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MAGNETO-ELECTRIC SUSCEPTIBILITIES OF DEGENERATE STATES. II. NUMERICAL CALCULATION OF SUSCEPTIBILITIES, AND INTERACTION PROCESSES BETWEEN ATOMS AND ELECTROMAGNETIC FIELDS IN THE PRESENCE OF A STATIC MAGNETIC FIELD

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Numerical calculations of electric ($\chi^{e}_{ij}(-\omega;\omega)$) and magneto-electric ($\chi^{eem}_{ijk}(-\omega;\omega,0)$) susceptibilities are performed for atoms at the frequencies of ruby and neodymium lasers and their second harmonics. The following processes, described by these susceptibilities, are considered: (*i*) normal and inverse Faraday effect; (*ii*) DC magnetic field-induced Rayleigh light scattering; (*iii*) magneto-electric splitting and shift of atomic levels.

1. *Introduction*

In our previous work [1] (to be referred to as I) dealing with the general structure of the magneto-electric susceptibilities χ_{ijk}, we derived expressions for the invariant atomic parameters R_{xy}, fully defining the χ_{ijk}'s. Here, we give calculations of the parameters R_{xy} for some atoms, constituting the non-linear media most usually used in experiments on their interaction with laser radiation. In Sections 3–5, we consider a variety of magneto-electric processes conveniently described in terms of R_{xy}, thus (*i*) the normal and inverse Faraday effect, (*ii*) light scattering in the presence of a static magnetic field, and (*iii*) shift and splitting of atomic levels under the concomitant action of a magnetic field and laser radiation on the atom.

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2. *The parameters R_{xy} for atomic terms of various symmetry*

The calculation of the parameters R_{xy}, given by formula (I, 2.23), depends on the type of coupling between the angular momenta in the atom. For a well-defined state of the latter with prescribed momentum J, the reduced matrix elements $\langle nJ\|M\|n'J'\rangle$ and $\langle nJ\|\boldsymbol{d}\|n'J'\rangle$ are to be calculated in conformity with the well known formulae of angular momentum theory [2]. As a result, the R_{xy} can be expressed as combinations of radial matrix elements, containing integrals of the radial part $R_{nJ}(r)$ of the wave func-

TABLE I

The parameters R_{01} and α_0 for inert gases in the ground state

Atom	R_{01}/α, in atomic units				α_0, in atomic units			
	ω_N	ω_R	$2\omega_N$	$2\omega_R$	ω_R	ωR	$2\omega_N$	$2\omega_R$
He	0.178	0.273	0.361	0.567	1.49	1.495	1.50	1.52
Ne	0.861	1.329	1.763	2.824	5.23	5.27	5.29	5.37
Ar	4.65	7.24	9.73	16.3	13.2	13.3	13.4	13.9
Kr	8.68	13.6	18.5	32.0	18.3	18.5	18.8	19.6
Xe	19.0	30.1	41.5	76.0	27.4	27.9	28.4	30.4

TABLE II

The parameters R_{xy} and α_p for some alkali atoms in the ground state (R_{xy} and α_p in atomic units)

Atom	ω	R_{01}/α	R_{10}/α	R_{12}/α	α_0	α_1
K	ω_N	-2.92×10^4	4.33×10^4	-1.17×10^4	610	6.61
	ω_R	-2.13×10^5	2.90×10^5	-8.57×10^4	-1296	46.5
$\mathrm{R_b}$	ω_N	-3.51×10^4	5.38×10^4	-1.34×10^4	706	35.5
	ω_R	-1.50×10^5	2.19×10^5	-7.24×10^4	-1146	137
$\mathrm{C_s}$	ω_N	-1.09×10^5	1.45×10^5	-2.15×10^4	1219	254
	ω_R	-5.75×10^4	8.62×10^4	-2.96×10^4	-720	124

TABLE III

The parameters R_{01} and α_0 for n^3P_0-states of Hg and Sr

ω	Hg, 6^3P_0		Sr, 5^3P_0	
	$\frac{1}{\alpha}R_{01}$, a.u.	α_0, a.u.	$\frac{1}{\alpha}R_{01}$, a.u.	α_0, a.u.
ω_N	554	55.8	1.4×10^4	28.6
ω_R	1368	65.8	2.8×10^6	1656
$2\omega_N$	4342	90.0	5.2×10^4	-8.42
$2\omega_R$	1.0×10^4	16.7	7.1×10^2	-45.2

TABLE IV

The parameters R_{xy} and α_p for n^3P_1-states of Hg and Sr

State	ω	$\frac{1}{\alpha} R_{01}$, a. u.	$\frac{1}{\alpha} R_{10}$, a. u.	$\frac{1}{\alpha} R_{12}$, a. u.	$\frac{1}{\alpha} R_{21}$, a. u.	α_0, a. u.	α_1, a. u..	α_2, a. u.
Hg, 6^3P_1	ω_N	396	1.24×10^4	2704.0	1.19×10^4	-65	13.1	88
	ω_R	970	2.86×10^4	1812.5	2.72×10^4	-80	29.5	117,7
	ω_{2N}	3262	8.45×10^4	$-7574{,}1$	7.70×10^4	-123	81.7	211.3
	ω_{2R}	-8235	-1.18×10^5	-15602	-1.16×10^5	-73.3	-148.3	-70.3
Sr, 5^3P_1	ω_N	7.78×10^4	2.73×10^6	1.57×10^5	2.73×10^6	-39	322.3	308
	ω_R	3.54×10^6	7.60×10^7	-2.57×10^7	6.26×10^7	-4000	6100	1.1×10^4
	ω_{2N}	-1.87×10^4	-2.09×10^7	-2.01×10^5	-2.07×10^7	-80	-250.7	-193.3
	ω_{2R}	2.43×10^3	6.20×10^4	-2.49×10^4	6.19×10^4	30.3	6.66	-35.7

TABLE V

The parameters R_{xy} and α_p for n^3P_2-states of Hg and Sr

State	ω	$\frac{1}{\alpha} R_{01}$, a. u.	$\frac{1}{\alpha} R_{10}$, a. u.	$\frac{1}{\alpha} R_{12}$, a. u.	$\frac{1}{\alpha} R_{21}$, a. u.	$\frac{1}{\alpha} R_{02}$, a. u.	α_0, a. u.	α_1, a. u.	α_2, a. u.
Hg, 6^3P_2	ω_N	-1.68×10^3	-5.67×10^2	-3.63×10^2	1.06×10^3	-1.13×10^3	-105.3	53.71	-178.4
	ω_R	-8.50×10^3	-3.09×10^3	5.87×10^3	4.35×10^3	3.70×10^3	-164.4	179.4	-330.5
	$2\omega_N$	-3.75×10^5	-1.50×10^5	-5.08×10^5	7.66×10^4	2.7×10^5	552.5	-1738	1828
	$2\omega_R$	-5.24×10^4	9.57×10^4	-2.30×10^4	2.08×10^3	1.95×10^4	228.0	227.7	109.7
Sr, 5^3P_2	ω_N	-1.39×10^4	-1.10×10^3	-8.45×10^4	1.01×10^5	1.12×10^5	-62.74	580.9	-402.8
	ω_R	-2.73×10^6	-1.11×10^6	4.66×10^6	-2.33×10^5	1.81×10^6	2130	-5370	6223
	$2\omega_N$	-1.43×10^6	4.02×10^5	-9.40×10^5	6.43×10^4	4.23×10^5	663.6	591.3	493.2
	$2\omega_R$	-7.89×10^2	-6.48	1.87×10^4	1.01×10^4	7.1×10^4	133.2	-81.59	123.2

