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MULTIPOLE EXPANSION OF THE RETARDED INTERATOMIC POTENTIAL ENERGY

V. DERIVATION WITH THE USE OF A DISPERSION RELATION

L.G. SUTTORP*

International Centre for Theoretical Physics, Trieste, Italia

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Synopsis

A dispersion relation technique is used to obtain the retarded dispersion energy of two neutral atoms in nondegenerate ground states. To that end multipole expansions are derived for the electromagnetic form factors occurring in the vertex function that characterizes the interaction of a neutral atom with a pair of photons on the mass shell. The long-range asymptotic expression for the dispersion energy is shown to contain electric and diamagnetic matrix elements in a symmetric way.

1. *Introduction.* In previous papers[1]) of this series the retarded dispersion energy of two neutral atoms in their ground states has been obtained by calculating, with the help of perturbation theory up to fourth order, the energy shift due to the interaction of the atoms with the radiation field. Recently, Feinberg and Sucher[2]) showed how dispersion-relation techniques may be used to evaluate the two-photon exchange contribution to the interaction energy of neutral particles. In fact, they found, by assuming analytical properties for certain functions and introducing a generalized electric dipole approximation, an expression for the interaction energy that contains the properties of the particles in the form of several spectral functions. Since these particles were not specified in more detail explicit expressions for these spectral functions could not be given.

In the present paper these dispersion-relation methods are applied to the derivation of the multipole expansion of the interatomic dispersion energy. By making use of the knowledge about the atomic structure the analytical properties of the Fourier transform of the dispersion energy may be studied in detail. As a result a dispersion relation can be written down for this Fourier transform, with a weight function containing the atomic vertex functions characterizing the emission of a

* On leave of absence from the Instituut voor Theoretische Physica, Universiteit van Amsterdam.

pair of real photons. These atomic vertex functions are determined by the electric and magnetic form factors of the atoms for which expansions in terms of multipole matrix elements may be derived by considering the emission of photons with definite helicities. In this way the explicit dependence of the form factors on the energy and the total momentum of the photons will be found. The interatomic dispersion energy will be obtained subsequently by evaluation of the dispersion integral for its Fourier transform. The expression derived in this way may be shown to be equivalent to that found in the previous papers of this series. The present treatment, however, exhibits clearly the symmetry between electric and magnetic phenomena. In particular, it follows that the long-range asymptotic form of the dispersion energy contains electric and (dia)magnetic matrix elements in a symmetric way; indeed, the electric quadrupole terms found earlier can be rewritten, with the help of a sum rule, so as to yield the diamagnetic contribution to the dispersion energy.

2. *The interatomic interaction energy due to two-photon exchange.* Let us consider two neutral one-electron atoms a and b with fixed nuclei and interacting with each other *via* the electromagnetic field coupled to both electrons and nuclei. Treating this interaction as a perturbation one finds* for the contribution in the scattering matrix due to the exchange of two photons between the electrons, with charges $-e$:

$$\begin{aligned} S_{\mathrm{fi}} = e^4 \int \mathrm{d}^4R_1 \cdots \mathrm{d}^4R_4 \bar{\psi}_{\alpha_\mathrm{f}}(R_1)\gamma^\mu S_{\mathrm{F},a}(R_1, R_2)\gamma^\nu \psi_{\alpha_\mathrm{i}}(R_2) \\ \times \bar{\psi}_{\beta_\mathrm{f}}(R_3)\gamma^{\mu'} S_{\mathrm{F},b}(R_3, R_4)\gamma^{\nu'}\psi_{\beta_\mathrm{i}}(R_4) \\ \times \{g_{\mu\mu'}g_{\nu\nu'}D_\mathrm{F}(R_1 - R_3)D_\mathrm{F}(R_2 - R_4) + g_{\mu\nu'}g_{\nu\mu'}D_\mathrm{F}(R_1 - R_4) \\ \times D_\mathrm{F}(R_2 - R_3)\}, \end{aligned} \tag{1}$$

