
\begin{aligned}
\delta E_a \sim -e^4\int&\frac{\mathrm{d}^3k}{(2\pi)^3}\,\frac{n_L\hbar\omega_L}{2\epsilon_0\mathcal{V}_L}\,\frac{\hbar\omega_{\boldsymbol{k}\,\lambda}}{2\epsilon_0}\left(\delta^{ij} - \frac{k^i\,k^j}{\boldsymbol{k}^{\,2}}\right)\\
&\times\Big\langle\phi\,\Big|z\,\frac{1}{H_A - E + \hbar\omega_{\boldsymbol{k}\,\lambda}}\,x^i\,\frac{1}{H_A - E - \hbar\omega_L + \hbar\omega_{\boldsymbol{k}\,\lambda}}\\
&\times x^j\,\frac{1}{H_A - E + \hbar\omega_{\boldsymbol{k}\,\lambda}}\,z\,\Big|\,\phi\Big\rangle\,.
\end{aligned}
$$
(21)

The $\sim$ sign implies that the terms which involve the virtual state $|\phi, n_L - 1, 1_{\boldsymbol{k}\lambda}\rangle$ still need to be isolated. We have traced out the photon degrees of freedom. We can isolate the imaginary contribution created by the virtual state $|\phi, n_L - 1, 1_{\boldsymbol{k}\lambda}\rangle$, with $\omega_{\boldsymbol{k}\lambda} = \omega_L$, as follows,

$$\frac{1}{H_A - E - \hbar\omega_L + \hbar\omega_{\boldsymbol{k}\,\lambda}}$$
$$\rightarrow \frac{1}{H_A - E - \hbar\omega_L + \hbar\omega_{\boldsymbol{k}\,\lambda} - \mathrm{i}\epsilon}$$
$$\rightarrow \frac{|\phi\rangle\langle\phi|}{-\hbar\omega_L + \hbar\omega_{\boldsymbol{k}\,\lambda} - \mathrm{i}\epsilon} \rightarrow \frac{\mathrm{i}\,\pi}{\hbar}\,\delta(\omega_{\boldsymbol{k}\,\lambda} - \omega_L)\,|\phi\rangle\langle\phi|\,. \quad (22)$$

Here, an infinitesimal imaginary part has been introduced according to the Feynman prescription for the positive-energy, virtual atomic states. Keeping track of the prefactors, one finally obtains

$$\mathrm{i}\,\mathrm{Im}(\delta E_a) = -\mathrm{i}\frac{I_L}{2\epsilon_0 c}\left\{\frac{\omega_L^3}{6\pi\epsilon_0 c^3}\right. \qquad (23)$$
$$\left. \times \left(\frac{e^2}{3}\left\langle\phi\left|x^i\,\frac{1}{H_A - E + \hbar\omega_L}\,x^i\right|\phi\right\rangle\right)^2\right\},$$

where the Einstein summation convention is used for repeated indices. The term in curly brackets, upon comparison with Eq. (17), can be identified as the imaginary part of the polarizability associated with diagram (a) of Fig. 1. The imaginary part $\mathrm{Im}(\delta E_a)$ of the energy shift associated with Fig. 1(a) is negative, as it should be for a decay process, while the imaginary part of the polarizability is positive [see the overall minus sign in Eq. (17)].

Diagram (b) in Fig. 1 generates an imaginary part in much the same way, but the "outer" virtual states have one virtual laser photon less than the intermediate state which generates the imaginary part, hence

$$\mathrm{i}\,\mathrm{Im}(\delta E_b) = -\mathrm{i}\frac{I_L}{2\epsilon_0 c}\left\{\frac{\omega_L^3}{6\pi\epsilon_0 c^3}\right. \qquad (24)$$
$$\left. \times \left(\frac{e^2}{3}\left\langle\phi\left|x^i\,\frac{1}{H_A - E - \hbar\omega_L}\,x^i\right|\phi\right\rangle\right)^2\right\}.$$

