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Nonlinear variations in the Faraday effect caused in atomic systems by a strong magnetic field

N. L. Manakov and V. D. Ovsiannikov
Voronezh State University, 394063 Voronezh, Union of Soviet Socialist Republics

Stanisław Kielich
Nonlinear Optics Division, Institute of Physics of A. Mickiewicz University, 60-780 Poznań, Poland

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A theory of nonlinear variations in the Faraday effect caused by a strong magnetic field is proposed. In atomic gases, the variation is defined by the nonlinear magneto-optical susceptibility tensor component $\chi^{\mathrm{em}}_{xyzzz}$, which is expressed in terms of integrals of the radial Green's functions $G_E(r_1,r_2)$ of valence electrons of the atom. Applying for $G_E(r_1,r_2)$ and the wave functions analytical expressions in approximation of the model potential method, the authors carried out the first numerical calculations of $\chi^{\mathrm{em}}_{xyzzz}$ for inert-gas, alkali, and hydrogen atoms. The calculations show that the variations in the Verdet constant induced by the square of the magnetic field are sufficiently large (even in frequency regions far from resonance) for detection by the currently available strong-magnetic-field pulse technique.

I. INTRODUCTION

Modern laboratories have at their disposal the equipment necessary to produce, on the one hand, strong magnetic fields and, on the other, intense oscillating electric fields, such as those conveyed by laser beams. The conditions are thus fulfilled for the experimental observation of various nonlinear magneto-electro-optical effects.[1,2] In this paper we concentrate exclusively on variations in the Faraday effect caused by intense laser light[3,4] and a strong magnetic field.[4,5] The laser beam intensity-dependent Faraday effect has been analyzed theoretically for gases and liquids[4,6] as well as crystals.[2,3] However, it is only recently that numerical calculations of the nonlinear electronic part of Verdet's constant have been performed, by Yu and Osborn,[7] for the simple model of a two-level molecule with one-photon transitions, and by Kielich, Manakov, and Ovsiannikov[8] for a general model with one- and two-photon transitions by the potential model method for radial Green's functions of the optical electrons.[9-12] The calculations, carried out by the last method for inert-gas, alkali, and hydrogen atoms, prove that the nonlinear variations in the Faraday effect can be observed in experiment, particularly within the nonlinear resonance region for frequencies ω and 2ω.

In this paper we carry out calculations of the nonlinear variations in the Faraday effect induced by a very strong magnetic field in atomic gases.

II. PHENOMENOLOGICAL THEORY

To begin, we deal with the problem in a phenomenological approach, starting from the equation for the electric permittivity tensor at frequency ω and wave vector $\vec{k}$ of the electromagnetic field:

$$[\epsilon_{ij}(\omega,\vec{k})-\delta_{ij}]E_j(\omega,\vec{k})=4\pi P_i(\omega,\vec{k}),$$

where $\vec{P}(\omega,\vec{k})$ is the vector of the electric polarization, induced in the medium by an electromagnetic field with the electric vector $\vec{E}(\omega,\vec{k})$ and magnetic vector $\vec{H}(\omega,\vec{k})$.

Similarly, the equation for the magnetic permittivity tensor is

$$[\mu_{ij}(\omega,\vec{k})-\delta_{ij}]H_j(\omega,\vec{k})=4\pi M_i(\omega,\vec{k}),$$

where $\vec{M}(\omega,\vec{k})$ is the vector of the magnetic polarization, induced generally by the field vectors $\vec{E}(\omega,\vec{k})$ and $\vec{H}(\omega,\vec{k})$.

Since in a medium with refractive index n the wave vector $\vec{k}=n(\omega/c)\vec{s}$ for a wave propagating in the direction $\vec{s}$, Maxwell's equations for the medium are, in tensor notation,

$$n(\omega,\vec{k})\delta_{ijk}s_jE_k(\omega,\vec{k})=\mu_{ij}(\omega,\vec{k})H_j(\omega,\vec{k}),$$
$$-n(\omega,\vec{k})\delta_{ijk}\,s_jH_k(\omega,\vec{k})=\epsilon_{ij}(\omega,\vec{k})E_j(\omega,\vec{k}),$$

where δ_{ijk} is the antisymmetric Levi-Cività tensor.