TABLE VI

The parameters R_{xy} and α_p for $n^2P_{1/2}$-states of Tl, Ga and Al

State	ω	$\frac{1}{\alpha}R_{01}$, a. u.	$\frac{1}{\alpha}R_{10}$, a. u.	$\frac{1}{\alpha}R_{12}$, a. u.	α_0, a. u.	α_2, a. u.
Tl, $6^2P_{1/2}$	ω_N	-7.55×10^1	1.46×10^2	2.84×10^3	46.3	-12.4
	ω_R	-1.74×10^2	1.21×10^2	3.81×10^3	53.5	-24.9
	$2\omega_N$	-4.70×10^2	-6.08×10^1	6.43×10^3	67.8	-50.6
	$2\omega_R$	-1.27×10^4	-3.65×10^3	1.92×10^4	-48.6	304
Ga, $4^2P_{1/2}$	ω_N	1.14×10^3	1.60×10^2	2.26×10^4	51.8	-15.6
	ω_R	2.35×10^3	1.01×10^2	2.91×10^4	61,2	-32.8
	$2\omega_N$	5.06×10^3	-3.39×10^2	4.67×10^4	83.3	-74.7
	$2\omega_R$	-2.52×10^4	1.28×10^3	-3.06×10^4	3.87	222
Al, $3^2P_{1/2}$	ω_N	5.18×10^3	2.92×10^2	1.57×10^5	55.9	-11.8
	ω_R	1.17×10^4	3.29×10^2	1.96×10^5	65.7	-25.0
	$2\omega_N$	2.61×10^4	1.93×10^2	2.90×10^5	87.4	-56.7
	$2\omega_R$	-1.54×10^5	8.58×10^3	-3.04×10^4	43.3	302

TABLE VII

The parameters R_{xy} and α_p for the $6^2P_{3/2}$-state of thallium (in atomic units)

ω	$\frac{1}{\alpha}R_{01}$	$\frac{1}{\alpha}R_{10}$	$\frac{1}{\alpha}R_{12}$	$\frac{1}{\alpha}R_{21}$	$\frac{1}{\alpha}R_{32}$	α_0	α_1	α_2
ω_N	-1.20×10^3	3.01×10^3	-1.80×10^3	8.44×10^2	3.32×10^3	0.96×10^2	0.52×10^2	1.95×10^2
ω_R	-5.81×10^3	9.88×10^3	-6.08×10^4	3.60×10^3	2.08×10^4	1.44×10^2	1.62×10^2	3.46×10^2
$2\omega_N$	-2.88×10^6	3.89×10^6	-6.46×10^6	1.26×10^6	1.21×10^7	-1.71×10^3	-4.96×10^3	-6.23×10^3
$2\omega_R$	-5.70×10^5	1.44×10^6	-7.42×10^5	1.22×10^5	3.6×10^5	-8.72×10^2	1.13×10^3	-4.24×10^2

tion of the initial state $|nJ\rangle$ of the atom, and the radial Green function $g_\gamma(E; r, r')$, where γ is the set of spin-angular quantum numbers permitted by the selection rules. In the general case, such integrals are of the form:

$$\varrho^{n_1 n_2 \dots n_{N+1}}_{\gamma_1, \gamma_2, \dots, \gamma_N}(\omega_1, \dots, \omega_N)$$

$$= \langle R_{nJ}(r_1)| r_1^{n_1} g_{\gamma_1}(E_n+\omega_1; r_1, r_2) r_2^{n_2} \cdot \dots \cdot r_N^{n_N} g_{\gamma_N}(E_n+\omega_N; r_N, r_{N+1}) r_{N+1}^{n_{N+1}} | R_{nJ}(r_{N+1})\rangle, \quad (2.1)$$

where $n_1, \dots, n_{N+1}$ are 1 or 0 for $\hat{d}$ and $\hat{\mu}$, respectively.

The R_{xy} are determined by integrals of two Green functions $g_\gamma(N = 2)$, whereas the parameters α_p of the linear susceptibilities $\chi_{ij}(-\omega; \omega)$ — by integrals of one $(N = 1)$. To calculate ϱ, we have recourse to the expression for $g_\gamma(E; r, r')$ in the approximation of the model potential method [3, 4], where g_γ is of the form of a series in Laguerre polynomials and $\varrho(\omega_1, \dots, \omega_N)$ is calculated similarly in the form of a rapidly convergent series of the hypergeometrical type easily accessible to summation by computer. Some details of calculations involving the Green function $g_y(E, \boldsymbol{r}, \boldsymbol{r}')$ are to be found in Ref. [4]. Formulae for α_p and R_{xy} expressed by way of integrals $\varrho(\omega_1, \ldots, \omega_N)$ for some atomic terms with different values of J (and hence with differently M-degenerate levels) are adduced in Appendix. The corresponding numerical values are given in Tables I–VII.

In the following Sections, we consider some magneto-electric effects, described by the quantities R_{xy} and α_p.

3. *The Faraday effect*

Among the physical processes described by the tensor $\chi_{ijk}^{\mathrm{eem}}(-\omega; \omega, 0)$, one of the most important is the Faraday effect, widely applied in practice with the aim to obtain information regarding the structure and properties of material media. A statisfactorily complete discussion of the available results for the Faraday effect in atomic and molecular systems is to be found in the review article [5]. The angle ϕ by which the polarisation plane is rotated per unit length of the active medium in the magnetic field $\boldsymbol{H}$

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

is determined by the Verdet constant $V(\omega)$, of a value dependent on the parameters of the medium and the radiation frequency. Above, $n_\pm$ is the refractive index for right (left) polarized light. In rarefied gases, and omitting spin and magnetic effects, V is given by the classical formula of Becquerel [6]:

$$V(\omega) = -\frac{\mu_{\mathrm{B}}}{\hbar c}\,\omega\,\frac{dn(\omega)}{d\omega}, \tag{3.1}$$

where $\mu_{\mathrm{B}} = \dfrac{|e|\hbar}{2mc}$ – Bohr's magneton, $n(\omega)$ is the refractive index of the medium. A closer, quantum-mechanical analysis shows [7, 8] Verdet's constant to consist, in general, of three terms:

$$V(\omega) = \frac{\pi\mu_{\mathrm{B}}}{\hbar c}\,\omega\left\{A(\omega)+B(\omega)+\frac{C(\omega)}{kT}\right\}, \tag{3.2}$$

where the constants $A(\omega)$ and $B(\omega)$ are different functions of ω, and $C(\omega)$ describes the temperature-dependent part of V[1].

The difficulty inherent in the quantum-mechanical calculation of $V(\omega)$ of specific atoms and molecules results from the circumstance that the expression for $n(\omega)$ in (3.1) involves single summation, whereas the coefficients A and B of (3.2) involve two-fold

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[1] Though denoted as A, B, C, the three quantities are here defined otherwise than in Ref. [5].

summation over the discrete spectrum and likewise integration over the continuous spectrum of the stationary states of the quantal system [5]. The problem becomes simpler only at resonance i. e. if ω is close to an absorption frequency of the system, when only resonance terms need to be included in the sums. Here, however, one has to take into account the widths Γ of the resonance levels [5] as well as the eventuality of a non-linear dependence of the angle ϕ on the magnetic field strength H (Maccaluso-Corbino effect [6]).