where the initial electron states are characterized by Dirac eigenfunctions ψ_{α_i}, ψ_{β_i} and the final states by ψ_{α_f}, ψ_{β_f}. In writing the expression (1) we supposed the wave functions of the electrons to have no appreciable overlap, so that electronic exchange could be neglected. The propagator for the electron of atom a in the electrostatic field of its nucleus is given by[3])

$$S_{\mathrm{F},a}(R_1, R_2) = -\frac{1}{2\pi}\sum_\alpha \psi_\alpha(\boldsymbol{R}_1)\bar{\psi}_\alpha(\boldsymbol{R}_2)\int_{-\infty}^{\infty}\mathrm{d}\omega_a \frac{\mathrm{e}^{\mathrm{i}\omega_a(t_1 - t_2)}}{E_\alpha(1 - \mathrm{i}0) + \omega_a}, \tag{2}$$

where α labels all positive- and negative-energy eigenfunctions $\psi_\alpha(R) = \psi_\alpha(\boldsymbol{R}) \times \exp(-\mathrm{i}E_\alpha t)$ of the electron in the field of its nucleus. The photon propagator in (1)

* The metric tensor $g^{\mu\nu}$ with components $g^{00} = -g^{ii} = 1$ $(i = 1, 2, 3)$, $g^{\mu\nu} = 0$ $(\mu \neq \nu)$ is chosen. Four-tensor indices run from 0 to 3, while summation over repeated indices is understood. The Dirac matrices γ^μ satisfy the anticommutation relations $\{\gamma^\mu, \gamma^\nu\} = 2g^{\mu\nu}$. Rationalized gaussian units, with $\hbar$ and c put equal to unity, are used throughout.

is defined as:

$$D_F(R_1 - R_2) = -\frac{1}{(2\pi)^4}\int d^4k\,\frac{e^{-ik\cdot(R_1-R_2)}}{k^2 + i0}. \tag{3}$$

The complete two-photon exchange contribution to the scattering matrix contains moreover terms describing the photon exchange between the nucleus of one atom and the electron or nucleus of the other. These terms will be added in a later stage of the treatment.

If the propagators (2) and (3) are inserted into (1) and the integrations over $t_i = R_i^0$ $(i = 1, \ldots, 4)$ and ω_a, ω_b are carried out one finds that the scattering matrix may be written as:

$$S_{fi} = -2\pi i\delta\,(E_{\alpha_f} + E_{\beta_f} - E_{\alpha_i} - E_{\beta_i})\,V_{fi}. \tag{4}$$

For equal initial and final states the quantity $V_{fi} = V$ may be interpreted[4]) as the interaction energy of the atoms due to two-photon exchange between the electrons. After introduction of relative coordinates $\boldsymbol{r}_a$, $\boldsymbol{r}_b$ of the electrons with respect to their nuclei, with position vectors $\boldsymbol{R}_a$, $\boldsymbol{R}_b$, the interaction energy is found to depend on the separation $\boldsymbol{R} = \boldsymbol{R}_b - \boldsymbol{R}_a$ in the following way:

$$V(\boldsymbol{R}) = (2\pi)^{-3}\int d\boldsymbol{Q}\,e^{i\boldsymbol{Q}\cdot\boldsymbol{R}}F(\boldsymbol{Q}), \tag{5}$$

where the Fourier transform has the form:

$$F(\boldsymbol{Q}) = \frac{i}{2(2\pi)^4}\int d^4k\,d^4k'\,\Gamma_{a,\mu\nu}(k, k')\,\Gamma_b^{\mu\nu}(-k, -k') \times \frac{\delta(k^0 + k'^0)\,\delta(\boldsymbol{k} + \boldsymbol{k}' - \boldsymbol{Q})}{(k^2 + i0)\,(k'^2 + i0)}. \tag{6}$$

