NON-LINEAR MAGNETO-ELECTRIC SUSCEPTIBILITIES AND LASER LIGHT INTENSITY DEPENDENT FARADAY EFFECT IN ATOMIC SYSTEMS

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Quantum-mechanical formulae for the magneto-electric susceptibility fifth-rank tensor are derived, providing the basis for first numerical calculations of contributions from these susceptibilities to non-linear variations in Faraday effect in atomic systems. The calculations predict that the laser intensity dependence of the Verdet constant is accessible to experimental measurement. In particular, for inert gases the Verdet constant can vary by as much as 40 per cent in light wave fields of $E \approx 10^7$ V/cm. For atomic hydrogen, the variation amounts to 20 per cent in a field of $E = 5 \times 10^5$ V/cm at a resonance mistuning of 30 cm^{-1}.

1. Introduction

The powerful sources of laser radiation now available permit the investigation, in addition to classical electro- and magneto-optical effects, of a variety of novel non-linear phenomena involving changes in the polarisational characteristics of radiation, propagating in non-linear media. Moreover, in the field of an intense light wave, intensity-dependent corrections to the Kerr, Faraday, Cotton-Mouton and other classical effects can become essential. The semi-macroscopic theory of the influence of intense laser fields on these effects has been developed in Refs [1–3]. Hitherto, however, no consequently quantum mechanical calculations of the relevant non-linear susceptibility tensors or their numerical values for specific non-linear media have been performed. Since the non-linear corrections are given by fifth- and higher-rank tensors and are essentially dependent on the radiation wavelength, approximate evaluations can prove rough thus making difficult the assessment of the magnitude of the effects to be expected as well as of the feasibility of their observation in experiment.

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In this paper, quantum mechanical expressions are derived for the non-linear corrections to Faraday's effect in an intense light field, and results of numerical computations of the respective quantities for atomic gases are given. The choice of the latter as non-linear media was determined by the following two circumstances: Firstly, correct quantum mechanical calculations of non-linear suceptibilities are, at present, feasible for gases only, and secondly, from the experimental viewpoint, rarefied diamagnetic atomic gases are the best adapted to accurate measurements of the purely electron non-linearities of the medium defined by the higher-order non-linear susceptibilities of the atom. The latter circumstance is due to the absence, in diamagnetic gases, of the temperature-dependent terms proportional to products of susceptibilities of lower orders which, in the case of most molecules and liquids, screen the effects due to electronic non-linearities of higher orders. The smallness of the non-linear coefficients of atoms by comparison with other non-linear media can be partly compensated by the use of highly intense incident radiation. Thus, e. g. in the case of inert gases the breakdown voltage is $E_0 \approx 5 \times 10^7$ V/cm, admitting of the use in experiments of the focussed radiation of pulse solid-state lasers.

2. Theoretical background

The variation in polarisational characteristics of radiation propagating in a medium is determined by the anisotropy of the refractive index $n(\omega)$. Such anisotropy can result from the intrinsic properties of the medium (natural activity) and can be induced by an external electromagnetic field. The change in refractive index due to the action of a field can be expressed in terms of the electric and magnetic polarization induced in the medium [2]. We shall be considering a rarefied gas, of volume V containing N atoms, isotropic in the absence of external fields. In a static magnetic field $\boldsymbol{H}$ and that of a monochromatic wave with electric vector $\boldsymbol{E}(t) = \mathrm{Re}(\boldsymbol{E}(\omega)e^{-i\omega t})$, its refractive index is determined by the component of electric dipole polarisation vector $\boldsymbol{P}(t)$ at the frequency ω which, with accuracy up to terms of order 4 in $\boldsymbol{E}$ and $\boldsymbol{H}$ inclusively, can be written in the following form[1]:

$$\boldsymbol{P}(t) = \mathrm{Re}\,(\boldsymbol{P}(\omega)e^{-i\omega t}),$$

$$\begin{aligned} P_i(\omega) = {} & \chi_{ij}(-\omega;\omega)E_j(\omega) + \chi_{ijk}(-\omega;\omega,0)E_j(\omega)H_k(0) \\ & + \chi_{ijkl}(-\omega;\omega,-\omega,\omega)E_j(\omega)E_k^*(\omega)E_l(\omega) + \chi_{ijkl}(-\omega;\omega,0,0)E_j(\omega)H_k(0)H_l(0) \\ & + \chi_{ijklm}(-\omega;\omega,-\omega,\omega,0)E_j(\omega)E_k^*(\omega)E_l(\omega)H_m(0) \\ & + \chi_{ijklm}(-\omega;\omega,0,0,0)E_j(\omega)H_k(0)H_l(0)H_m(0) + \ldots \end{aligned} \tag{1}$$

The susceptibilities χ describe various processes of interaction between the wave and the atoms, as well as Rayleigh light scattering (not affecting the spectral composition of the radiation), and changes in the polarisational parameters of the light wave propaga-

[1] We take into consideration electric dipolar interaction between the system and wave only, since in atomic gases the contribution from magnetic and higher multipolar terms is usually negligible [1, 2].

ting in the gas. In particular, the first two terms of (1) define the linear Faraday effect. The general structure of the tensors χ_{ij} and χ_{ijk} as well as numerical calculations for some atoms are given in Ref. [4]. Next, $\chi_{ijkl}(-\omega;\omega,-\omega,\omega)$ is the hypersusceptibility tensor, calculated for atomic gases and related with the constants of the optical Kerr effect and self-induced rotation of the polarisation ellipse in Refs [5, 6]. Moreover, Ref. [5] contains calculations of $\chi_{ijkl}(-\omega;\omega,0,0)$, the tensor describing the Cotton-Mouton effect. Whereas the last two terms of (1) are corrections, non-linear in E and H, to the magneto-electric susceptibility tensor χ_{ijk}. Their investigation is of interest from a double point of view. Firstly, in the presence of intense light fields (or a strong static magnetic field) they can contribute significantly to the total Faraday rotation and be well accessible to experimental observation thus providing novel information, inherent in the tensor χ_{ijklm}, on the properties of the medium. Secondly, by calculating the higher-order corrections to the physical characteristics which define the process in the first non-vanishing order of perturbation calculus (to χ_{ijk}, in the case of Faraday rotation) we are able to determine those critical field strengths E_{crit} and H_{crit} up to which perturbation theory is still valid as a description of field-system interaction. Series of the type (1), commonly applied in non-linear optics, are menaingful as long as corrections of higher-orders are small. This, in general, requires that the relationship:

$$E \ll E_{\text{at}}, \quad H \ll H_{\text{at}}$$

shall be fulfilled (E_{at} and H_{at} being characteristic intra-atomic fields, amounting in most atoms to about $\sim 10^8$ V/cm and 10^8 Gs, respectively). The correct value of the critical fields, e.g. E_{crit}, can be found from the relation:

$$|\chi_{ijk}(-\omega;\omega,0)| \approx |\chi_{ijklm}(E_{\text{crit}})_l(E_{\text{crit}})_m|. \tag{2}$$

Because of the considerable magnitude of the non-linear coefficients, E_{crit} can be much lower than E_{at}. Thus, for example, with regard to the alkali atoms perturbation calculus ceases to be applicable for light shift already at field strengths of $E \approx 5 \times 10^6$ V/cm [6], typical for pulse solid-state lasers. Hence, when studying electro- and magneto-optical effects in the fields of modern, highly powerful lasers, one has to keep in mind that traditional series expansions of the type (1) can prove inadequate and that new, non-perturbative methods for the theoretical description of the processes in question have to be developed.

We now proceed to consider the non-linear corrections in intense light fields. The action of strong magnetic fields will be the subject of our next paper.