We shall be considering the non-resonance case only, when $V(\omega)$ is determined by the components of the magneto-electric susceptibility tensor $\chi_{ijk}^{\mathrm{eem}}(-\omega;\omega,0)$ of the medium. The majority of existing calculations of V are for the simplest bi-atomic molecules: hydrogen [9–12], nitrogen [13], and oxygen [14], as well as for certain more highly complicated ones [10]. The authors cited had recourse to various methods and approximations, permitting the calculation of A, B, C only in the range of frequencies up to that of the first resonance. With regard to atoms, use is commonly made of the approximate formula (3.1), though some general relations have been derived by Rosenfeld [7] for A, B, C within the framework of LS-coupling. The derivative $\frac{dn}{d\omega}$ has been calculated for atomic hydrogen [15–17], the inert gases [17–19], and alkali atoms [19]. Experiment shows that for frequencies ω remote from those, ω_{nm}, of atomic transitions the values measured for V differ but unessentially from those calculated using the formula (3.1) (see, e. g. [20]); however, as ω tends to ω_{nm}, the discrepancy becomes significant. Thus, in the case of alkali atoms, measurements have been carried out in the region of the resonance doublet $nP_{1/2}$, $nP_{3/2}$ (for references, see the review article [5]) and the contribution from the B-term of Eq. (3.2) was found to be comparable in magnitude with A, so that formula (3.1) is no longer applicable.

Consequently, it is interesting to perform strict calculations of the Verdet constant (3.2) throughout a wide range of frequencies ω with the aim to elucidate the relative contributions from A and B and to determine the accuracy with which the approximate formula (3.1) is fulfilled in atomic gases. Besides, the problem of measuring the temperature-dependent terms $C(\omega)$ for isolated atoms presents considerable interest. With regard to atoms and ions in complexes of the transition metals, the important role of the temperature-dependent terms at low T has been noted in Ref. [5]. Rosenfeld [7] showed that in states with orbital moment $L > 0$ the term $C(\omega)$ of (3.2) is directly proportional to the paramagnetic susceptibility of the atom. Nonetheless, even in S_J-states (such as are the ground states of most atoms) $C(\omega)$ can be non-zero at spin moment $s = J$ different from zero. This situation can occur, in particular, in the heavy alkali atoms Rb and Cs, for which the antisymmetric polarizability α_1 (see [1]) becomes considerable in the optical frequency range [21].

The tensor $\chi_{ijk}^{\mathrm{eem}}(-\omega;\,\omega,\,0)$ is analogical to the tensor $\chi_{kij}^{\mathrm{mee}}(0;\,-\omega,\,\omega)$, (I, 2.10), defining the inverse Faraday effect [22–24]. In the general case, these tensors depend on the spatial dispersion, related with multipolar electric and magnetic transitions [25].

By having recourse to the formula

$$(n_{\pm}^2-1)E_{\pm}(\omega) = 4\pi P_{\pm}(\omega)$$

of classical electrodynamics and applying the expansion (I, 2.4, 5), we write the rotation angle ϕ for the isolated atom in the following form:

$$\phi = -\frac{\omega}{2c}\frac{4\pi i P_y(\omega)}{nE_x(\omega)} = -i\frac{2\pi\omega}{nc}\{\chi^{\mathrm{e}}_{yx}(-\omega;\omega)+\chi^{\mathrm{eem}}_{yxz}(-\omega;\omega,0)H_z\}, \tag{3.3}$$

where $\boldsymbol{H}$ is assumed as directed along the z-axis and $\boldsymbol{E}$ as polarized along x. When considering an ensemble of atoms with the density N_0 at the temperature T, Eq. (3.3) has to be averaged with the statistical distribution function:

$$f_r = C\exp\left\{-\frac{\Delta E}{kT}\right\}, \tag{3.3}$$

where $\Delta E = \langle nJM| - (\boldsymbol{\mu}\cdot\boldsymbol{H})|nJM\rangle = \mu_{\mathrm{B}} g_J MH$, with g_J denoting the Landé factor of the atom. As a result, χ^{e}_{yx} of (3.3) is multiplied by $-\dfrac{\Delta E}{kT}$. Next, the expression thus derived has to be averaged over the projections M of J. This is easy to perform if use is made of the following relation, resulting from (I, 2.22):

$$\overline{\chi^{\mathrm{eme}}_{ijk}(-\omega;0,\omega)} = \frac{1}{2J+1}\sum_{M=-J}^{J}\chi^{\mathrm{eme}}_{ijk}(-\omega;0,\omega) = \begin{pmatrix}1 & 1 & 1\\ j & i & k\end{pmatrix}R_{01}. \tag{3.5}$$

In particular, on averaging over all orientations, the magneto-electric susceptibility takes a form analogical to that of the non-degenerate case (I, 2.24) i.e. is determined by the sole atomic parameter R_{01}. Averaging of the product $\Delta E\,\chi^{\mathrm{e}}_{ij}(-\omega;\omega)$ is performed using (I, 2.14) and leads to a non-zero α_1-term. The final expression for the Verdet constant can be written in the form:

$$V(\omega) = \frac{\phi}{H(0)} = \frac{2\pi\omega}{\hbar c}N_0\left(\frac{1}{\sqrt{6}}R_{01} - \frac{1}{3kT}\mu_{\mathrm{B}} g_J\cdot\sqrt{\frac{J(J+1)}{6}}\cdot\alpha_1\right). \tag{3.6}$$

We now proceed to apply the formula (3.6) to alkali atoms and inert gases.

For alkali atoms, the moment J of the ground state is 1/2 and, with regard to (I, 2.22, 23), we obtain for R_{01}:

$$\begin{aligned}R_{01} = -\tfrac{1}{2}\sum_{J_1,J_2}\begin{Bmatrix}1 & 1 & 1\\ J_1 & J_2 & \tfrac{1}{2}\end{Bmatrix}&(\langle n\tfrac{1}{2}\,\|\boldsymbol{d}\{g_{J_1}(E_n-\omega;\boldsymbol{r}_1,\boldsymbol{r}_2)\boldsymbol{\mu}g_{J_2}(E_n-\omega;\boldsymbol{r}_2,\boldsymbol{r}_3)\\ &-g_{J_1}(E_n+\omega;\boldsymbol{r}_1,\boldsymbol{r}_2)\boldsymbol{\mu}g_{J_2}(E_n+\omega;\boldsymbol{r}_2,\boldsymbol{r}_3)\}\boldsymbol{d}\|n\tfrac{1}{2}\rangle\\ &+\langle n\tfrac{1}{2}\,\|\boldsymbol{\mu}\|n\tfrac{1}{2}\rangle\,\langle n\tfrac{1}{2}\,\|\boldsymbol{d}\{g_{J_1}(E_n-\omega;\boldsymbol{r}_1,\boldsymbol{r}_2)g_{J_1}(E_n-\omega;\boldsymbol{r}_2,\boldsymbol{r}_3)\\ &-g_{J_1}(E_n+\omega;\boldsymbol{r}_1,\boldsymbol{r}_2)g_{J_1}(E_n+\omega;\boldsymbol{r}_2,\boldsymbol{r}_3)\}\boldsymbol{d}\|n\tfrac{1}{2}\rangle).\end{aligned} \tag{3.7}$$