The vertex function $\Gamma_a^{\mu\nu}$ appearing here describes the emission of a pair of virtual photons with energy-momenta k^μ, k'^μ satisfying $k^0 = -k'^0$, by the electron of atom a:

$$\Gamma_a^{\mu\nu}(k, k') = e^2\sum_\alpha\left\{\frac{J^\mu_{\alpha_0\alpha}(\boldsymbol{k})\,J^\nu_{\alpha\alpha_0}(\boldsymbol{k}')}{E_\alpha(1 - i0) - k^0 - E_{\alpha_0}} + \frac{J^\nu_{\alpha_0\alpha}(\boldsymbol{k}')\,J^\mu_{\alpha\alpha_0}(\boldsymbol{k})}{E_\alpha(1 - i0) + k^0 - E_{\alpha_0}}\right\}. \tag{7}$$

It contains the current matrix elements $J^\mu_{\alpha_0\alpha}$ between the initial (or final) state labelled α_0 and an arbitrary energy state α of the electron of atom a; these matrix elements are defined as:

$$J^\mu_{\alpha_0\alpha}(\boldsymbol{k}) = \langle\alpha_0\,|\gamma^0\gamma^\mu\,e^{-i\boldsymbol{k}\cdot\boldsymbol{r}_a}|\,\alpha\rangle. \tag{8}$$

From (7) the symmetry property

$$\Gamma_a^{\mu\nu}(k, k') = \Gamma_a^{\nu\mu}(k', k) \tag{9}$$

of the vertex function follows immediately.

To find the interaction energy due to two-photon exchange between a pair of atoms that are described in a nonrelativistic way the two-photon vertex function need to be studied only in the approximation obtained by performing a Foldy–Wouthuysen transformation for a Dirac particle in an external Coulomb field and retaining terms of lowest order in the inverse of the electron mass m. This transformation leads to a Schrödinger form for the hamiltonian, while the charge-current matrix elements become then:

$$\begin{aligned} J^0_{\alpha_0\alpha}(\boldsymbol{k}) &= \langle \alpha_0 | e^{-i\boldsymbol{k}\cdot\boldsymbol{r}_a} | \alpha \rangle, \\ J^i_{\alpha_0\alpha}(\boldsymbol{k}) &= \langle \alpha_0 | \tfrac{1}{2}\gamma^0 \{p_a^i/m + \gamma^i, e^{-i\boldsymbol{k}\cdot\boldsymbol{r}_a}\} | \alpha \rangle \qquad (i = 1, 2, 3), \end{aligned} \tag{10}$$

where the curly brackets denote an anticommutator and where $\boldsymbol{p}_a$ is the momentum operator of the electron of atom a. By adding an extra term -1 to the operator in the charge matrix element before its insertion into the two-photon vertex function the contribution of the charged nuclei to that vertex function can be taken into account easily. For nondegenerate positive-energy states α_0 the components of the atomic vertex functions, characterizing two-photon emission in nonrelativistic approximation, then get the form:

$$\begin{aligned} \Gamma_a^{00}(k, k') &= e^2 \sum_\alpha{}' Q_\alpha(\boldsymbol{k})\, Q_\alpha^*(-\boldsymbol{k}')\, \{(k_\alpha - k^0 - i0)^{-1} + (k_\alpha + k^0 - i0)^{-1}\}, \\ \Gamma_a^{0i}(k, k') &= \Gamma_a^{i0}(k', k) = (e^2/m) \sum_\alpha{}' Q_\alpha(\boldsymbol{k})\, P_\alpha^{*i}(-\boldsymbol{k}') \\ &\quad \times \{(k_\alpha - k^0 - i0)^{-1} - (k_\alpha + k^0 - i0)^{-1}\}, \\ \Gamma_a^{ij}(k, k') &= (e^2/m^2) \sum_\alpha{}' P_\alpha^i(\boldsymbol{k})\, P_\alpha^{*j}(-\boldsymbol{k}') \\ &\quad \times \{(k_\alpha - k^0 - i0)^{-1} + (k_\alpha + k^0 - i0)^{-1}\} - (e^2/m)\,\delta^{ij} D(\boldsymbol{k} + \boldsymbol{k}'), \end{aligned} \tag{11}$$