We assume the medium in the absence of external fields as isotropic, with the scalar permittivities ϵ and μ. If a static magnetic field $H(0)$ is applied externally to the medium along the axis z of laboratory coordinates, its electric permittivity takes the form

$$(\epsilon_{ij})=\begin{bmatrix}\epsilon & \epsilon_{xy} & 0\\ -\epsilon_{xy} & \epsilon & 0\\ 0 & 0 & \epsilon\end{bmatrix}.$$

The magnetic permittivity tensor is similar.

With these assumptions Maxwell's equations lead with a satisfactory approximation to the following expression for the difference in refractive indices for waves circularly polarized in the negative and positive senses:

$$n_-(\omega,\vec{k})-n_+(\omega,\vec{k}) = i[(\mu/\epsilon)^{1/2}\epsilon_{xy}(\omega,\vec{k})+(\epsilon/\mu)^{1/2}\mu_{xy}(\omega,\vec{k})].$$

By definition, the rotation angle of the polarization plane per unit length of the medium is

$$\Theta(\omega,k)=\tfrac{1}{2}(\omega/c)[n_-(\omega,\vec{k})-n_+(\omega,\vec{k})],$$

whence for a wave propagating in the z direction we obtain the following formula, of relevance to our further considerations:

$$\Theta(\omega,k)=2\pi i\left(\frac{\omega}{c}\right)\left[\left(\frac{\mu}{\epsilon}\right)^{1/2}\frac{\partial P_x(\omega,\vec{k})}{\partial E_y(\omega,\vec{k})} +\left(\frac{\epsilon}{\mu}\right)^{1/2}\frac{\partial M_x(\omega,\vec{k})}{\partial H_y(\omega,\vec{k})}\right]. \tag{1}$$

Formula (1) is valid with the assumption that the polarization vectors $\vec{P}(\omega,\vec{k})$ and $\vec{M}(\omega,\vec{k})$ are linear functions of the light vectors $\vec{E}(\omega,\vec{k})$ and $\vec{H}(\omega,\vec{k})$, whereas their dependence on the strength of the static magnetic field $H(0)$ can be, in general, nonlinear. $\vec{P}(\omega,\vec{k})$ and $\vec{M}(\omega,\vec{k})$ contain in addition to the dipolar terms, quite generally all higher multipolar electric as well as magnetic contributions,[3] which, however, are related to spatial dispersion, a subject beyond the scope of this paper.[13] Restricting our considerations to the dipolar electric and magnetic approximation, we write the vector component of electric polarization in the form

$$\begin{aligned}P_i(\omega)=[&\chi^{\mathrm{e}}_{ij}(-\omega,\omega)+\chi^{\mathrm{em}}_{ijk}(-\omega,\omega,0)H_k(0)\\ &+\chi^{\mathrm{em}}_{ijkl}(-\omega,\omega,0,0)H_k(0)H_l(0)\\ &+\chi^{\mathrm{em}}_{ijkln}(-\omega,\omega,0,0,0)H_k(0)H_l(0)H_n(0)\\ &+\cdots]E_j(\omega),\end{aligned} \tag{2}$$

neglecting the terms involving linear dependence on the magnetic light vector $\vec{H}(\omega)$.

The second-rank tensor χ^{e}_{ij} defines linear electric susceptibility; the third-rank pseudotensor χ^{em}_{ijk}, magnetoelectric susceptibility, defining the linear Faraday effect; the fourth-rank tensor $\chi^{\mathrm{em}}_{ijkl}$, the Cotton-Mouton effect and magnetostriction; but the fifth-rank pseudotensor $\chi^{\mathrm{em}}_{ijkln}$ of nonlinear magnetoelectric susceptibility defines the nonlinear variations in the Faraday effect which are the subject of our investigation.

An expansion similar to that of the electric polarization (2) can be written for the magnetic polarization. Since in the case of isotropic diamagnetics its contribution to the rotation angle (1) is negligible, we refrain from considering it here.