Finally, the sum of diagrams (c) and (d) generates the following expression,

$$\mathrm{i}\,\mathrm{Im}(\delta E_{c+d}) = -\mathrm{i}\frac{I_L}{2\epsilon_0 c}\left\{\frac{e^4\,\omega_L^3}{27\pi\epsilon_0 c^3}\right.$$
$$\times \left\langle\phi\left|x^i\,\frac{1}{H_A - E + \hbar\omega_L}\,x^i\right|\phi\right\rangle$$
$$\left. \times \left\langle\phi\left|x^j\,\frac{1}{H - E - \hbar\omega_L}\,x^j\right|\phi\right\rangle\right\}. \quad (25)$$

The end result for the imaginary part of the polarizability, generated by the fourth-order diagrams in Fig. 1, reads as

$$\mathrm{i}\,\mathrm{Im}(\delta E^{(4)}) = \mathrm{i}\,\mathrm{Im}(\delta E_{a+b+c+d}) = -\mathrm{i}\frac{I_L}{2\epsilon_0 c}\left\{\frac{\omega_L^3}{6\pi\epsilon_0 c^3}\right.$$
$$\times \left(\frac{e^2}{3}\left\langle\phi\left|x^i\,\frac{1}{H_A - E + \hbar\omega_L}\,x^i\right|\phi\right\rangle\right.$$
$$\left.\left. + \frac{e^2}{3}\left\langle\phi\left|x^j\,\frac{1}{H_A - E - \hbar\omega_L}\,x^j\right|\phi\right\rangle\right)^2\right\}. \tag{26}$$

After matching with Eq. (17), it can be summarized in the following, compact result,

$$\mathrm{Im}[\alpha(\omega_L)] = \mathrm{Im}[\alpha_R(\omega_L)] + \frac{\omega_L^3}{6\pi\epsilon_0 c^3}\,[\alpha(\omega_L)]^2\,, \qquad (27a)$$

where $\mathrm{Im}[\alpha_R(\omega_L)]$ is the sum of the resonance peaks, according to Eq. (18e), and in the last term, the square of the polarizability is calculated using propagators without any added "damping terms" for the virtual state energies, i.e., under the identification $[\alpha(\omega_L)]^2 \equiv \mathrm{Re}[\alpha(\omega_L)]^2$ [see Eq. (26)]. The result (27) is manifestly odd in the argument ω_L, as it should be [see Eq. (1)].

Let us dwell on the precise form of this result a little more. As shown below in Eq. (48), the second term on the right-hand side of Eq. (27a) is a correction of relative order α_{QED}^3, where α_{QED} is the fine-structure constant. Furthermore, the contribution given in Eq. (18e) can be identified, in terms of a quantum electrodynamic formalism, as the "tree-level" contribution to the imaginary part, or, as the imaginary part of the "tree-level" polarizability $\alpha_{\mathrm{TL}}(\omega)$ itself. If we absorb in our definition of $\alpha_{\mathrm{TL}}(\omega)$ relativistic corrections to the dipole polarizability of relative order α_{QED}^2 (see Ref. [18]) and all radiative corrections without cuts in the self-energy photon lines (of relative order α_{QED}^3, see Refs. [16, 19–21]), then we can write Eq. (27a) alternatively as

$$\frac{\mathrm{Im}[\alpha(\omega)]}{\alpha(\omega)} = \frac{\mathrm{Im}[\alpha_{\mathrm{TL}}(\omega)]}{\alpha_{\mathrm{TL}}(\omega)} + \frac{\omega^3}{6\pi\epsilon_0 c^3}\,\alpha_{\mathrm{TL}}(\omega) + \mathcal{O}(\alpha_{\mathrm{QED}}^4)\,. \tag{27b}$$

We can "sum" the second term into a denominator,

$$\mathrm{Im}[\alpha(\omega)] \approx \mathrm{Im}\left[\frac{\alpha_{\mathrm{TL}}(\omega)}{1 - \mathrm{i}\,\dfrac{\omega^3}{6\pi\epsilon_0 c^3}\,\alpha_{\mathrm{TL}}(\omega)}\right]. \qquad (27c)$$