where the matrix elements

$$\begin{aligned} P_\alpha^i(\boldsymbol{k}) &= \langle \alpha_0 | \tfrac{1}{2}\{p_a^i, e^{-i\boldsymbol{k}\cdot\boldsymbol{r}_a}\} | \alpha \rangle, \\ Q_\alpha(\boldsymbol{k}) &= \langle \alpha_0 | e^{-i\boldsymbol{k}\cdot\boldsymbol{r}_a} - 1 | \alpha \rangle, \\ D(\boldsymbol{k} + \boldsymbol{k}') &= \langle \alpha_0 | e^{-i(\boldsymbol{k}+\boldsymbol{k}')\cdot\boldsymbol{r}_a} | \alpha_0 \rangle \end{aligned} \tag{12}$$

have been introduced. The primes at the summation signs in (11) limit the sums to intermediate states with positive energies: $E_\alpha = E_{\alpha_0} + k_\alpha$ with $k_\alpha \geqslant 0$. In deriving

(11) the reality of the nonrelativistic atomic hamiltonian in the coordinate representation has been used; it implies that projection operators on fixed energy E_α are real in that representation. For a nondegenerate state α_0 it follows then that the sum

$$\sum_{\alpha\,(\text{fixed } k_a)} Q_\alpha(k)\, Q_\alpha^*(-k') \tag{13}$$

is invariant under an interchange of k and k', so that the energy denominators could be combined in the way indicated in the first line of (11). Similar arguments were employed to bring the other components of $\Gamma_a^{\mu\nu}$ into a simple form. The "direct transition" matrix element $D(k + k')$ in Γ_a^{ij} arose from the "odd" part of the current operator, with $\gamma^0\gamma^i$, in (10).

The expressions (11) with (12) give the two-photon vertex function for a one-electron atom in the nonrelativistic approximation. The extension to many-electron atoms may be achieved by replacing the operators in the matrix elements (12) by sums of similar operators, one for each electron.

In the following the dispersive contribution to the two-photon exchange interaction energy for atoms in ground states will be evaluated. To obtain that contribution only the dispersive part of the two-photon vertex function (again denoted by $\Gamma_a^{\mu\nu}$) has to be considered; it is found from (11) by suppressing contributions with ground-level intermediate states in the sums. For atoms in nondegenerate ground states, to which the treatment will be confined from now on, it may be shown that the two-photon exchange interaction energy is completely dispersive.

The Fourier transform of the interatomic dispersion energy is determined now by inserting the dispersive vertex functions following from (11) into (6). Using the rotational invariance of the atomic hamiltonians one may show that for atoms in nondegenerate ground states the product $\Gamma_{a,\mu\nu}(k, k')\,\Gamma_b^{\mu\nu}(-k, -k')$ will in fact depend on k^2, k'^2, $k \cdot k'$ and k^{02}, for $k^0 = -k'^0$. As a consequence $F(Q)$ will depend only on Q^2 and thus, according to (5), the interaction energy only on the internuclear distance $R = |\boldsymbol{R}|$, as could be expected.

After substitution of the vertex functions (11) into (6) the analytical properties of $F(Q^2)$ as a function of Q^2 may be investigated. This variable is in fact, apart from a minus sign, the total momentum transfer $t = -(k + k')^2$ from one atom to the other, due to the exchange of the two photons. In appendix A it is shown that $F(t)$ is regular in the complex t plane except for a cut along the positive t axis, starting in a branchpoint at $t = 0$. Moreover, it is found there that the discontinuity across the cut may be given by the expression:

$$F(t + \mathrm{i}0) - F(t - \mathrm{i}0)$$

$$= -\frac{\mathrm{i}}{8\pi} \int_0^{\frac{1}{2}\sqrt{t}} \varkappa_\perp \, \mathrm{d}\varkappa_\perp \, \frac{\bar{\Gamma}_{a,\mu\nu}^{+}(k, k')\, \bar{\Gamma}_b^{-\mu\nu}(-k, -k')}{[t(\frac{1}{4}t - \varkappa_\perp^2)]^{\frac{1}{2}}} \qquad (t > 0), \tag{14}$$