By (1), (2), and the definition of the Verdet constant,

$$V(\omega)=\tfrac{1}{2}(\omega/c)[n_-(\omega)-n_+(\omega)]/H_z(0),$$

we obtain in the presence of a strong static magnetic field

$$\begin{aligned}V(\omega,H_0)=i[2\pi/n(\omega)](\omega/c)&\\ \times[\chi^{\mathrm{em}}_{xyz}(-\omega,\omega,0)&\\ +\chi^{\mathrm{em}}_{xyzzz}(-\omega,\omega,0,0,0)H_0^2+\cdots].&\end{aligned} \tag{3}$$

The preceding phenomenological formula describes the variation in the Verdet constant induced by the second power of the magnetic field. We now work out a microscopic theory permitting the numerical calculation of the nonlinear magnetoelectric susceptibility component $\chi^{\mathrm{em}}_{xyzzz}$. Like χ^{em}_{xyz}, the nonlinear susceptibility in general consists, in addition to a part not directly dependent on temperature, of temperature-dependent terms, related to, e.g., reorientation of the molecules in the magnetic field.[5,6] However, we restrict our quantum-mechanical calculations to atomic gases, where in the absence of internal interaction, no temperature-dependent terms appear.

III. MICROSCOPIC THEORY AND NUMERICAL RESULTS FOR ATOMIC GASES

In order to derive the quantum-mechanical formula for the magnetoelectric susceptibility tensor $\chi^{\mathrm{em}}_{ijklm}$, we use the Hamiltonian of the dipole approximation,

$$H=H_0-\hat{d}_iE_i(t)-\hat{m}_iH_i(0)-\tfrac{1}{2}\hat{\chi}^{\mathrm{d}}_{ij}H_i(0)H_j(0)-\cdots, \tag{4}$$

where $\hat{d}$ and $\hat{m}$ are respectively the electric and magnetic moment operators and[14]

$$\hat{\chi}^{\mathrm{d}}_{ij}=\sum_{\nu}\frac{e_\nu^2}{4m_\nu c^2}(r_{\nu i}r_{\nu j}-r_\nu^2\delta_{ij}) \tag{5}$$

is the diamagnetic susceptibility tensor operator for a system of spinless particles, with charge e_ν and mass m_ν.

When we calculate the mean value of the electric moment $\vec{P}(\omega)$ up to terms $\sim EH^3$, by analogy with the case of an intense light field[8] we can write $\chi^{\mathrm{em}}_{ijklm}$ in the form of the sum

$$\chi^{\mathrm{em}}_{ijklm}(-\omega,\omega,0,0,0)=\chi^{\mathrm{p}}_{ijklm}+\chi^{\mathrm{d}}_{ijklm},$$

with χ^{p} being the component determined by magneticdipole interaction and χ^{d} being that determined by interference of magnetodipole and diamagnetic interactions. For a system in the nondegenerate state $|0\rangle$ with energy E_0, we obtain

$$\chi^{\mathrm{p}}_{xyzzz}=\langle 0|\hat{d}_xG_{E_0+\omega}\hat{m}_zG_{E_0+\omega}\hat{m}_zG_{E_0+\omega}\hat{m}_zG_{E_0+\omega}\hat{d}_y|0\rangle+\langle 0|\hat{d}_yG_{E_0-\omega}\hat{m}_zG_{E_0-\omega}\hat{m}_zG_{E_0-\omega}\hat{m}_zG_{E_0-\omega}\hat{d}_x|0\rangle, \tag{6}$$