The reduced matrix elements of the operators $\boldsymbol{d}$ and $\boldsymbol{\mu}$ are easy to calculate applying well known formulae of angular momentum theory, as a result of which $V(\omega)$ for states with $J = \frac{1}{2}$ takes the form (for the sake of convenience, our formula is expressed in the atomic system of units with $e = m = \hbar = 1$):

$$V(\omega) = \pi\alpha^2 N_0 \omega \left[\tfrac{1}{27} (7\Delta_{\frac{3}{2},\frac{3}{2}} + 4\Delta_{\frac{1}{2},\frac{1}{2}} - 2\Delta_{\frac{1}{2},\frac{3}{2}}) + \frac{\alpha_1}{3\sqrt{2}\,kT} \right],$$

$$\begin{aligned}\Delta_{J_1,J_2} = \langle R_{n\frac{1}{2}}(r_1) | r_1 \{ & g_{J_1}(E_n+\omega;\, r_1, r_2) g_{J_2}(E_n+\omega;\, r_2, r_3) \\ & - g_{J_1}(E_n+\omega;\, r_1, r_2) g_{J_2}(E_n-\omega;\, r_2, r_3) \} r_3 | R_{n\frac{1}{2}}(r_3) \rangle .\end{aligned} \tag{3.8}$$

Above, the Δ_{J_1,J_2} are radial integrals, and α_1 — the vectorial polarizability.

If, in Δ_{J_1,J_2}, the dependence of the Green function on the moments J_1, J_2 is neglected (i. e. if g is dealt with as a hydrogen-like Green function with orbital moment $l = 1$), α_1 vanishes in this approximation [21] and (3.8) goes over into the Becquerel formula (3.1). Consequently, the distribution between (3.1) and (3.2) resides in spin-orbital splitting of the excited states of the atom — a factor obviously not taken into account in the classical theory of Faraday's effect. It is moreover worth noting that to the B-term of (3.2) corresponds the term in $\Delta_{1/2,3/2}$ of (3.8), and to the A-term — the sum of those in $\Delta_{1/2,1/2}$ and $\Delta_{3/2,3/2}$. As a consequence, in the non-resonance case when spin-orbit splitting is unessential, the relation $B(\omega) \approx -\frac{2}{11} A(\omega)$ holds for the terms A and B of Serber's formula (3.2). On comparing the calculations based on formula (3.8) and those of Ref. [19] based on (3.1) we find that for frequencies remote from resonance (when frequency dis-

TABLE VIII

The dispersion of Verdet constant for Cs and comparison of the exact results with Becqerel formula

$\omega \times 10^{-3}$, cm^{-1}	$y(\omega)$ in atomic units	$\dfrac{y(\omega)}{d\alpha/d\omega}$
10.6	1.133×10^6	1.053
10.7	1.597×10^6	1.068
10.8	2.395×10^6	1.089
10.9	2.196×10^6	1.121
11.0	9.762×10^6	1.171
11.1	4.972×10^7	1.248
11.178	∞	—
11.2	6.904×10^8	1.360
11.3	2.722×10^7	1.472
11.4	1.357×10^7	1.490
11.5	1.538×10^7	1.392
11.6	3.608×10^7	1.274
11.7	5.374×10^8	1.189
11.732	∞	—
11.8	1.175×10^8	1.132
11.9	1.892×10^7	1.096
12.0	7.466×10^6	1.072
12.1	3.985×10^6	1.056

tuning exceeds spin-orbit splitting) the discrepancy is very small, amounting to about $(1 \div 2)\%$. However, closer and closer to resonance, it becomes quite significant. As an example, Table VIII shows the dispersional dependence $y(\omega) = \frac{V(\omega)}{\pi\alpha^2 N_0 \omega}$ within the region of the resonance doublet $6^2S_{1/2}-6^2P_{1/2,3/2}$ of the cesium atom as calculated from formulae (3.1) and (3.8).

The temperature-dependent term C in (3.2) is related with the vectorial polarizability α_1 and can contribute significantly in the optical frequency region, where α_1 becomes considerable [21]. Thus, at the neodymium laser frequency $\omega_N = 9440\,\mathrm{cm}^{-1}$, the contribution from $C(\omega)$ to $V(\omega)$ can amount to as much as $\sim 15\%$ in the case of the cesium atom under normal conditions at $T = 300\mathrm{K}$.

In the ground states of the inert gases, the total momentum of the atom is $J = 0$; the temperature-dependent term is absent, and R_{01} takes the form (A.1). Calculations with formula (3.6) show that, in the optical frequency region, the divergence from Becquerel's formula does not exceed $\sim 1\%$. This was to be expected, since for inert gases the absorption lines originating in the ground state lie in the ultraviolet region and the multiplet structure of the excited states, if taken into account, does not affect essentially the magnitude of $V(\omega)$. In connection with the aforesaid it is worth noting that, in the experiments of Ingersol and Liebenberg [20], the divergence of V_{exp} from $V_{\mathrm{Becq.}}$ for neon amounted to $\sim 10\%$ and decreased with increasing ω. A divergence like that cannot be explained as resulting from an inadequacy of formula (3.1) but can be related, for instance, with some inaccuracy incurred when approximating the frequency-dependence of the refractive index by means of the single-term formula:

$$n(\omega) = \frac{q}{\omega_0^2 - \omega^2}, \tag{3.9}$$

applied in Ref. [20]. The accuracy of such an approximation increases as ω tends to a resonance frequency of the atom, and this can serve to explain why the divergence between V_{exp} and $V_{\mathrm{Becq.}}$ decreases with increasing ω.

The Verdet constant (3.6) is moreover related with that of the inverse Faraday effect [22–24] defining the static magnetic moment $\boldsymbol{m}(0)$ induced in the medium by an electromagnetic wave with a non-zero circularly polarized component. Hence, irrespective of whether investigation of atoms in degenerate states proceeds by the normal or inverse Faraday effect, only one of the five atomic parameters, namely R_{01}, can be measured.

4. *Rayleigh scattering of light by atoms in magnetic fields*

In Rayleigh scattering of light by a system in a magnetic field $\boldsymbol{H}$ the cross-section for scattering contains terms proportional to $\boldsymbol{H}$ which lead to an anisotropy of the scattering process and to a difference in the magnitudes of $\frac{d\sigma}{d\Omega}$ for left and right circularly polarized incident light. The problem of Rayleigh scattering in the presence of a field

$\boldsymbol{H}$ has been considered previously in Ref. [26] where, however, only general formulae for $\frac{d\sigma}{d\Omega}$ are adduced in terms of the susceptibilities χ_{ij}, χ_{ijk}. Further on, we shall derive expressions for the scattering cross-section in terms of the invariant parameters α_p, R_{xy}.