The latter functional form is in agreement with various results for polarizabilities of nanoparticles (not atoms) and other structures, corrected for "radiation damping" [see Eq. (5) of Ref. [22], Eq. (27) of Ref. [23], Eq. (5) of Ref. [24], Eq. (4) of Ref. [25], and Eq. (34) of Ref. [26]]. The term from the denominator in Eq. (27c) has the required functional from for "radiation damping" due to the Abraham–Lorentz radiation reaction force [see Eq. (1) and (10) of Ref. [27], and Refs. [28, 29]]. The tree-level

term is identified with the "damping by absorption" of the nanoparticle,

$$\text{Im}[\alpha_{\text{TL}}(\omega)] \approx \text{Im}[\alpha(\omega)] - \frac{\omega^3}{6\pi\epsilon_0 c^3} \left[\alpha_{\text{TL}}(\omega)\right]^2 . \quad (28)$$

or, as the "non-radiative" imaginary part in the sense of Ref. [22]. Note that the "tree-level" for a nanoparticle term is denoted as $\alpha_0(\omega)$ in Ref. [22], which however, for an atom, would be associated with the "monopole" polarizability which is vanishing (canonically, one denotes by $\alpha_\ell(\omega)$ the 2^ℓ-pole polarizability of an atom, see Ref. [30]).

A possible interconnection of the imaginary part of the polarizability with the square of the polarizability has previously been formulated in Refs. [29] and [31]. A term with the square of the polarizability is added to the imaginary part of the polarizability in Eqs. (G2) and (G3) of Ref. [29] [see also Eq. (49) of Ref. [28]. Within QED, the term with the square of the polarizability is identified as being due to a one-loop correction, while the resonant term is added to the results given in Refs. [28, 29, 31] and constitutes a tree-level contribution.

3 Velocity Gauge

The question of the choice of gauge in electrodynamic interactions has been raised over a number of decades [32], with a particularly interesting analysis being presented in Ref. [33]. In general, and in accordance with the now famous remark by Lamb on p. 268 of Ref. [34], the wave function of a bound state therefore preserves its probabilistic interpretation only in the length gauge, and it is this gauge which should be used for off-resonance excitation processes [33, 35]. According to Table 1 of of Ref. [36], the dipole matrix elements for dipole transitions from state $|i\rangle$ to state $|f\rangle$ in the dipole approximation are related by the

$$\left\langle f \left| \frac{e}{m} \boldsymbol{p} \cdot \boldsymbol{A} \right| i \right\rangle = \text{i} \frac{E_f - E_i}{\hbar\omega} \left\langle f \left| e\boldsymbol{r} \cdot \boldsymbol{E} \right| i \right\rangle , \quad (29)$$

as can be shown with the help of the commutation relation $\boldsymbol{p} = \frac{\text{i}m}{\hbar}[H_A, \boldsymbol{r}]$. The two forms are equivalent only at resonance $\hbar\omega = E_f - E_i$. In quantum dynamic calculations, which often involve nonresonant driving of atomic transitions, according to Lamb's remark of p. 268 of Ref. [34], "a closer examination shows that the usual interpretation of probability amplitudes is valid only in the former [the length] gauge, and no additional factor $[(E_f - E_i)/(\hbar\omega)]$ occurs." Namely, the momentum operator $\boldsymbol{p} = -\text{i}\hbar\boldsymbol{\nabla}$ describes the mechanical momentum of the matter wave in the length gauge, where the kinetic and canonical momentum operators assume the same form [14, 33]. In the velocity gauge, the canonical momentum $\boldsymbol{p} - e\boldsymbol{A}$ assumes the role of the conjugate momentum of the position operator in the equations of motion. One can also argue that the electric field used in the length-gauge interaction is gauge invariant, while the vector potential in the velocity-gauge term is not [14, 33, 37–40]. However, for processes such as

energy shifts, where the initial and final states have the same energy and the sum of the exchanged photon energies need to add up to zero, both gauges should lead to the same results (see Sec. 3.3 of Ref. [14], and Refs. [41–44]). While limits of gauge invariance have recently been discussed in Ref. [45], it is a nontrivial exercise to check the gauge invariance of the imaginary part of the polarizability. Similar questions have been discussed in the context of the QED radiative correction to laser-dressed states (Mollow spectrum, see Refs. [14, 38–40]).