$$\begin{aligned}\chi^{\mathrm{d}}_{xyzzz} = {} & \langle 0|\hat{d}_x G_{E_0+\omega}\hat{m}_z G_{E_0+\omega}\hat{d}_y G_{E_0}\hat{\chi}^{\mathrm{d}}_{zz}|0\rangle + \langle 0|\hat{d}_y G_{E_0-\omega}\hat{m}_z G_{E_0-\omega}\hat{d}_x G_{E_0}\hat{\chi}^{\mathrm{d}}_{zz}|0\rangle \\ & + \langle 0|\hat{\chi}^{\mathrm{d}}_{zz} G_{E_0}\hat{d}_x G_{E_0+\omega}\hat{m}_z G_{E_0+\omega}\hat{d}_y|0\rangle + \langle 0|\hat{\chi}^{\mathrm{d}}_{zz} G_{E_0}\hat{d}_y G_{E_0-\omega}\hat{m}_z G_{E_0-\omega}\hat{d}_x|0\rangle \\ & + \langle 0|\hat{d}_x G_{E_0+\omega}\hat{m}_z G_{E_0+\omega}\tilde{\chi}^{\mathrm{d}}_{zz} G_{E_0+\omega}\hat{d}_y|0\rangle + \langle 0|\hat{d}_y G_{E_0-\omega}\hat{m}_z G_{E_0-\omega}\tilde{\chi}^{\mathrm{d}}_{zz} G_{E_0-\omega}\hat{d}_x|0\rangle \\ & + \langle 0|\hat{d}_x G_{E_0+\omega}\tilde{\chi}^{\mathrm{d}}_{zz} G_{E_0+\omega}\hat{m}_z G_{E_0+\omega}\hat{d}_y|0\rangle + \langle 0|\hat{d}_y G_{E_0-\omega}\tilde{\chi}^{\mathrm{d}}_{zz} G_{E_0-\omega}\hat{m}_z G_{E_0-\omega}\hat{d}_x|0\rangle\,, \end{aligned} \tag{7}$$

where $\tilde{\chi}^{\mathrm{d}}_{zz} = \hat{\chi}^{\mathrm{d}}_{zz} - \langle 0|\hat{\chi}^{\mathrm{d}}_{zz}|0\rangle$. Above, $G_{E_0\pm\omega}$ is a Green's function, accounting for summation over intermediate states. Taking into consideration interaction of the field and the external optical electron only, and on performing integrations in (6) and (7) over angular coordinates in the LS-coupling picture, we obtain χ^{p} and χ^{d} as follows in terms of the radial matrix elements

$$\begin{aligned}&\rho^{n_1 n_2,\cdots,n_{N+1}}_{L_1,L_2,\cdots,L_N}(\omega_1,\omega_2,\cdots,\omega_N) \\ &\qquad = \langle 0|r_1^{n_1} g_{L_1}(E_0+\omega_1; r_1, r_2) r_2^{n_2} g_{L_2}(E_0+\omega_2; r_2, r_3)\cdots r_N^{n_N} g_{L_N}(E_0+\omega_N; r_N, r_{N+1}) r_{N+1}^{n_{N+1}}|0\rangle\,,\end{aligned}$$

with the radial part of the Green's function $g_L(\boldsymbol{E}; r, r')$ given by formula (A2) of the Appendix:

$$\chi^{\mathrm{p}} = i\frac{\alpha^3}{24}\Delta^{10001}_{1111}(\omega,\omega,\omega,\omega)\,,$$

$$\begin{aligned}\chi^{\mathrm{d}} = i\frac{\alpha^3}{180}\{ & 6\Delta^{1021}_{111}(\omega,\omega,\omega) + \Delta^{1021}_{112}(\omega,\omega,0) \\ & + 5[\Delta^{1012}_{110}(\omega,\omega,0) - \overline{r^2}\Delta^{1001}_{111}(\omega,\omega,\omega)]\}\,,\end{aligned} \tag{8}$$

with $\alpha = \frac{1}{137}$. Here

$$\overline{r^2} = \langle 0|r^2|0\rangle\,,$$

$$\begin{aligned}&\Delta^{\cdots}_{\cdots}(\omega_1,\omega_2,\ldots,\omega_N) \\ &\quad = \rho^{\cdots}_{\cdots}(\omega_1,\omega_2,\ldots,\omega_N) - \rho^{\cdots}_{\cdots}(-\omega_1,-\omega_2,\ldots,-\omega_N)\,.\end{aligned}$$

With the preceding expressions the Verdet constant in a strong magnetic field takes the form (in atomic units)

$$V(\omega) = \pi N_0\alpha^2\omega\{Y_0(\omega) + (\alpha H)^2[Y_2^{\mathrm{p}}(\omega) + Y_2^{\mathrm{d}}(\omega)]\}\,, \tag{9}$$

as in the case of an intense light field.[8] In addition,

$$Y_2^{\mathrm{p(d)}}(\omega) = -i(2/\alpha^3)\chi^{\mathrm{p(d)}}.$$

The quantity $Y_0(\omega)$ is defined and calculated in Ref. 8. The numerical values of $Y_2(\omega)$, as well as all details of their calculation, are given in the Appendix.