3. *General formulae for the magneto-electric susceptibility* $\chi_{ijklm}(-\omega;\omega,-\omega,\omega,0) \equiv \chi^{\text{em}}$

To calculate the susceptibility χ^{em}, we have to extract from the dipole polarization vector

$$\boldsymbol{P}(t) = \langle \Psi_{nJM}(\boldsymbol{r},t)|\hat{\boldsymbol{d}}|\Psi_{nJM'}(\boldsymbol{r},t)\rangle \tag{3}$$

the component $\boldsymbol{P}(\omega)$ at frequency ω proportional to the field strength product E^3H. The

wave functions $\Psi(r, t)$ are solutions of the Schrödinger equation of the atom within the framework of perturbation theory with the interaction Hamiltonian $V_e + V_h$, where:

$$V_e = -(d \cdot E(t)) = -\mathrm{Re}\,((d \cdot E)e^{-i\omega t}),$$
$$V_h = -(\mu \cdot H(0)), \tag{4}$$

which, in the absence of interactions, go over into the unperturbed wave function of the atom $|nJ\rangle$ with n — the principal quantum number of the atomic level for which χ^{em} is calculated, and J — the total angular momentum. Since states with $J > 0$ are degenerate in the projection M of J on the quantization axis, the mean values of the moment d in (3) is to be calculated over functions with different M, M', corresponding to the unperturbed functions $|nJM\rangle$ and $|nJM'\rangle$. Hence $p(t)$, and consequently χ^{em}, are dependent on the indices M, M'; however, for brevity, we refrain from specifying the latter. The susceptibilities χ_{ij}, χ_{ijk} are defined similarly for degenerate states [4].

Inasmuch as V_e is periodical in t, $\Psi(r, t)$ can be expanded in a Fourier series with the components $\Psi^{(k\omega)}(r)$, $k = 0, \pm 1, \ldots$, which, in turn, are expanded in series in powers of E and H. We shall be denoting the expansion coefficients of $\Psi^{(k\omega)}$, proportional to $E^n H^m$, by $\Psi^{(k\omega)}_{ne,mh}(r)$ (obviously, $n \geqslant k$). With regard to the aforesaid, $p(\omega)$ is easily shown to be of the form:

$$\begin{aligned} P(\omega) = P^*(-\omega) = {} & \langle nJM|d|\Psi^{(\omega)}_{3e,h}\rangle + \langle \Psi^{(0)}_{0,h}|d|\Psi^{(\omega)}_{3e,0}\rangle + \langle \Psi^{(-\omega)}_{e,0}|d|\Psi^{(0)}_{2e,h}\rangle \\ & + \langle \Psi^{(0)}_{2e,0}|d|\Psi^{(\omega)}_{e,h}\rangle + \langle \Psi^{(\omega)}_{e,h}|d|\Psi^{(2\omega)}_{2e,0}\rangle + \langle \Psi^{(\omega)}_{e,0}|d|\Psi^{(2\omega)}_{2e,h}\rangle \\ & + \langle \Psi^{(-\omega)}_{3e,h}|d|nJM'\rangle + \langle \Psi^{(-\omega)}_{3e,0}|d|\Psi^{(0)}_{0,h}\rangle + \langle \Psi^{(0)}_{2e,h}|d|\Psi^{(\omega)}_{e,0}\rangle \\ & + \langle \Psi^{(-\omega)}_{e,h}|d|\Psi^{(0)}_{2e,0}\rangle + \langle \Psi^{(-2\omega)}_{2e,0}|d|\Psi^{(-\omega)}_{e,h}\rangle + \langle \Psi^{(-2\omega)}_{2e,h}|d|\Psi^{(-\omega)}_{e,0}\rangle. \end{aligned} \tag{5}$$

It should be stated clearly that the functions $\langle\ |$ and $|\ \rangle$ correspond to distinct values M and M' of the projection of the momentum J.