In the velocity gauge, the interactions are formulated in terms of the vector potentials $\boldsymbol{A}_L$ and $\boldsymbol{A}_I$, which denote the laser field and the interaction with the quantized radiation field. We denote the corresponding interaction Hamiltonians, in the dipole approximation, by the lower-case letters h_L and h_I and write

$$\boldsymbol{A}_L = \hat{e}_z \sqrt{\frac{\hbar}{2\omega_L\epsilon_0\mathcal{V}_L}} \left(\text{i}a_L - \text{i}a_L^+\right) = \hat{e}_z A_L , \quad (30\text{a})$$

$$\boldsymbol{A}_I = \sum_{\boldsymbol{k}\lambda} \hat{\epsilon}_{\boldsymbol{k}\lambda} \sqrt{\frac{\hbar}{2\omega_{\boldsymbol{k}\lambda}\epsilon_0\mathcal{V}}} \left(\text{i}a_{\boldsymbol{k}\lambda} - \text{i}a_{\boldsymbol{k}\lambda}^+\right) \quad (30\text{b})$$

$$h_L = -\frac{e A_L p_z}{m} , \qquad h_I = -\frac{e \boldsymbol{A}_I \cdot \boldsymbol{p}}{m} , \quad (30\text{c})$$

From the seagull term, we isolate the terms which involve at least one interaction with the laser mode and write

$$h_S = e^2 \frac{(\boldsymbol{A}_L + \boldsymbol{A}_I)^2}{2m} \to h_{LI} + h_{LL} ,$$

$$h_{LI} = e^2 \frac{\boldsymbol{A}_L \cdot \boldsymbol{A}_I}{m} , \qquad h_{LL} = e^2 \frac{A_L^2}{2m} . \quad (31)$$

In the length gauge, the second-order shift in the laser field is given by Eq. (17). In the velocity gauge, one obtains an additional term, as follows,

$$\delta E^{(2)} = \langle h_{LL} \rangle - \left\langle h_L \frac{1}{H_0 - E_0} h_L \right\rangle . \quad (32)$$

We trace out the laser photon degrees of freedom and obtain

$$\delta E^{(2)} = -e^2 \frac{n_L\hbar\omega_L}{2\epsilon_0\mathcal{V}_L} \left(\frac{1}{\omega_L^2} \left\langle \phi \left| \frac{p_z}{m} \left(\frac{1}{H_A - E + \hbar\omega_L} \right) \frac{p_z}{m} \right. \right. $$
$$\left. \left. + \frac{p_z}{m} \left(\frac{1}{H_A - E - \hbar\omega_L} \right) \frac{p_z}{m} \right| \phi \right\rangle - \frac{1}{m\,\omega_L^2} \right) , \quad (33)$$
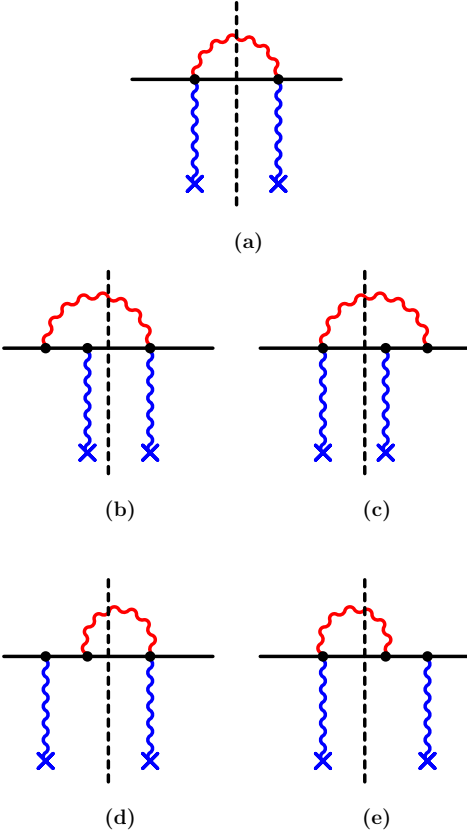
where p_z is the z component of the momentum operator. One might conclude that this form is manifestly different from Eq. (17). Using the operator identity