The poles of Y_0 and Y_2 coincide with the energies of the P states of the atom. The stronger frequency dependence of $Y_2(\omega)$ compared with $Y_0(\omega)$ causes the correction terms in (9) near resonance to be significant in fields of 10^4–10^5 Oe. The diamagnetic term $Y_2^{\mathrm{d}}(\omega)$ can be neglected near the poles, since χ^{d} contains three resonance denominators, whereas χ^{p} contains four [cf. Eqs. (6) and (7)]. It is worth noting that in Ref. 15 the influence of diamagnetic interaction χ^{d}_{ij} is altogether omitted when considering corrections to Verdet's constant in a strong magnetic field—an omission which is hardly justified far from resonance (in the region of optical transparency). In particular, for the hydrogen atom in the optical region, Y_2^{d} is two or three times larger than Y_2^{p}. For atoms having a low ionization potential(e.g., the alkali metals) one has, as a rule, $|Y_2^{\mathrm{d}}| \ll |Y_2^{\mathrm{p}}|$, although, far from resonance, the contribution of Y_2^{d} also can be significant (cf. Table I). Figure 1 shows graphs of Y_2^{p} and Y_2^{d} vs frequency for the ground state of hydrogen.

$Y_2^{\mathrm{d}}(\omega)$ undergoes a change in sign as ω passes through resonance. Hence the function has a zero in each inter-resonance interval. In the case of the hydrogen atom the first zero of $Y_2^{\mathrm{d}}(\omega_0)$ corresponds to the frequency $\omega_0 = 90\,200$ cm^{-1}. Y_2^{p} is positive everywhere and $Y_2^{\mathrm{p}} \gg |Y_2^{\mathrm{d}}|$ except for a narrow region in the vicinity of the minimum of Y_2^{p}, where Y_2^{d} exceeds Y_2^{p} by a factor of 1.5–2.

In a strong light field, $V(\omega)$ can be written in the form (cf. Ref. 8)

$$\begin{aligned}V(\omega) = \pi N_0\alpha^2\omega\{ & Y_0(\omega) + |E(\omega)|^2 Y_1(\omega) \\ & + [\alpha H(0)]^2 Y_2(\omega)\}\,,\end{aligned} \tag{10}$$

with $Y_2(\omega) = Y_2^{\mathrm{p}} + Y_2^{\mathrm{d}}$ and $Y_1(\omega)$ calculated in Ref. 8. A comparison of the numerical values of $Y_1(\omega)$ and $Y_2(\omega)$ shows that for equal field strengths $E(\omega)$ and $H(0)$ the effects of the electric field are stronger than those of the magnetic field inasmuch as in the region of frequencies comparable with the excitation frequency of the atom, $Y_1(\omega)$ exceeds $Y_2(\omega)$ 10 to 10^3 times.

It should be noted that our calculations are inapplicable at exact resonance, at which point the two-level resonance approximation is used. So for the results obtained above to be usable, the mistuning of ω from the eigenfrequencies of the atom must be essentially more than the resonance levels widths, or should have a value $\gtrsim 10$ cm^{-1}.

Concerning the possibility of an experimental identification of our nonlinear corrections, we point out that for the case of resonance mistuning ~100 cm^{-1} the correction to the Verdet constant is about 10% for a magnetic field strength of

TABLE I. Nonlinear corrections to Verdet's constant[a] in a strong magnetic field, for atomic systems from Eq. (9): $V(\omega)=\pi N_0\alpha^2\omega[Y_0+(\alpha H)^2(Y_2^{\mathrm{p}}+Y_2^{\mathrm{d}})]$.