In the notation of formula (I, 2.16), the scattering cross-section can be expressed as follows up to terms linear in $\boldsymbol{H}$:

$$\frac{d\sigma}{d\Omega_{e'}} = \frac{1}{2J+1} \sum_{M,M'} \left| \sum_{ijk} e_i'^{*} e^k \{\chi^{\mathrm{e}}_{ik}(-\omega;\omega) + \chi^{\mathrm{eme}}_{ijk}(-\omega;0,\omega) H^j\} \right|^2 \left(\frac{\omega}{c}\right)^4$$

$$= \left(\frac{d\sigma}{d\Omega_{e'}}\right)_0 + \frac{1}{2J+1} \sum_{M,M'} \left\{ \sum_{ijki'k'} e_i'^{*} e^k e'_{i'} e^{k'*} H^j \chi^{\mathrm{e}}_{i'k'} \chi^{\mathrm{eme}}_{ijk}(-\omega;0,\omega) + \text{compl. conj.} \right\}, \quad (4.1)$$

where $\left(\frac{d\sigma}{d\Omega_{e'}}\right)_0$ is the cross-section at $H = 0$ given by (I, 2.16) with $\omega = \omega'$. By (I, 2.16, 22) and applying irreducible tensor operator technique [2], (4.1) can be brought to invariant form:

$$\frac{d\sigma}{d\Omega_{e'}} - \left(\frac{d\sigma}{d\Omega_{e'}}\right)_0 = -\sum_{x,y} \alpha_x^{*} Q_{xy}(\{\{\boldsymbol{e}^{*}\otimes\boldsymbol{e}'\}_x \otimes \{\boldsymbol{e}'^{*}\otimes\boldsymbol{e}\}_y\}_1 \boldsymbol{H})_0 + \text{compl. conj.}, \quad (4.2)$$

where

$$Q_{xy} = \frac{1}{2x+1}\left[\frac{3}{2y+1}\right]^{1/2} R_{xy}.$$

By (4.2) we have $x \neq 3$ and the parameter R_{32} does not contribute to the scattering process. Inasmuch as we consider nonresonance scattering at a frequency ω less than the ionisation potential of the state $|nJ\rangle$, the parameters α_x and Q_{xy} are real. Moreover, the wave vector $\boldsymbol{k}$ of the incident photon is directed along $\boldsymbol{H}$ (the z-axis), whence $(\boldsymbol{e}\cdot\boldsymbol{H}) = 0$, and:

$$([\boldsymbol{e}\times\boldsymbol{e}^{*}]\cdot\boldsymbol{H}(0)) = -iH(0)\xi_2, \quad (4.3)$$

where ξ_2 is the Stokes parameter of the incident radiation defining the degree of its circular polarisation. Formula (4.3) follows from the well known relation [27]:

$$\xi_2 = i([\boldsymbol{e}\times\boldsymbol{e}^{*}]\cdot\boldsymbol{n}), \quad \text{with} \quad \boldsymbol{n} = \frac{\boldsymbol{k}}{|\boldsymbol{k}|}.$$

With regard to the aforesaid and to a formula of Ref. [2] reducing the tensorial product of four vectors in Eq. (4.2) to usual scalar and vector products, we re-write $\frac{d\sigma}{d\Omega}$

as follows:

$$\Delta = \frac{d\sigma}{d\Omega_{e'}} - \left(\frac{d\sigma}{d\Omega_{e'}}\right)_0$$

$$= \sqrt{\tfrac{2}{3}}\left(\alpha_0 R_{01} + \frac{1}{\sqrt{3}}\,\alpha_1 R_{10}\right) \operatorname{Im}\left((\boldsymbol{e}^* \cdot \boldsymbol{e}')\,([\boldsymbol{e}\times\boldsymbol{e}'^*]\cdot\boldsymbol{n})\right)H$$

$$+\tfrac{1}{5}\sqrt{\tfrac{2}{15}}\left(\alpha_1 R_{12}\,\frac{1}{\sqrt{15}} + 3\alpha_2 R_{21}\right)\left\{\tfrac{1}{3}\operatorname{Im}\left((\boldsymbol{e}\cdot\boldsymbol{e}'^*)\,([\boldsymbol{e}^*\times\boldsymbol{e}']\cdot\boldsymbol{n})\right) + \tfrac{1}{2}\,\xi_2 |(\boldsymbol{n}\cdot\boldsymbol{e}')|^2\right\}H. \qquad (4.4)$$

For the non-degenerate state, only the parameter R_{01} differs from zero, and Δ takes the form:

$$\Delta = \sqrt{\tfrac{2}{3}}\,\alpha_0 R_{01} \operatorname{Im}\left((\boldsymbol{e}^*\cdot\boldsymbol{e}')\,([\boldsymbol{e}\times\boldsymbol{e}'^*]\cdot\boldsymbol{H})\right)$$

for arbitrary orientation of the vectors $\boldsymbol{e}$, $\boldsymbol{e}'$ and $\boldsymbol{H}$. Obviously, now, R_{01} is related with the Verdet constant:

$$R_{01} = \frac{c\sqrt{6}}{\pi\omega}\,V(\omega).$$

As shown if Ref. [26], it is of primary interest to determine the difference $\Delta^+ - \Delta^-$ between the cross-sections for incident, right ($\boldsymbol{e} = \boldsymbol{e}_{+1}$) and left ($\boldsymbol{e} = \boldsymbol{e}_{-1}$) circularly polarized light. With respect to circularly polarized light, the following relation holds obviously:

$$\operatorname{Im}\left((\boldsymbol{e}^*\cdot\boldsymbol{e}')\,(\boldsymbol{n}\cdot[\boldsymbol{e}\times\boldsymbol{e}'^*])\right) = \mp|(\boldsymbol{e}'^*\cdot\boldsymbol{e})|^2.$$

The upper and lower signs correspond, respectively, to the right and left sense of circular polarisation. Finally, we obtain for $\Delta^+ - \Delta^-$:

$$\Delta^+ - \Delta^- = \frac{\sqrt{2}}{5}\left(\alpha_1 R_{12} + \sqrt{\tfrac{3}{5}}\,\alpha_2 R_{21}\right)|(\boldsymbol{e}'\cdot\boldsymbol{n})|^2 H$$

$$+\frac{4}{\sqrt{6}}\left[\frac{1}{5\sqrt{3}}\left(\alpha_1 R_{12} + \sqrt{\tfrac{3}{5}}\,\alpha_2 R_{21}\right) - \alpha_1 R_{10} - \frac{1}{\sqrt{6}}\,\alpha_0 R_{01}\right]|(\boldsymbol{e}'^*\cdot\boldsymbol{e})|^2 H. \qquad (4.5)$$

5. *The splitting and shift of atomic levels in crossed light and magnetic fields*

When considering the modifications of the atomic spectrum under the simultaneous action of a light laser wave and a static magnetic field $\boldsymbol{H}$, we shall be assuming that:

1. Zeeman splitting is large compared with quadratic Stark splitting (I, 2.20, 21);
2. H is sufficiently weak for Zeeman splitting to be small compared with multiplet splitting of the isolated atom and for usual, anomalous Zeeman splitting to occur in the absence of the light wave;

3. terms quadratic in H (quadratic Zeeman effect) are small compared with the Stark shift and are thus negligible; and

4. the light frequency ω is remote from the atomic transition frequencies, so that the perturbation of atomic levels is of the non-resonance kind (certain specific features of the atomic spectrum in a magnetic and resonance light field have been studied in Refs (28, 29]).