$$\frac{p^i}{m} = \frac{\text{i}}{\hbar} \left[H - E + \hbar\omega_L, r^i\right] , \quad (34)$$

one may show the relation

$$\frac{1}{3} \left\langle \phi \left| \frac{p^i}{m} \frac{1}{H - E + \hbar\omega_L} \frac{p^i}{m} \right| \phi \right\rangle = \frac{1}{2m} \quad (35)$$

$$- \frac{\omega_L}{3\hbar} \left\langle \phi \left| \boldsymbol{r}^2 \right| \phi \right\rangle + \frac{\omega_L^2}{3} \left\langle \phi \left| x^i \frac{1}{H - E + \hbar\omega_L} x^i \right| \phi \right\rangle ,$$

Fig. 2. (Color online.) Additional Feynman diagrams involving the seagull graph which have to be considered in the velocity gauge, in order to show the gauge invariance of the imaginary part of the polarizability.

where $\boldsymbol{r}^2 = x^i x^i$. With the help of this relation, and assuming a spherically symmetric atomic reference state $|\phi\rangle$, one may confirm that the second-order shifts (33) and (17) are equal.

In the additional diagrams which persist in the velocity gauge (see Fig. 2), the seagull graph enters "in disguise", with the virtual photon of the self-energy loop and the laser photon emerging from the same vertex. We may anticipate that diagram (a) in Fig. 2 generates a constant term, while diagrams (b) and (c) generate terms with one propagator, with a propagator denominator of the form $H - E + \omega_L$ (the spontaneously emitted photon is present in the "outer" lines). By contrast, the diagrams in Fig. 2 (c) and (d) generate propagator denominators $H - E - \omega_L$ for the internal line of the diagram.

From the diagram in Fig. 2(a), we have the energy shift

$$\delta\mathcal{E}_a = -\left\langle \phi_0 \left| h_{LI} \frac{1}{H_0 - E_0} h_{LI} \right| \phi_0 \right\rangle . \quad (36)$$

After tracing out the photon degrees of freedom, the imaginary part can be extracted as follows,

$$\mathrm{Im}(\delta\mathcal{E}_a) = -\mathrm{i} \frac{I_L}{2\epsilon_0 c} \frac{e^4}{6\pi\epsilon_0 c^3 m^2 \omega_L} . \quad (37)$$

From the diagram in Fig. 2(b) and (c), we have the energy shift

$$\delta\mathcal{E}_{b+c} = 2\left\langle \phi_0 \left| h_I \frac{1}{H_0 - E_0} h_L \frac{1}{H_0 - E_0} h_{LI} \right| \phi_0 \right\rangle . \quad (38)$$

The imaginary part of the energy, generated by the diagrams Fig. 2(b) and (c), can conveniently be written as follows,

$$\mathrm{Im}(\delta\mathcal{E}_{b+c}) = \mathrm{i} \frac{I_L}{2\epsilon_0 c} \frac{e^2}{3\pi\epsilon_0 c^3 m} \omega_L \quad (39)$$
$$\times \left(\frac{e^2}{3\,\omega_L^2} \left\langle \phi \left| \frac{p^i}{m} \frac{1}{H - E + \hbar\omega_L} \frac{p^i}{m} \right| \phi \right\rangle \right) .$$