ω	ω_N		ω_R		$2\omega_N$		$2\omega_R$	
Atom	Y_2^{p}	Y_2^{d}	Y_2^{p}	Y_2^{d}	Y_2^{p}	Y_2^{d}	Y_2^{p}	Y_2^{d}
H	7.885	27.34	13.08	44.12	19.10	62.18	41.52	120.8
He	0.098	0.476	0.152	0.736	0.204	0.980	0.334	1.56
Li	3.61×10^6	8.77×10^5	5.01×10^{10}	1.06×10^9	1.29×10^7	-2.03×10^6	1.24×10^5	3.92×10^4
Na	1.03×10^6	5.21×10^5	7.65×10^7	1.33×10^7	2.54×10^8	-3.27×10^7	1.04×10^7	9.59×10^6
K	3.18×10^7	7.22×10^6	1.26×10^9	-1.15×10^8	4.19×10^6	-1.44×10^6	6.09×10^{10}	1.31×10^{10}
Rb	4.24×10^7	9.60×10^6	8.77×10^8	-9.83×10^7	4.17×10^6	-1.34×10^6	6.85×10^6	-2.77×10^6
Cs	2.25×10^8	3.86×10^7	1.23×10^8	-2.53×10^7	3.21×10^6	1.01×10^6	7.53×10^9	-1.92×10^8

[a] Numerical values (in atomic units) of the corrections Y_2^{p} and Y_2^{d} for alkali, hydrogen, and helium atoms at laser frequencies and their harmonics: $\omega_N=9440\ \mathrm{cm}^{-1}$, neodymium laser; $\omega_R=14\,400\ \mathrm{cm}^{-1}$, ruby laser. To obtain $V(\omega)$ in units of μmin/Oe cm, $V(\omega)_{\mathrm{a.u.}}$ must be multiplied by the conversion factor:

$$\frac{10.8}{(3.1416)(1.714)(0.52917)}\times10^{10} .$$

When calculating $V(\omega)_{\mathrm{a.u.}}$, we must take all quantities in the formula for $V(\omega)$ in atomic units: $N_0\ (\mathrm{a.u.})=N_0\ (\mathrm{cm}^{-3})a_0^3$, $\omega\ (\mathrm{a.u.})=\omega\ (\mathrm{cm}^{-1})\ /2.195\times10^5$, $H\ (\mathrm{a.u.})=H\ (\mathrm{Oe})\ /1.714\times10^7$, and Bohr radius $a_0=5.29\times10^{-9}$ cm.

$H\simeq10^5$ Oe. In the typical nonresonant situations to which the data of Table I relate, a change of 10% is achieved in a field of $H\simeq10^6$ Oe. From the above and our previous results (Ref. 8) it follows that the nonlinear corrections to the Faraday effect in atomic gases predicted here are well accessible to experimental observation with the optical electric and static magnetic field strengths now at the disposal of most laboratories.

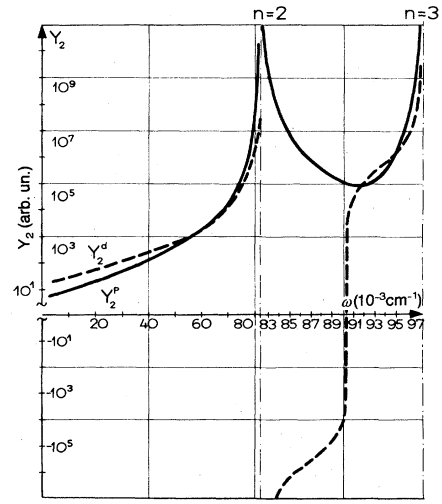

FIG. 1. Nonlinear corrections $Y_2^p(\omega)$ and $Y_2^d(\omega)$ to Verdet's constant as functions of frequency, for the hydrogen atom in its ground state. (Dot-dashed curve, frequency $\omega_{\mathrm{res}}=82\,303\ \mathrm{cm}^{-1}$ of the 1s-2p transition). For $\omega>\omega_{\mathrm{res}}$ the scale is magnified five-fold.