The functions $\Psi^{(k\omega)}_{ne,mh}(r)$ can be calculated by time-dependent perturbation theory methods as solutions of the relevant inhomogeneous differential equations (see, the review [7]). The tedious calculations involved by those methods can, however, be avoided by having recourse to the procedure of solving Schrödinger's equation in a monochromatic field developed in Refs [6, 8]. In this case, the $\Psi_{ne,mh}(r)$ are obtained formally from the expression for the $(n+m)$-th order wave function of time-independent perturbation theory. To illustrate this procedure, we shall calculate the second-order function Ψ_2 which, for the stationary perturbation $V = V_1 + V_2$, is of the well known form [9]:

$$\begin{aligned} \Psi_2(r) = {} & \sum_{p_1, p_2 \neq n} |p_2\rangle \frac{\langle p_2|V_1 + V_2|p_1\rangle \langle p_1|V_1 + V_2|n\rangle}{(E_{p_2} - E_n)(E_{p_1} - E_n)} \\ & - \langle n|V_1 + V_2|n\rangle \sum_{p \neq n} |p\rangle \frac{\langle p|V_1 + V_2|n\rangle}{E_p - E_n} \\ & - \tfrac{1}{2}|n\rangle \sum_{p \neq n} \frac{\langle n|V_1 + V_2|p\rangle \langle p|V_1 + V_2|n\rangle}{(E_p - E_n)^2}, \end{aligned} \tag{6}$$

where $(|p\rangle, E_p)$ is the complete set of eigen-functions and energies of the non-perturbed atom. If V_1 and V_2 depend harmonically on t with the frequency ω then, with regard to Refs [6, 8], $\Psi_2(r, t)$ can be obtained formally from (6) on replacement of the basis $(|p\rangle, E_p)$ by the new set of "unperturbed" functions and energies:

$$\{|p, k\rangle, E_{p,k}\} = \{|p\rangle e^{ik\omega t}, E_p + k\omega\}, \text{ where } k = 0, \pm 1, \ldots, \tag{7}$$

and of the matrix elements $\langle P_1|V_1+V_2|P_2\rangle$ by the "averaged" matrix elements:

$$\langle\langle p_1, k_1|V_1+V_2|p_2, k_2\rangle\rangle = \frac{1}{T}\int_0^T dt\langle p_1 e^{ik_1\omega t}|V_1+V_2|p_2 e^{ik_2\omega t}\rangle. \tag{8}$$

In particular, putting $V_1 = V_e$ and $V_2 = V_h$ from (4), we get for $\Psi_2(r, t)$ with regard to Eqs (6)—(8):

$$\begin{aligned}\Psi_2(r, t) = {} & \Psi^{(0)}_{2e,0} + \Psi^{(0)}_{0,2h} + \Psi^{(\omega)}_{e,h} e^{i\omega t} + \Psi^{(-\omega)}_{e,h} e^{-i\omega t} \\ & + \Psi^{(2\omega)}_{2e,0} e^{i2\omega t} + \Psi^{(-2\omega)}_{2e,0} e^{-i2\omega t},\end{aligned} \tag{9}$$

where:

$$\Psi^{(2\omega)}_{2e,0}(r) = \sum_{p_2;p_1,k_1}{}' \frac{|p_2\rangle \langle\langle p_2, 2|(d\cdot E(t))|p_1, k_1\rangle\rangle \langle\langle p_1, k_1|(d\cdot E(t))|n, 0\rangle\rangle}{(E_{p_2,2}-E_{n,0})(E_{p_1,k_1}-E_{n,0})}$$

$$= \tfrac{1}{4} G_{E_n-2\omega}(r, r_1)(d\cdot E^*) G_{E_n-\omega}(r_1, r_2)(d\cdot E^*)|n\rangle, \tag{10}$$

$$\Psi^{(\omega)}_{e,h}(r) = \tfrac{1}{2} G_{E_n-\omega}(r, r_1)(d\cdot E^*) G_{E_n}(r_1, r_2)(\mu\cdot H)|n\rangle$$

$$+\tfrac{1}{2} G_{E_n-\omega}(r, r_1)(\mu\cdot H) G_{E_n-\omega}(r_1, r_2)(d\cdot E^*)|n\rangle, \tag{11}$$