Finally, from the diagrams in Fig. 2(d) and (e), we have the energy shift

$$\delta\mathcal{E}_{d+e} = 2\left\langle \phi_0 \left| h_L \frac{1}{H_0 - E_0} h_I \frac{1}{H_0 - E_0} h_{LI} \right| \phi_0 \right\rangle \quad (40)$$

where the sequence of the dipole coupling h_L and h_I is reversed in comparison to the diagrams in Fig. 2(b) and (c). The imaginary part of the energy shift is the same,

$$\mathrm{Im}(\delta\mathcal{E}_{d+e}) = \mathrm{Im}(\delta\mathcal{E}_{b+c}) . \quad (41)$$

The diagrams in Fig. 2 all involve at least one occurrence of the combined laser-quantized-field seagull term h_{LI}.

Finally, we can write the additional imaginary part due to the seagull terms, denoted by $\mathrm{Im}(\delta\mathcal{E}_2) = \mathrm{Im}(\delta\mathcal{E}_{a+b+c+d+e})$, in the velocity gauge as follows,

$$\mathrm{Im}(\delta\mathcal{E}_2) = -\mathrm{i} \frac{I_L}{2\epsilon_0 c} \frac{1}{6\pi\epsilon_0 c^3} \frac{e^4}{m^2 \omega_L}$$
$$+ \mathrm{i} \frac{I_L}{2\epsilon_0 c} \frac{e^2}{3\pi\epsilon_0 c^3 m} \omega_L$$
$$\times \left[\frac{e^2}{3\,\omega_L^2} \left(\left\langle \phi \left| \frac{p^i}{m} \frac{1}{H - E + \hbar\omega_L} \frac{p^i}{m} \right| \phi \right\rangle \right. \right.$$
$$\left. \left. + \left\langle \phi \left| \frac{p^i}{m} \frac{1}{H - E - \hbar\omega_L} \frac{p^i}{m} \right| \phi \right\rangle \right) \right]$$
$$= \mathrm{i} \frac{I_L}{2\epsilon_0 c} \frac{e^2}{6\pi\epsilon_0 c^3 m} \left[\frac{e^2}{m\,\omega_L} + 2\,\omega_L\,\alpha(\omega_L) \right] . \quad (42)$$

We have used the identity (35). To this result we have to add the contribution of the diagrams in Fig. 1, this time evaluated with the dipole Hamiltonians replaced by their velocity-gauge counterparts ($H_L \to h_L$ and $H_I \to h_I$),

$$\mathrm{i}\,\mathrm{Im}(\delta\mathcal{E}_1) = -\mathrm{i} \frac{I_L}{2\epsilon_0 c} \frac{1}{6\pi\epsilon_0 c^3} \frac{1}{\omega_L}$$
$$\times \left[\frac{e^2}{3} \sum_{\pm} \left\langle \phi \left| \frac{p^i}{m} \frac{1}{H - E \pm \hbar\omega_L} \frac{p^i}{m} \right| \phi \right\rangle \right]^2 \quad (43)$$

Again, with the help of (35), we can write this as

$$\mathrm{i}\,\mathrm{Im}(\delta\mathcal{E}_1) = -\mathrm{i} \frac{I_L}{2\epsilon_0 c} \frac{e^2}{6\pi\epsilon_0 c^3 m} \left\{ \frac{e^2}{m\,\omega_L} + 2\,\omega_L\,\alpha(\omega) \right\}$$
$$- \mathrm{i} \frac{I_L}{2\epsilon_0 c} \frac{1}{6\pi\epsilon_0 c^3} \omega_L^3 \left[\alpha(\omega_L) \right]^2 . \quad (44)$$

Finally, one obtains in the velocity gauge

$$\mathrm{Im}(\delta\mathcal{E}_1) + \mathrm{Im}(\delta\mathcal{E}_2) = -\frac{I_L}{2\epsilon_0 c}\frac{1}{6\pi\epsilon_0 c^3}\omega_L^3\,[\alpha(\omega_L)]^2\,, \quad (45)$$

confirming the result (26) once more [the oscillator strength in Eqs. (18) is in itself gauge invariant, and the resonant contribution (18e) needs to be added to the off-resonant contribution (45)].