APPENDIX: PROCEDURE OF NUMERICAL CALCULATION OF COMPOUND MATRIX ELEMENTS IN THE APPROXIMATION OF THE MODEL POTENTIAL METHOD

In numerical calculations of matrix elements in higher orders of perturbation theory, use is made of the model potential method (MPM) developed in Refs. 9–11 for the description of states of the valence electron in the atom. In the MPW the wave functions and radial Green's function $g_\gamma^E(r,r')$ are of the form

$$R_{n\gamma}(r)=(2Z^{3/2}/\nu_{n\gamma}^2)f_{n\gamma}(2Zr/\nu_{n\gamma}) , \tag{A1}$$

where $n=0,1,2,\ldots$ is a radial quantum number and

$$g_\gamma^E(r,r')=\frac{4Z}{\nu}\sum_{k=0}^{\infty}\frac{f_{k\gamma}(2Zr/\nu)f_{k\gamma}(2Zr'/\nu)}{1+k+\lambda_\gamma-\nu} , \tag{A2}$$

where

$$f_{k\gamma}(x)=\left(\frac{k!}{\Gamma(k+2\lambda_\gamma+2)}\right)^{1/2}x^{\lambda_\gamma}e^{-x/2}L_k^{2\lambda_\gamma+1}(x) ,$$

$$\nu=Z/(-2E)^{1/2} ,$$

and $L_k^\alpha(x)$ is a Laguerre polynomial; the parameter λ_γ is determined for $E=E_{n\gamma}$ from the experimentally available spectrum of the atom $E_{n\gamma}$, whereas for $E\neq E_{n\gamma}$ it is determined by interpolation from known values $\lambda_\lambda(E_{n\gamma})$.

Applying (A1) and (A2) and well-known integrals of products of Laguerre polynomials, we express the composite matrix element ρ of Eq. (8) in the following form:

$$\rho_{n_f\,'\gamma_1\gamma_2\cdots\gamma_{N_1}-n_i}^{n_1 n_2\cdots n_{N+1}}(\omega_1\omega_2\cdots\omega_N)=(Z^2/\nu_f\nu_i^2)S(N+1,n_i)\,, \tag{A3}$$

where the function S can be calculated from the recurrent formula

$$S(p,k_p)=\sum_{k_{p-1}=0}^{\infty}\frac{S(p-1,k_{p-1})\Phi_{k_{p-1}k_p}^{n_p}(\nu_{p-1}\lambda_{p-1};\nu_p\lambda_p)}{1+k_{k-1}+\lambda_{p-1}-\nu_{p-1}}\,,\quad p=2,3,\ldots,N+1,\quad S(1,k_1)=\Phi_{k_0k_1}^{n_1}(\nu_0\lambda_0;\nu_1\lambda_1)\,. \tag{A4}$$

Above

$$\begin{aligned}\Phi_{kk'}^{n}(\nu\lambda;\nu'\lambda')&\equiv\frac{4Z}{\nu}\left\langle f_{k\gamma}\left(\frac{2Zr}{\nu}\right)\middle|r^n\middle|f_{k'\gamma'}\left(\frac{2Zr}{\nu'}\right)\right\rangle\\&=\frac{1}{4Z^3\nu}\left(\frac{\nu\gamma'}{Z(\nu+\nu')}\right)^{n-1}\left(\frac{2\nu}{\nu+\nu'}\right)^{\lambda'+2}\left(\frac{2\nu'}{\nu+\nu'}\right)^{\lambda+2}\frac{\Gamma(\lambda+\lambda'+3+n)}{\Gamma(2\lambda+2)\Gamma(2\lambda'+2)}\left(\frac{\Gamma(k+2\lambda+2)\Gamma(k'+2\lambda'+2)}{k!k'!}\right)^{1/2}\\&\quad\times F_2\left(\lambda+\lambda'+3+n;-k,-k';2\lambda+2,2\lambda'+2;\frac{2\nu'}{\nu+\nu'};\frac{2\nu}{\nu+\nu'}\right);\end{aligned} \tag{A5}$$

and $k_{N+1}=n_i,\nu_{N+1}=\nu_i,\lambda_{N+1}=\lambda_i$ and $k_0=n_f,\nu_0=\nu_f,\lambda_0=\lambda_f$ are the radial quantum number, "effective" principal quantum number, and orbital parameter of respectively the initial and final states of the atom. ν_p, λ_p, with $p\neq 0,N+1$ are analogical quantities for the pth Green's function in (A3). F_2 is a generalized hypergeometrical series of two variables (Appel function[16]) which in our case is of the form of a double finite sum over the indices $-k$ and $-k'$. In our calculations, use is made of the expression of F_2 in terms of a finite sum of hypergeometrical functions ${}_2F_1$:

$$\begin{aligned}&F_2(\alpha;-k,-k';\beta,\beta';x,x')\\&\quad=\sum_{m=0}^{k}\frac{(-k)_m(\alpha)_m}{(\beta)_m m!}\,\chi^{\mathrm{m}}\,{}_2F_1(\alpha+m,-k';\beta';x')\,.\end{aligned}$$