$$\Psi^{(0)}_{2e,0}(r) = \tfrac{1}{4} G_{E_n}(r, r_1)(d\cdot E) G_{E_n-\omega}(r_1, r_2)(d\cdot E^*)|n\rangle$$

$$+\tfrac{1}{4} G_{E_n}(r, r_1)(d\cdot E^*) G_{E_n+\omega}(r_1, r_2)(d\cdot E)|n\rangle$$

$$-\tfrac{1}{8}|n\rangle\{\langle n|(d\cdot E) G_{E_n-\omega}(r_1, r_2) G_{E_n-\omega}(r_2, r_3)(d\cdot E^*)|n\rangle$$

$$+\langle n|(d\cdot E^*) G_{E_n+\omega}(r_1, r_2) G_{E_n+\omega}(r_2, r_3)(d\cdot E)|n\rangle\}, \tag{12}$$

$$\Psi^{(0)}_{0,2h}(r) = G_{E_n}(r, r_1)(\tilde{\mu}\cdot H) G_{E_n}(r_1, r_2)(\mu\cdot H)|n\rangle$$

$$-\tfrac{1}{2}|n\rangle\langle n|(\mu\cdot H) G_{E_n}(r_1, r_2) G_{E_n}(r_2, r_3)(\mu\cdot H)|n\rangle. \tag{13}$$

Above, we introduced the notation $\tilde{\mu} = \mu - \langle n|\mu|n\rangle$ whereas for summation over the virtual states in (6) we have recourse to the Green function:

$$G_E(r, r') = \sum_p \frac{|p\rangle\langle p|}{E_p - E}. \tag{14}$$

The functions $\Psi^{(-\omega)}$, $\Psi^{(-2\omega)}$ of (9) are derived from (10), (11) by changing the sign at ω and interchanging $E^* \to E$. Similarly, the other functions $\Psi_{ne,mg}$ of (5) can be derived

by applying the well known formulae of time-independent perturbation theory of $(n+m)$-th order.

The final expression for $P(\omega)$ (and hence χ^{em}) can be written in the form of a sum of terms of three types:

$$P(\omega) = \{\Pi\} + \{S\} + \{N\}. \tag{15}$$

I. $\{\Pi\}$ — these are matrix elements of the 5-th rank involving 4 Green functions G_E. For example:

$$\langle nJM| (\boldsymbol{d} \cdot \boldsymbol{E}^*) G_{E_n+\omega}(\boldsymbol{r}_1, \boldsymbol{r}_2) d G_{E_n+2\omega}(\boldsymbol{r}_2, \boldsymbol{r}_3) (\tilde{\boldsymbol{\mu}} \cdot \boldsymbol{H})$$

$$\times G_{E_n+2\omega}(\boldsymbol{r}_3, \boldsymbol{r}_4) (\boldsymbol{d} \cdot \boldsymbol{E}) G_{E_n+\omega}(\boldsymbol{r}_4, \boldsymbol{r}_5) (\boldsymbol{d} \cdot \boldsymbol{E}) |nJM'\rangle.$$

This term can be represented by a Feynman graph of the type of Fig. 1. The other terms of the type $\{\Pi\}$ can be obtained from Fig. 1 by transposition of the photon lines and vertical lines – – –, corresponding to interaction with the field H; the energies E of the Green

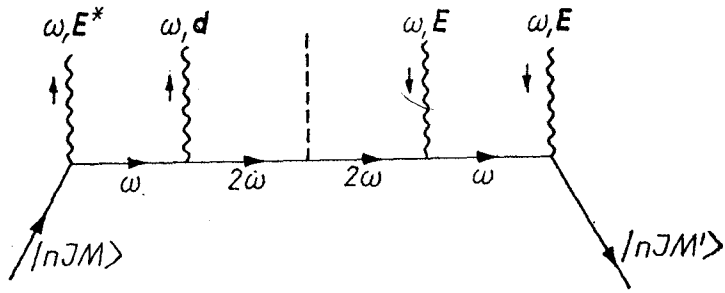

Fig. 1. The Feynman diagram for the fourth-order magneto-electric susceptibility

functions G_E being determined in conformity with the energy conservation law for each vertex. Obviously, the number of possible distinct terms is $5/2! = 60$ where 2! intervenes because 2 photon lines are identical.