The situation under consideration takes place e. g. in alkali atoms in fields $H\sim(10^3\div 10^4)$ Oe, $E=(10^5\div 10^6)$ V/cm, typical for experiment. In such cases, the change in energy of a level $|nJ\rangle$ reduces to Zeeman splitting proportional to H into $(2J+1)$ equidistant components with different projections M of the momentum $\boldsymbol{J}$ onto the direction of $\boldsymbol{H}$ as well as to a Stark shift of each of the sub-levels. Besides the usual Stark shift, proportional to $|E|^2$, there occur interferencial terms proportional to $H|\boldsymbol{E}|^2$, defined by the magnitude of the magneto-electric susceptibility $\chi^{\text{eem}}_{ijk}\,(-\omega;\omega,0)$. The latter terms will be the object of our study.

Quite generally, the change in energy ΔE_{nJM} is of the form:

$$\Delta E_{nJM}=\mu_{\mathrm{B}}g_J MH-\tfrac{1}{4}A_{JM}|\boldsymbol{E}|^2+\tfrac{1}{4}B_{JM}H|\boldsymbol{E}|^2. \tag{5.1}$$

The quantities A_{JM} and B_{JM} are given by the compound matrix elements of 2-nd and 3-rd order perturbation calculus for quasi-energetical states of the system in the monochromatic wave (cf., Ref. [30]); expressed by way of the susceptibilities χ, they are of the form:

$$A_{JM}|\boldsymbol{E}|^2=\sum_{i,j=0,\pm 1}\chi^{\mathrm{e}}_{ij}(-\omega;\omega)E_i(\omega)E_j^*(\omega) \tag{5.2}$$

$$B_{JM}H|\boldsymbol{E}|^2=\sum_{i,k,j}\chi^{\mathrm{eem}}_{ijk}(-\omega;\omega,0)E_i(\omega)E_j^*(\omega)H_k(0). \tag{5.3}$$

With regard to (I,2.14), we obtain:

$$A_{JM}|\boldsymbol{E}|^2=\sqrt{3(2J+1)}\,(-1)^{J+M}\sum_{p=0,1,2}\begin{pmatrix} J & J & p\\ -M & M & 0\end{pmatrix}\frac{\alpha_p}{\sqrt{2p+1}}\{\boldsymbol{E}\otimes\boldsymbol{E}^*\}_{p,0} \tag{5.4}$$

Contrary to (I,2.20, 21), the preceding expression is valid at arbitrary elliptical polarisation $\boldsymbol{E}(t)$ since the projection of $\boldsymbol{J}$ onto $\boldsymbol{H}$ remains a good quantum number. To obtain explicitly the dependence of ΔE_{nJM} on the mutual orientation of the fields $\boldsymbol{E}$ and $\boldsymbol{H}$, we have recourse to the well known relations [2, 27]:

$$\{\boldsymbol{E}\otimes\boldsymbol{E}^*\}_{20}=\frac{1}{\sqrt{6}}|\boldsymbol{E}|^2\left(\frac{3|(\boldsymbol{E}\cdot\boldsymbol{H})|^2}{|\boldsymbol{E}|^2\boldsymbol{H}^2}-1\right), \tag{5.5a}$$

and

$$i[\boldsymbol{E}\times\boldsymbol{E}^*]_z=\xi_2|\boldsymbol{E}|^2\cos\beta, \tag{5.5b}$$

where ξ_2 is the degree of circular polarization of $\boldsymbol{E}(t)$ (4.3), and β is the angle between the propagation direction of the wave and the vector $\boldsymbol{H}$. Finally, we obtain:

$$A_{JM}=\alpha_0+\frac{\alpha_1 M}{\sqrt{2J(J+1)}}\xi_2\cos\beta-\tfrac{1}{2}\alpha_2\frac{3M^2-J(J+1)}{\sqrt{5J(J+1)(2J+3)(2J-1)}}(3\cos^2\theta-1), \tag{5.6}$$

where

$$|(\boldsymbol{E}\cdot\boldsymbol{H})| = |\boldsymbol{E}|\,|\boldsymbol{H}|\cos\theta.$$

Along similar lines, Eq. (5.3) becomes:

$$B_{JM} = \frac{R_{10}}{\sqrt{J(J+1)}}M + \left(R_{01} - R_{21}\frac{3M^2 - J(J+1)}{[J(J+1)(2J+3)(2J-1)]^{1/2}}\right)\frac{\xi_2\cos\beta}{\sqrt{6}}$$

$$+\tfrac{1}{5}\left(\frac{R_{12}}{\sqrt{J(J+1)}} - 4R_{32}\{3J(J+1)-1-5M^2\}\left[\frac{3(2J+1)(2J-3)!}{2(2J+4)!}\right]^{1/2}\right)M(3\cos^2\theta - 1). \tag{5.7}$$

Thus, the modification of the spectrum proportional to $H|\boldsymbol{E}|^2$ is defined by the five parameters R_{xy}; here, in contradistinction to the case of scattering in a magnetic field (Section 4), the parameter R_{32} contributes also. It follows from (5.6, 7) that ΔE_{nJM} depends essentially on the mutual orientation of $\boldsymbol{E}(t)$ and $\boldsymbol{H}$. In particular, at linear polarization $\boldsymbol{E}(t)$ ($\xi_2 = 0$) when $\theta = \arccos\frac{\sqrt{3}}{3}$ the sublevels are equidistant as in the case of usual Zeeman splitting.

The action of the light wave now causes a Stark shift in "centre of mass" of the Zeeman multiplet and a renormalisation of the Landé factor proportional to the wave intensity.

$$g_J^{\text{mod}} = g_J + \frac{1}{\mu_{\text{B}}}\frac{R_{10}}{4\sqrt{J(J+1)}}|\boldsymbol{E}|^2. \tag{5.8}$$

Clearly, the experimental observation of the interferencial terms, proportional to $H|\boldsymbol{E}|^2$, may be beset with difficulties inasmuch as they contribute but weakly to the quadratic Stark effect, (5.6). In order to eliminate the Stark effect one can e. g. proceed to measure the splitting of sublevels with the projections $+M$ and $-M$ in a linearly polarized field $\boldsymbol{E}(t)$. In this case the terms proportional to $|\boldsymbol{E}|^2$ contribute nothing (except for a shift in "centre of mass"), and the change in Zeeman splitting $\Delta_0 = 2\mu_{\text{B}}g_JMH$ is defined by the terms involving R_{12}, R_{32} of (5.7). Fig. 1 visualizes qualitatively the situation for the level

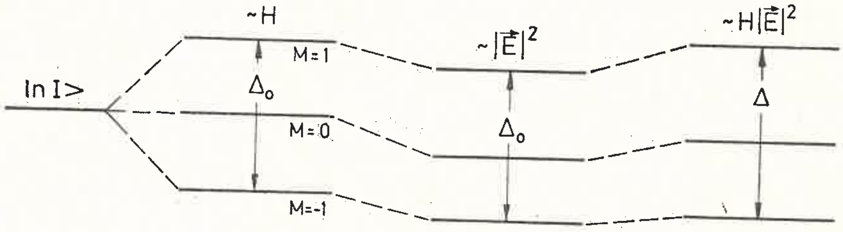

Fig. 1. Qualitative behaviour of the level with $J = 1$ in crossed electric and magnetic fields

with $J = 1$. The sign of $\Delta-\Delta_0$ in Fig. 1 depends on the angle θ. By varying θ, the difference $\Delta-\Delta_0$ can be made positive or negative.