The polynomials ${}_2F_1$ are calculated using relationships recurrent in both upper indices.[16]

If the energy of one of the Green's functions of Eqs. (6) and (7) coincides with that of the initial or final state of the atom, the term corresponding to the state in question does not occur in the expansion (A2), and supplementary terms appear. A reduced Green's function of this kind is derived explicitly in Ref. 10. The relation (A4) in this situation undergoes a modification. Thus, if in $\rho_{\vdots\vdots}^{\vdots}$ the function $g_{\gamma q}^{E_2}(r_q,r_{q+1})$ is a reduced one, (A4) must be replaced for $p=q+1$ by the following expression (with $n=n_i$ or n_f, depending on the state with whose energy that of the Green's function coincides):

$$\begin{aligned}S(q+1,k_{q+1})=\sum_{k_q\neq n}&\frac{S(q,k_q)\Phi_{k_qk_{q+1}}^{n_{q+1}}(\nu_q\lambda_q;\nu_{q+1}\lambda_{q+1})}{1+k_q+\lambda_q-\nu_q}\\&+S(q,n)\{[(5/2\nu_q)+2\lambda_q]\Phi_{nk_{q+1}}^{n_{q+1}}(\nu_q\lambda_q;\nu_{q+1}\lambda_{q+1})-(Z/\nu_q)\Phi_{nk_{q+1}}^{n_{q+1}+1}(\nu_q\lambda_q;\nu_{q+1}\lambda_{q+1})\\&\qquad-(2Z\nu_qn)^{1/2}\Phi_{n-1k_{q+1}}^{n_{q+1}+1/2}(\nu_q\lambda_q+\tfrac{1}{2};\nu_{q+1}\lambda_{q+1})\}\\&-[(Z/\nu_q)S_1(q,n)+(2Z\nu_qn)^{1/2}S_2(q,n)]\Phi_{nk_{q+1}}^{n_{q+1}}(\nu_q\lambda_q;\nu_{q+1}\lambda_{q+1})\,.\end{aligned} \tag{A6}$$

The quantities

$$\begin{aligned}S_1(q,n)&\equiv\sum_{k_{q-1}=0}^{\infty}\frac{S(q-1,k_{q-1})\Phi_{k_{q-1}n}^{n_{q+1}}(\nu_{q-1}\lambda_{q-1};\nu_q\lambda_q)}{1+k_{q-1}+\lambda_{q-1}-\nu_{q-1}}\,,\\S_2(q,n)&\equiv\sum_{k_{q-1}=0}^{\infty}\frac{S(q-1,k_{q-1})\Phi_{k_{q-1}n-1}^{n_{q+1/2}}(\nu_{q-1}\lambda_{q-1};\nu_q\lambda_q+\frac{1}{2})}{1+k_{q-1}+\lambda_{q-1}-\nu_{q-1}}\end{aligned} \tag{A7}$$

must be calculated simultaneously with $S(q,n)$ in the preceding step of utilization of the formula (A6).

The expressions (A4)–(A7) permit the calculation of composite matrix elements of arbitrary order for transitions between discrete states of the atom. The data for the calculations comprise the sets of energy values of the initial, final, and intermediate states (E_p), the related orbital parameters $\lambda_p(p=0,1,\ldots,N+1)$, and exponents of the power of the radius r,n_j $(j=1,2,\ldots,N+1)$. A similar recurrent procedure for the calculation of matrix elements, defining the probabilities of many-photon ionization of atoms and not involving reduced Green's functions, has been applied in Ref. 17. Calculations of $Y_2(\omega)$ in (9) for atomic ground states ($n_i = n_f = 0$) show that it is sufficient to take into consideration the first ($\nu_p + 4$) terms in the sum over k_p to obtain the numerical value of the series (A4) with an accuracy of 0.01%. The same holds for the series (A6) and (A7).

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