II. $\{S\}$ — these are "secular" terms, occurring due to the circumstance that the corrections to the wave function in the second and higher orders of perturbation theory involve terms accounting for the change in energy of the level $|n\rangle$ in the field (cf., the review [7]). Some of these terms — those corresponding to the Zeeman effect — have been taken into account in $\{\Pi\}$ by the interchange $\mu \to \tilde{\mu}$. Moreover, $P(\omega)$ contains contributions from terms corresponding to the AC Stark effect and to the magneto-electric shift in level proportional to E^2H [4]. In the general case, $\{S\}$ comprises 36 terms. As an example, we adduce the term:

$$-\langle nJM| d G_{E_n+\omega}(\boldsymbol{r}_1, \boldsymbol{r}_2) G_{E_n+\omega}(\boldsymbol{r}_2, \boldsymbol{r}_3) (\boldsymbol{d} \cdot \boldsymbol{E}) |nJM'\rangle$$

$$\times \langle nJM'| (\boldsymbol{d} \cdot \boldsymbol{E}^*) G_{E_n+\omega}(\boldsymbol{r}_1, \boldsymbol{r}_2) (\boldsymbol{d} \cdot \boldsymbol{E}) G_{E_n}(\boldsymbol{r}_2, \boldsymbol{r}_3) (\boldsymbol{\mu} \cdot \boldsymbol{H}) |nJM'\rangle + \ldots$$

III. $\{N\}$ — these are normalization terms, accounting for the effect of wave function normalization. In particular, in (12), the third term is a normalisation term. Usually,

normalization terms do not occur in susceptibilities of lower rank (up to the fourth); however, in higher orders, they have to be taken into account.

Thus, the general formula for χ^{em} is highly complicated and so bulky that we refrain from adducing it here explicitly[2]. We note nonetheless that all the terms of χ^{em} are given by integrals of the G_E and, if a sufficiently simple analytical expression is available for G_E as that derived in Refs [10, 11], can be calculated numerically by computer. The quantum mechanical formula for χ^{em} can also be obtained following a different path, namely by expanding the well known expression for the hypersusceptibility tensor $\chi_{ijkl}(-\omega; \omega, -\omega, \omega)$ [12] in a series in H, but the result cannot be expressed directly in terms of Green function integrals of the type (15), and rather tedious intermediate transformations are required.

With regard to the third-rank susceptibility, defining Faraday's effect, the correspondence between the two forms of expression of χ_{ijk} is established in Ref. [13].

4. The non-linear correction to the Faraday effect, and numerical results

The non-linear corrections to usual Faraday rotation can be of relevance in the following two cases:

(*i*) when studying the rotation of the polarisation plane of a weak (probe) electromagnetic wave with electric vector $\boldsymbol{E}(t) = \boldsymbol{E}\cos\omega t$, propagating in the direction of the magnetic field $\boldsymbol{H}$, the gas being illuminated with intense light of frequency $\omega_L \neq \omega$. In this situation, the rotation angle becomes dependent on the laser field intensity in a way described by the susceptibility $\chi_{ijklm}(-\omega; \omega, -\omega_L, \omega_L, 0)$. This mechanism is considered in Refs [14, 15],

(*ii*) when the field $\boldsymbol{E}(t)$ itself is intense (laser light) and the nonlinearities, induced thereby in the gas, cause an additional rotation, proportional to the light intensity. Since here no probe beam is required, and the rotation effect bears on the polarisation plane of the intense laser wave, this setup presents some valuable simplifications for the experimenter. We now proceed to analyze the case (*ii*) and the relation between the non-linear correction to Verdet's constant $V(\omega)$ and the susceptibility χ^{em}, investigated in Section 3.