The general formulae (5.6, 7) simplify considerably in the non-degenerate case, and ΔE_{nJM} now is:

$$\Delta E_{n,J=0} = -\tfrac{1}{4}\,\alpha_0|E|^2+\tfrac{1}{4}\,\frac{R_{01}}{\sqrt{6}}\,H|E|^2\xi_2\cos\beta. \tag{5.9}$$

Above, the first term represents the usual Stark shift, defined by the scalar polarizability α_0, whereas the second term is non-zero only if the degree of circular polarisation of $\boldsymbol{E}(t)$ differs from zero, and vanishes in a static electric field E. The latter part of the shift depends on the interaction between the magnetic moment $\mu_{\text{mag}} \approx |E|^2\,\boldsymbol{K}$, induced in the atom by the inverse Faraday effect, and the field $\boldsymbol{H}$.

6. *Conclusion*

The preceding analysis and examples show conclusively that the formalism of decomposition of the magneto-electric susceptibility tensors for degenerate states into irreducible parts, leading to the specification of invariant atomic paramaters, is highly convenient with regard to the investigation of various processes of interaction between atoms and external fields.

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APPENDIX

On introduction of the following notation for sums and differences of the integrals of Eq. (2.1):

$$\Delta(\omega_1, \ldots, \omega_N) \equiv \varrho(\omega_1, \ldots, \omega_N)-\varrho(-\omega_1, \ldots, -\omega_N),$$

$$\sigma(\omega_1, \ldots, \omega_N) \equiv \varrho(\omega_1, \ldots, \omega_N)+\varrho(-\omega_1, \ldots, -\omega_N),$$

the quantities α_p and R_{xy} for well-defined atomic terms take the forms adduced further on sub (a)–(e). For the sake of conveniency, we give in formula (A.1)–(A.5) the compound matrix elements T_p and T_{xy} by way of which the R_{xy} are expressed in conformity with formula (I, 2.23) (in atomic units, $\alpha = 1/137$):

$$R_{xy} = \alpha(2y+1)\left[\frac{2x+1}{3(2J+1)}\right]^{1/2}\left(T_{xy}-\frac{1}{\alpha}\langle nJ\|\boldsymbol{\mu}\|nJ\rangle\begin{Bmatrix}1 & y & x\\ J & J & J\end{Bmatrix}T_y\right).$$

The parameters α_p of linear susceptibilities are given by the formula:

$$\alpha_p = \frac{2p+1}{\sqrt{3(2J+1)}}\,T_p^0,$$

where T_p^0 is of the same form as T_p on performing the interchange:

$$\Delta^{101}_{\gamma,\gamma}(\omega, \omega) \to \Delta^{11}_{\gamma}(\omega), \qquad \sigma^{101}_{\gamma,\gamma}(\omega, \omega) \to \sigma^{11}_{\gamma}(\omega).$$

Thus, we refrain from writing the formula for T_p^0.

We apply the spectroscopic notation of atomic levels currently used in handbooks on atomic spectroscopy:

(a) $np^5\ {}^1S_0$ — ground states of the inert gases:

$$\alpha_1 = \alpha_2 = 0, \qquad R_{10} = R_{12} = R_{21} = R_{32} = 0,$$

$$T_0 = \frac{1}{9\sqrt{3}}\left[\sigma^{101}_{\gamma_1\gamma_1}(\omega, \omega) + 2\sigma^{101}_{\gamma_2\gamma_2}(\omega, \omega) + 2\sigma^{101}_{\gamma_3\gamma_3}(\omega, \omega) + 2\sigma^{101}_{\gamma_4\gamma_4}(\omega, \omega) + 2\sigma^{101}_{\gamma_5\gamma_5}(\omega, \omega)\right],$$

$$T_{01} = \frac{1}{81\sqrt{2}}\left[4\Delta^{101}_{\gamma_1\gamma_1}(\omega, \omega) + 5\Delta^{101}_{\gamma_2\gamma_2}(\omega, \omega) + 7\Delta^{101}_{\gamma_3\gamma_3}(\omega, \omega) + 8\Delta^{101}_{\gamma_4\gamma_4}(\omega, \omega) + 5\Delta^{101}_{\gamma_5\gamma_5}(\omega, \omega)\right.$$
$$\left. - 2\Delta^{101}_{\gamma_1\gamma_3}(\omega, \omega) - 2\Delta^{101}_{\gamma_2\gamma_4}(\omega, \omega) - 2\Delta^{101}_{\gamma_4\gamma_5}(\omega, \omega) + 4\Delta^{101}_{\gamma_2\gamma_5}(\omega, \omega)\right]. \tag{A.1}$$

Above, $\gamma = \{j_0 l [K]_J\}$ is the set of quantum numbers within the framework of Jl-coupling: j_0 — momentum of the core, l — orbital momentum of the electron, K — "intermediate" momentum, J — total momentum.

$$\gamma_1 = \{\tfrac{1}{2}\,0[\tfrac{1}{2}]_1\}, \qquad \gamma_2 = \{\tfrac{1}{2}\,2[\tfrac{3}{2}]_1\}, \qquad \gamma_3 = \{\tfrac{3}{2}\,0[\tfrac{3}{2}]_1\},$$
$$\gamma_4 = \{\tfrac{3}{2}\,2[\tfrac{1}{2}]_1\}, \qquad \gamma_5 = \{\tfrac{3}{2}\,2[\tfrac{3}{2}]_1\}.$$

(b) $n^2S_{1/2}$-states of alkali atoms:

Above, and henceforth: $\gamma = \{JL\}$, J and L — total and orbital momenta of the valence electron.

$$\alpha_2 = 0, \qquad R_{21} = R_{32} = 0, \qquad \langle n\,\tfrac{1}{2}\,\|\mu\|\,n\,\tfrac{1}{2}\rangle = \alpha\sqrt{3/2}.$$

$$T_0 = \sqrt{\tfrac{2}{27}}\left[\sigma^{101}_{1\frac{1}{2},1\frac{1}{2}}(\omega, \omega) + 2\sigma^{101}_{1\frac{3}{2},1\frac{3}{2}}(\omega, \omega)\right],$$

$$T_1 = \tfrac{2}{9}\left[\Delta^{101}_{1\frac{1}{2},1\frac{1}{2}}(\omega, \omega) - \Delta^{101}_{1\frac{3}{2},1\frac{3}{2}}(\omega, \omega)\right].$$

$$T_{01} = \tfrac{1}{27}\left[\Delta^{101}_{1\frac{1}{2},1\frac{1}{2}}(\omega, \omega) - 2\Delta^{101}_{1\frac{1}{2},1\frac{3}{2}}(\omega, \omega) + 10\Delta^{101}_{1\frac{3}{2},1\frac{3}{2}}(\omega, \omega)\right],$$

$$T_{10} = \frac{1}{27\sqrt{6}}\left[\sigma^{101}_{1\frac{1}{2},1\frac{1}{2}}(\omega, \omega) - 8\sigma^{101}_{1\frac{1}{2},1\frac{3}{2}}(\omega, \omega) - 20\sigma^{101}_{1\frac{3}{2},1\frac{3}{2}}(\omega, \omega)\right],$$

$$T_{12} = \frac{1}{27\sqrt{6}}\left[-2\sigma^{101}_{1\frac{1}{2},1\frac{1}{2}}(\omega, \omega) + \sigma^{101}_{1\frac{1}{2},1\frac{3}{2}}(\omega, \omega) + 4\sigma^{101}_{1\frac{3}{2},1\frac{3}{2}}(\omega, \omega)\right]. \tag{A.2}$$