where $\bar{\Gamma}_a^{\pm\mu\nu}(\boldsymbol{k}, \boldsymbol{k}')$ is defined to be equal to $\Gamma_a^{\mu\nu}(k, k')$ for $k^0 = -k'^0$, $k^{02} = k^2 = k'^2$, $k^0 \gtrless 0$. By writing $\boldsymbol{k} = \frac{1}{2}\boldsymbol{Q} + \boldsymbol{\kappa}_\perp$, $\boldsymbol{k}' = \frac{1}{2}\boldsymbol{Q} - \boldsymbol{\kappa}_\perp$, the product of vertex functions may be converted to a function of $\kappa_\perp^2$ and $Q^2 = -t$; this function has to be continued to $t > 0$ before being substituted into the integral.

In view of the analytical structure of $F(t)$, described above, it will be assumed to satisfy an unsubtracted dispersion relation:

$$F(t) = \int_0^\infty \mathrm{d}t' \, \frac{\varrho(t')}{t' - t}, \tag{15}$$

for t not lying on the positive t axis. Inserting this relation into the expression (5), with $t = -Q^2$, one finds for the interatomic dispersion energy the integral representation[2]):

$$V(R) = \frac{1}{4\pi R} \int_0^\infty \mathrm{d}t \, \mathrm{e}^{-\sqrt{t}R} \varrho(t). \tag{16}$$

It is completely determined by the weight function $\varrho(t)$ the form of which follows from (15):

$$\varrho(t) = (2\pi\mathrm{i})^{-1} \{F(t + \mathrm{i}0) - F(t - \mathrm{i}0)\}. \tag{17}$$

Thus, in view of (14), one may limit oneself to a discussion of the vertex functions $\bar{\Gamma}_a^{+\mu\nu}$ and $\bar{\Gamma}_b^{-\mu\nu}$ on the photon mass shells. This study, which entails the introduction of electromagnetic form factors to describe the atoms, will be the subject of the next section.

3. *Electromagnetic form factors.* The general vertex function $\Gamma_a^{\mu\nu}$, and hence the mass-shell vertex function $\bar{\Gamma}_a^{\mu\nu}$, satisfies the orthogonality relations

$$\Gamma_a^{\mu\nu}(k, k') \, k_\mu = 0, \qquad \Gamma_a^{\mu\nu}(k, k') \, k'_\nu = 0. \tag{18}$$

The first equation may be proved by inserting the expressions (11) and using the relation

$$\boldsymbol{k} \cdot \boldsymbol{P}_\alpha(\boldsymbol{k}) = -m \langle \alpha_0 | [H(a), \mathrm{e}^{-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}_a} - 1] | \alpha \rangle = m k_\alpha Q_\alpha(\boldsymbol{k}) \tag{19}$$

[with $H(a)$ the atomic hamiltonian of atom a] between the matrix elements in (12). Due to the symmetry property (9) the second equation in (18) follows directly from the first.

As a consequence of the properties (18) of the vertex function the product of mass-shell vertex functions $\bar{\Gamma}_{a,\mu\nu}^{+}(\boldsymbol{k}, \boldsymbol{k}') \, \bar{\Gamma}_b^{-\mu\nu}(-\boldsymbol{k}, -\boldsymbol{k}')$ may be found by calculat-

ing first, for arbitrary polarization vectors $\boldsymbol{\varepsilon}$ and $\boldsymbol{\varepsilon}'$ satisfying

$$\boldsymbol{\varepsilon} \cdot \boldsymbol{k} = 0, \qquad \boldsymbol{\varepsilon}' \cdot \boldsymbol{k}' = 0, \tag{20}$$