4 Conclusions

Our main result concerns the imaginary part of the atomic polarizability, which is the sum of a resonant term $\mathrm{Im}[\alpha_R(\omega_L)]$ and of an off-resonant driving of an atom with the dynamic dipole polarizability $\alpha(\omega)$. According to Eq. (27), the result is given as follows,

$$\mathrm{Im}[\alpha(\omega_L)] = \mathrm{Im}[\alpha_R(\omega_L)] + \frac{\omega_L^3}{6\pi\epsilon_0 c^3}\,\mathrm{Re}[\alpha(\omega_L)]^2\,. \quad (46)$$

The clarification of the functional dependence of the non-resonant contribution to the imaginary part for small ω has been a matter of discussion in the past (see Chap. XXI of Ref. [46] and Refs. [9, 47]), with both a linear dependence on ω and an ω^3 dependence being discussed as candidate formulas. Initially, one would assume that the ω^3 dependence is favored in the "length gauge", whereas the linear term is obtained in the "velocity gauge" (see the discussion in Ref. [9]), but the formulation presented here in terms of the imaginary part of a (necessarily gauge-invariant) fourth-order energy shift removes the ambiguity. So, our result (46) settles the question. While the calculations have been described for a single-electron atom (hydrogen atom), the results hold more generally because one may simply add, in the calculation of the polarizability, the dipole couplings of the other electrons, according to the replacement $x^i \to \sum_a x_a^i$ where a denotes the subscript of the electron. The full inclusion of all diagrams given in Figs. 1 and 2 is crucial in confirming the gauge invariance (see Secs. 2 and 3).

Our results indicate that the imaginary part of the atomic polarizability cannot be obtained on the basis of a replacement in the propagator denominators of the polarizability function,

$$\alpha(\omega) = \frac{e^2}{3}\sum_{\pm}\frac{|\langle\phi|x^i|\phi_m\rangle|^2}{E_m - E \pm \hbar\omega}$$
$$\to \frac{e^2}{3}\sum_{\pm}\frac{|\langle\phi|x^i|\phi_m\rangle|^2}{E_m - \frac{\mathrm{i}}{2}\Gamma_m - E \pm \hbar\omega}\,. \quad (47)$$

The latter prescription cannot possibly lead to a consistent result for the imaginary part of the polarizability, no matter how one extrapolates the decay width $\Gamma_m \equiv \Gamma_m(\omega)$ off resonance. However, we may at least observe that if the Γ_m represent the one-photon decay widths of the virtual states, then the replacement (47) generates the right

order-of-magnitude for the imaginary part off resonance, in agreement with Eq. (46).

The specification of the real part of the square of the polarizability in Eq. (46) serves to maintain the relevant symmetry: Namely, $\mathrm{Im}[\alpha(\omega_L)]$ needs to be an odd function under a sign change of its argument. However, the off-resonant (one-loop) contribution to the imaginary part of the polarizability is in itself a radiative correction and should thus be suppressed by powers of the fine-structure constant α_{QED}. Hence, to leading order in α_{QED}, we can leave out the additional specification of the real part in the second term in Eq. (46). To put this statement into context, we switch to natural units with $\hbar = c = \epsilon_0 = 1$ and refer to Ref. [48] where it is shown that the imaginary part of the one-loop self-energy describes an imaginary energy contribution of order $\alpha_{\mathrm{QED}}^5\,m$, where α_{QED} is the fine-structure constant [in contrast to $\alpha(\omega)$ which denotes the polarizability]. The radiative correction is of relative order α_{QED}^3 in comparison to the Schrödinger energy; the latter is of order $\alpha_{\mathrm{QED}}^2\,m$ in natural units.