(c) $n^2P_{1/2}$-states (for example, the atoms Tl, Al, Ga, ...):

$$\alpha_2 = 0, \qquad R_{21} = R_{32} = 0, \qquad \langle n\,\tfrac{1}{2}\,\|\mu\|\,n\,\tfrac{1}{2}\rangle = \frac{\alpha}{\sqrt{6}}.$$

$$T_0 = \sqrt{\tfrac{2}{27}}\left[\sigma^{101}_{0\frac{1}{2},0\frac{1}{2}}(\omega, \omega) + 2\sigma^{101}_{2\frac{1}{2},2\frac{1}{2}}(\omega, \omega)\right],$$

$$T_1 = \tfrac{2}{9}\left[\Delta^{101}_{0\frac12,0\frac12}(\omega,\omega) - \Delta^{101}_{2\frac12,2\frac12}(\omega,\omega)\right].$$

$$T_{10} = \frac{1}{3\sqrt{6}}\left[\sigma^{101}_{0\frac12,0\frac12}(\omega,\omega) - 4\sigma^{101}_{2\frac32,2\frac32}(\omega,\omega)\right],$$

$$T_{01} = \tfrac{1}{9}\left[\Delta^{101}_{0\frac12,0\frac12}(\omega,\omega) + 2\Delta^{101}_{2\frac32,2\frac32}(\omega,\omega) - \tfrac{2}{3}\,\Delta^{011}_{1\frac32,0\frac12}(0,\omega) + \tfrac{2}{3}\,\Delta^{011}_{1\frac32,2\frac32}(0,\omega)\right],$$

$$T_{12} = -\frac{2}{9\sqrt{6}}\left[\sigma^{101}_{0\frac12,0\frac12}(\omega,\omega) - \tfrac{2}{5}\,\sigma^{101}_{2\frac32,2\frac32}(\omega,\omega) + \sigma^{011}_{1\frac32,0\frac12}(0,\omega) + \tfrac{1}{5}\,\sigma^{011}_{1\frac32,2\frac32}(0,\omega)\right]. \quad \text{(A.3)}$$

(d) n^3P_0-state (metastable states Hg, Sr, ...):

$$\alpha_1 = \alpha_2 = 0, \quad R_{10} = R_{12} = R_{21} = R_{32} = 0.$$

$$T_0 = \frac{1}{3\sqrt{3}}\left[\sigma^{101}_{01,01}(\omega,\omega) + 2\sigma^{101}_{21,21}(\omega,\omega)\right],$$

$$T_{01} = \frac{\sqrt{2}}{9}\left[\Delta^{101}_{01,01}(\omega,\omega) + \tfrac{1}{2}\,\Delta^{101}_{21,21}(\omega,\omega) + \Delta^{011}_{11,01}(0,\omega) - \Delta^{011}_{11,21}(0,\omega)\right]. \quad \text{(A.4)}$$

(e) n^3P_1 — the ground states of Hg, Sr, ...:

$$R_{32} = 0, \quad \langle n1\|\boldsymbol{\mu}\|n1\rangle = \alpha\,\frac{3\sqrt{6}}{4}.$$

$$T_0 = -\tfrac{1}{3}\left[\sigma^{101}_{01,01}(\omega,\omega) + \tfrac{1}{2}\,\sigma^{101}_{21,21}(\omega,\omega) + \tfrac{3}{2}\,\sigma^{101}_{22,22}(\omega,\omega)\right],$$

$$T_1 = \tfrac{1}{6}\left[\Delta^{101}_{01,01}(\omega,\omega) + \tfrac{1}{2}\,\Delta^{101}_{21,21}(\omega,\omega) - \tfrac{3}{2}\,\Delta^{101}_{22,22}(\omega,\omega)\right],$$

$$T_2 = \tfrac{1}{6}\left[\sigma^{101}_{01,01}(\omega,\omega) + \tfrac{1}{2}\,\sigma^{101}_{21,21}(\omega,\omega) - \tfrac{3}{10}\,\sigma^{101}_{22,22}(\omega,\omega)\right].$$

$$T_{01} = \frac{1}{3\sqrt{6}}\left[\Delta^{101}_{01,01}(\omega,\omega) - \tfrac{1}{8}\,\Delta^{101}_{21,21}(\omega,\omega) - \tfrac{21}{8}\,\Delta^{101}_{22,22}(\omega,\omega) + \tfrac{3}{4}\,\Delta^{101}_{21,22}(\omega,\omega)\right.$$

$$\left. - \tfrac{5}{6}\,\Delta^{011}_{12,01}(0,\omega) + \tfrac{1}{12}\,\Delta^{011}_{12,21}(0,\omega) + \tfrac{3}{4}\,\Delta^{011}_{12,22}(0,\omega) - \tfrac{2}{3}\,\Delta^{011}_{10,01}(0,\omega) + \tfrac{2}{3}\,\Delta^{011}_{10,21}(0,\omega)\right],$$

$$T_{10} = -\frac{1}{3\sqrt{6}}\left[\sigma^{101}_{01,01}(\omega,\omega) - \tfrac{1}{8}\,\sigma^{101}_{21,21}(\omega,\omega) - \tfrac{3}{4}\,\sigma^{101}_{22,21}(\omega,\omega) - \tfrac{21}{8}\,\sigma^{101}_{22,22}(\omega,\omega)\right],$$

$$T_{12} = -\frac{1}{3\sqrt{6}}\left[\sigma^{101}_{01,01}(\omega,\omega) - \tfrac{1}{8}\,\sigma^{101}_{21,21}(\omega,\omega) - \tfrac{3}{10}\,\sigma^{101}_{21,22}(\omega,\omega) + \tfrac{21}{40}\,\sigma^{101}_{22,22}(\omega,\omega)\right.$$

$$\left. - \tfrac{3}{4}\,\sigma^{011}_{12,01}(0,\omega) + \tfrac{3}{40}\,\sigma^{011}_{12,21}(0,\omega) - \tfrac{9}{40}\,\sigma^{011}_{12,22}(0,\omega)\right],$$

$$T_{21} = \frac{1}{3\sqrt{6}}\left[\Delta^{101}_{01,01}(\omega,\omega) - \tfrac{1}{8}\,\Delta^{101}_{21,21}(\omega,\omega) + \tfrac{3}{10}\,\Delta^{101}_{21,22}(\omega,\omega) + \tfrac{21}{40}\,\Delta^{101}_{22,22}(\omega,\omega)\right.$$

$$\left. - \tfrac{1}{12}\,\Delta^{011}_{12,01}(0,\omega) + \tfrac{1}{120}\,\Delta^{011}_{12,21}(0,\omega) + \tfrac{3}{40}\,\Delta^{011}_{12,22}(0,\omega) - 4\Delta^{011}_{10,01}(0,\omega) + 4\Delta^{011}_{10,21}(0,\omega)\right]. \quad \text{(A.5)}$$

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