the expressions $\boldsymbol{\varepsilon} \cdot \bar{\boldsymbol{\Gamma}}_a^+ (\boldsymbol{k}, \boldsymbol{k}') \cdot \boldsymbol{\varepsilon}'$ and $\boldsymbol{\varepsilon} \cdot \bar{\boldsymbol{\Gamma}}_b^- (-\boldsymbol{k}, -\boldsymbol{k}') \cdot \boldsymbol{\varepsilon}'$. In fact, one may write, for four-vectors $\varepsilon_\pm^\mu(\boldsymbol{k}) = (0, \boldsymbol{\varepsilon}_\pm(\boldsymbol{k}))$ with space components satisfying $\boldsymbol{\varepsilon}_\pm(\boldsymbol{k}) \cdot \boldsymbol{k} = 0$ and $\boldsymbol{\varepsilon}_+(\boldsymbol{k}) \cdot \boldsymbol{\varepsilon}_-(\boldsymbol{k}) = 0$, the relation:

$$\sum_{\lambda = \pm} \varepsilon_\lambda^\mu(\boldsymbol{k})\, \varepsilon_\lambda^\nu(\boldsymbol{k}) = -g^{\mu\nu} - \frac{k^\mu k^\nu}{(k \cdot \eta)^2} + \frac{k^\mu \eta^\nu + k^\nu \eta^\mu}{k \cdot \eta}, \tag{21}$$

with k^μ on the mass shell ($k^2 = 0$) and $\eta^\mu = (1, \mathbf{0})$. Thus the contracted product of mass-shell vertex functions may be rewritten, in view of (18), in the following way:

$$\bar{\Gamma}_{a,\mu\nu}^+ (\boldsymbol{k}, \boldsymbol{k}')\, \bar{\Gamma}_b^{-\mu\nu} (-\boldsymbol{k}, -\boldsymbol{k}')$$

$$= \sum_{\lambda, \lambda' = \pm} \boldsymbol{\varepsilon}_\lambda(\boldsymbol{k}) \cdot \bar{\boldsymbol{\Gamma}}_a^+ (\boldsymbol{k}, \boldsymbol{k}') \cdot \boldsymbol{\varepsilon}_{\lambda'}(\boldsymbol{k}')\, \boldsymbol{\varepsilon}_\lambda(\boldsymbol{k}) \cdot \bar{\boldsymbol{\Gamma}}_b^- (-\boldsymbol{k}, -\boldsymbol{k}') \cdot \boldsymbol{\varepsilon}_{\lambda'}(\boldsymbol{k}'). \tag{22}$$

The contracted vertex functions occurring here follow from the last line of (11) by replacing k^{02} by $k^2 = k'^2$:

$$\boldsymbol{\varepsilon} \cdot \bar{\Gamma}_a^+ (\boldsymbol{k}, \boldsymbol{k}') \cdot \boldsymbol{\varepsilon}' = (e^2/m^2) \sum_{\alpha(\neq \alpha_0)} \{2k_\alpha/(k_\alpha^2 - k^2 - \mathrm{i}0)\}$$

$$\times\, \boldsymbol{\varepsilon} \cdot \boldsymbol{P}_\alpha(\boldsymbol{k})\, \boldsymbol{\varepsilon}' \cdot \boldsymbol{P}_\alpha^*(-\boldsymbol{k}') - (e^2/m)\, \boldsymbol{\varepsilon} \cdot \boldsymbol{\varepsilon}' D(\boldsymbol{k} + \boldsymbol{k}'), \tag{23}$$

while $\boldsymbol{\varepsilon} \cdot \bar{\boldsymbol{\Gamma}}_a^- (\boldsymbol{k}, \boldsymbol{k}') \cdot \boldsymbol{\varepsilon}'$ has the same form.