Applying the usual definition of the Verdet constant $V(\omega)$:

$$V(\omega) = \frac{\omega}{2c}\,\frac{n_- - n_+}{H}$$

and taking into account that in a satisfactory approximation

$$(n_\pm^2 - 1)E_\pm(\omega) = 4\pi P_\pm(\omega) = \frac{\mp 4\pi}{\sqrt{2}}\{P_x(\omega) \pm iP_y(\omega)\},$$

---

[2] For non-degenerate states, the structure of χ^{em} is somewhat simpler, inasmuch as for levels with $J = 0$ one has the equalities

$$\langle 0|(\boldsymbol{\mu}\cdot\boldsymbol{H})|0\rangle \equiv 0,$$

$$\langle 0|(\boldsymbol{\mu}\cdot\boldsymbol{H})G_E \ldots = \ldots G_E(\boldsymbol{\mu}\cdot\boldsymbol{H})|0\rangle \equiv 0$$

we obtain by (1) the following expression:

$$V(\omega) = i\frac{2\pi\omega N_0}{cn}\left[\chi_{xyz}(-\omega;\omega,0)+\chi_{xyyyz}(-\omega;\omega,-\omega,\omega,0)\,|E|^2\right] \tag{16}$$

for the setup when the field $E(t) = E\cos\omega t$ propagates in the direction of the vector H (along the z-axis). Above, N_0 is the number density of atoms in the medium, and n the refractive index (with satisfactory accuracy, equal to unity).

Eq. (16) holds only for non-degenerate states with $J = 0$, when $\chi_{xy}(-\omega;\omega) = 0$. At degeneracy $V(\omega)$ contains temperature-dependent terms which, if effects proportional to $|E|^2$ are taken into account in $V(\omega)$, become rather bulky [14]. In this paper, we shall consider but the ground states of hydrogen, the inert gases, and alkali atoms for which the temperature-dependent terms do not contribute to $V(\omega)$. (Though these terms are non-zero for alkali metals [4], their contribution is not decisive). To simplify the calculations, the multiplet structure of the excited levels can be neglected; this does not affect $V(\omega)$ far from resonances [4]. In this approximation, χ_{xyz} is proportional to the derivative $d\alpha(\omega)/d\omega$ of the dynamical polarizability in conformity with Becquerel's formula whereas χ_{xyyyz}, in conformity with the procedure of Section 3, is expressed in terms of integrals of the radial Green function of the optical electron. For the latter function, use is made of the expression derived in the approximation of the model potential method [10, 11].

In order to provide an example of the numerical results, it is convenient to re-write Eq. (16) in atomic units, as follows:

$$V(\omega) = \pi N_0\alpha^2\omega\{Y_0(\omega)+Y_1(\omega)\,|E|^2\}, \tag{17}$$

where $\alpha = 1/137$ is the fine structure constant, and $Y_0 = d\alpha(\omega)/d\omega$.

Table I shows the frequency-dependence of $Y_0(\omega)$ and $Y_1(\omega)$ for hydrogen. $Y_1(\omega)$ is found to depend on ω more strongly than $Y_0(\omega)$. In particular, Y_1 exhibits a resonance at $\omega = \dfrac{E_{3s}-E_{1s}}{2} = 48772\,\text{cm}^{-1}$ which is absent in Y_0. The subsequent resonances of Y_1 are given by the relation $\omega_n = \dfrac{E_n-E_{1s}}{2}$, with $n \geqslant 4$.

Tables II and III show Y_0 and Y_1 for the alkali atoms and inert gases at the fundamental frequencies and harmonics of pulse ruby and neodymium lasers. These results permit the prediction that the non-linear corrections to $V(\omega)$ are accessible to observation in experiment. Thus, for xenon, the non-linear effects contribute about 40% in a field of 5×10^7 V/cm. Such fields are currently in use in experimental work on the ionization of inert gases [16]. Close to resonance the role of the non-linear effects increases steeply. In fact, in the case of hydrogen at a mistuning of about 30 cm^{-1} ($\omega = 48800\,\text{cm}^{-1}$) the term with $Y_1(\omega)$ contributes a correction of about 20% in a field of but $E \approx 5\times10^5$ V/cm.