Let us apply this program to the polarizability. Taking into account that the Bohr radius is of order $1/(\alpha_{\mathrm{QED}}\,m)$ in natural units (i.e., about 137 times larger than the reduced Compton wavelength of the electron), we obtain the following order-of-magnitude estimates:

$$\alpha(\omega) \sim \frac{\alpha_{\mathrm{QED}}}{(\alpha_{\mathrm{QED}}m)^2}\frac{1}{\alpha_{\mathrm{QED}}^2 m} = \frac{1}{(\alpha_{\mathrm{QED}}m)^3}\,. \quad (48)$$

In order to perform the estimate for $\mathrm{Im}[\alpha(\omega)] \sim m^{-3}$, we need to take into account that a typical atomic driving frequency is of order $\omega_L \sim \alpha_{\mathrm{QED}}^2\,m$, and the ratio again is of the order of

$$\frac{\mathrm{Im}[\alpha(\omega)]}{\alpha(\omega)} \sim \alpha_{\mathrm{QED}}^3\,, \quad (49)$$

as it should be for a one-loop radiative correction [here, $\mathrm{Im}[\alpha(\omega)]$ is restricted to the second term in Eq. (46)].

Because of the general nature of the obtained result, it is useful to discuss the conversion to other unit systems. In natural units (n.u.), with $\hbar = c = \epsilon_0 = 1$, the result (46) simplifies to

$$\mathrm{Im}[\alpha(\omega)]\big|_{\mathrm{n.u.}} = \mathrm{Im}[\alpha_R(\omega)]\big|_{\mathrm{n.u.}} + \frac{\omega^3\big|_{\mathrm{n.u.}}}{6\pi}\,[\alpha(\omega)]^2\big|_{\mathrm{n.u.}}\,. \quad (50)$$

Using the relation $e^2 = 4\pi\alpha_{\mathrm{QED}}$, we can write the dipole polarizability, in natural units, as follows,

$$\alpha(\omega)\big|_{\mathrm{n.u.}} = \frac{4\pi\alpha_{\mathrm{QED}}}{\alpha_{\mathrm{QED}}^4}$$
$$\times\left\{\frac{1}{3}\sum_{\pm}\left\langle\phi\left|\alpha_{\mathrm{QED}}\,m\,x^j\,\frac{\alpha_{\mathrm{QED}}^2 m}{H - E \pm \omega_L}\,\alpha_{\mathrm{QED}}\,m\,x^j\right|\phi\right\rangle\right\}\,. \quad (51)$$

Here, the sum over the signs $\pm$ denotes the two terms generated by virtual absorption and emission of the laser photon, as in Eq. (17b). For an atom, all quantities in the term in curly brackets are of order unity; e.g., $\alpha_{\mathrm{QED}}\,m\,x^j =$

$x^j/a_0 = \rho^j$ where a_0 is the Bohr radius. The numerical values (reduced quantities) associated with the polarizability, in atomic units (a.u.) and natural units (n.u.), are thus related by

$$\alpha(\omega)|_{\mathrm{n.u.}} = \frac{4\pi}{\alpha_{\mathrm{QED}}^3} \ \alpha(\omega)|_{\mathrm{a.u.}} \ . \tag{52}$$

Atomic transition frequencies are measured in Hartrees in atomic units, and thus we have

$$\omega_L|_{\mathrm{n.u.}} = \alpha_{\mathrm{QED}}^2 \, m \ \omega_L|_{\mathrm{a.u.}} \ . \tag{53}$$

So, in atomic units, our main result given in Eq. (27) and (50) reads as

$$\mathrm{Im}[\alpha(\omega)]|_{\mathrm{a.u.}} = \mathrm{Im}[\alpha_R(\omega)]|_{\mathrm{a.u.}} + \frac{2\alpha_{\mathrm{QED}}^3}{3} \left[\omega^3 \, \alpha^2(\omega)\right]\big|_{\mathrm{a.u.}} \ . \tag{54}$$

The imaginary part of the polarizability enters the description of a number of dissipative processes, such as the quantum friction due to interaction with a bath of blackbody photons (4), or, with a dielectric surface [see Eq. (5)]. The imaginary part of the polarizability is manifestly non-vanishing off resonance and, for small driving frequency ω, is proportional to ω^3.

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