Eq. (2) moreover leads to the magnitude of the critical field E_{crit}, discussed in the Introduction. Typically, E_{crit} for inert gases lies at $\sim10^8$ V/cm, and for alkali atoms at $(5\times10^6 \div 10^7)$ V/cm. It is of interest that these values are of the same order as in the case of the non-linear corrections to the level shift discussed in Ref. [6].

TABLE I

Dispersion of the coefficients $Y_0(\omega)$ and $Y_1(\omega)$ for the ground state of the hydrogen atom

$\omega \times 10^{-3}$, cm^{-1}	$Y_0(\omega)$, at. units	$Y_1(\omega)$, at. units
9.44	0.390	28.73
14.4	0.615	49.38
18.88	0.841	75.78
28.8	1.476	203.3
40.0	15.86	6.33×10^3
41.0	16.73	7.907×10^3
42.0	17.67	1.015×10^4
43.0	18.67	1.352×10^4
44.0	19.75	1.896×10^4
45.0	20.90	2.875×10^4
46.0	22.13	4.979×10^4
47.0	23.47	1.124×10^5
47.2	23.75	1.404×10^5
47.4	24.04	1.811×10^5
47.6	24.33	2.439×10^5
47.8	24.62	3.486×10^5
48.0	24.92	5.438×10^5
48.2	25.23	9.764×10^5
48.4	25.54	2.281×10^6
48.6	25.85	1.057×10^7
48.8	26.17	4.030×10^8
49.0	26.50	6.055×10^6
49.2	26.82	1.747×10^6
49.3	26.99	1.164×10^6
49.5	27.33	6.401×10^5
49.7	27.68	4.230×10^5
49.9	28.03	3.192×10^5
50.0	28.20	2.895×10^5
50.1	28.38	2.701×10^5
50.3	28.75	2.566×10^5

TABLE II

Coefficients Y_0 and Y_1 for atoms of the alkali metals at the radiation frequencies of neodymium ($\omega_N = 9440\ \text{cm}^{-1}$) and ruby ($\omega_R = 4400\ \text{cm}^{-1}$) lasers

Atom	ω_N		ω_R	
	Y_0	Y_1	Y_0	Y_1
Li	8.5×10^3	-9.19×10^6	1.05×10^6	-4.71×10^{11}
Na	4.52×10^3	1.71×10^7	4.18×10^4	9.67×10^{10}
K	3.12×10^4	2.54×10^6	2.26×10^5	-1.46×10^{10}
Rb	3.9×10^4	8.9×10^{10}	1.8×10^5	-1.4×10^{10}
Cs	9.7×10^4	-7.5×10^9	7.2×10^4	2.6×10^{10}

TABLE III

Coefficients Y_0 and Y_1 for inert gases at the fundamental and second-harmonic radiation frequencies of the neodymium and ruby laser

Atom	ω_N		ω_R		$2\omega_N$		$2\omega_R$	
	Y_0	Y_1	Y_0	Y_1	Y_0	Y_1	Y_0	Y_1
He	0.146	1.48	0.223	2.31	0.295	3.13	0.463	5.32
Ne	0.704	12.26	1.08	19.6	1.44	27.1	2.30	50.2
Ar	3.80	~~1.97~~	5.91	329	7.95	486	13.3	1150
Kr	7.10	538	11.1	927	15.1	1432	26.1	4040
Xe	15.5	1913	24.6	3474	34.0	5780	62.0	24100

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Accordingly, when studying magneto-electric phenomena in the radiation fields of powerful lasers, non-linear effects contribute essentially to the atomic constants measured. At still higher intensities of the laser beam, they can make the interpretation of the phenomena in terms of classical non-linear susceptibilities impossible.

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