To employ the rotational invariance of the atomic hamiltonians the spherical tensor formalism will be used in the following. Thus the atomic states α will be labelled from now on by angular-momentum quantum numbers L_a, M_a and a third quantum number N_a, while the non-degenerate ground state is characterized by $L_a = 0$, $M_a = 0$, and $N_a = 0$; as a consequence the energy difference $k_\alpha = E_\alpha - E_{\alpha_0}$ will depend only on N_a. With the help of the expansion of a plane wave in spherical harmonics and spherical Bessel functions one may write then for the spherical component $\varkappa$ of the matrix element $\boldsymbol{P}_\alpha(\boldsymbol{k}) = \boldsymbol{P}_{N,L,M}(\boldsymbol{k})$ (omitting the index a referring to atom a in the remaining part of this section):

$$P_{N,L,M}^\varkappa(\boldsymbol{k}) = 4\pi \sum_{L_1, M_1, s_1} \frac{(-)^{M_1} (-\mathrm{i}k)^{L_1 + 2s_1}}{(2s_1)!!\, (2L_1 + 2s_1 + 1)!!}$$

$$\times \langle 0 \,|\tfrac{1}{2} \{p^\varkappa, r^{L_1 + 2s_1} Y_{L_1}^{M_1}(\hat{\boldsymbol{r}})\}|\, N, L, M \rangle\, Y_{L_1}^{-M_1}(\hat{\boldsymbol{k}}), \tag{24}$$

with $\hat{\boldsymbol{r}}$ and $\hat{\boldsymbol{k}}$ unit vectors in the direction of $\boldsymbol{r}$ and $\boldsymbol{k}$, respectively. With the use of the Wigner–Eckart theorem[5]) and the introduction of reduced matrix elements

$\Omega_{N,L}(L_1, s_1)$ by writing [v. (II.23)]*

$$\frac{1}{(2s_1)!!\,(2L_1 + 2s_1 + 1)!!}\left(\frac{4\pi}{2L_1 + 1}\right)^{\frac{1}{2}} \langle 0\,|\tfrac{1}{2}\{p^{\varkappa}, r^{L_1+2s_1} Y_{L_1}^{M_1}(\hat{r})\}|\,N, L, M\rangle$$

$$= \begin{pmatrix} L_1 & 1 & L \\ M_1 & \varkappa & M \end{pmatrix} \Omega_{N,L}(L_1, s_1) \tag{25}$$

(the first parameter between the brackets of the Ω-matrix elements of paper II could be dropped for brevity in the present case of nondegenerate ground-state atoms), an expression for the tensor $\mathsf{T}_N(\boldsymbol{k}, \boldsymbol{k}')$, contained in the first term of (23) and defined as

$$\mathsf{T}_N(\boldsymbol{k}, \boldsymbol{k}') = \sum_{LM} \boldsymbol{P}_{N,L,M}(\boldsymbol{k})\, \boldsymbol{P}^*_{N,L,M}(-\boldsymbol{k}') \tag{26}$$

may be found. In fact, from (24) and (25) the spherical components of this tensor follow as:

$$T_N^{\varkappa\lambda}(\boldsymbol{k}, \boldsymbol{k}') = \sum_{L_1, L_2, L, s_1, s_2} 4\pi(-)^{L+L_2+1}\,(-k^2)^{\frac{1}{2}(L_1+L_2)+s_1+s_2}$$
$$\times \{(2L_1 + 1)(2L_2 + 1)\}^{\frac{1}{2}}\, \Omega_{N,L}(L_1, s_1)$$
$$\times \Omega^*_{N,L}(L_2, s_2)\, \hat{T}^{\varkappa\lambda}_{L_1,L_2,L}(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}'), \tag{27}$$

where the ancillary tensor

$$\hat{T}^{\varkappa\lambda}_{L_1,L_2,L}(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}') = \sum_{M_1, M_2, M} (-)^{M_1+M_2+M}$$
$$\times \begin{pmatrix} L_1 & 1 & L \\ M_1 & \varkappa & M \end{pmatrix} \begin{pmatrix} L_2 & 1 & L \\ M_2 & \lambda & -M \end{pmatrix} Y_{L_1}^{-M_1}(\hat{\boldsymbol{k}})\, Y_{L_2}^{-M_2}(\hat{\boldsymbol{k}}') \tag{28}$$

has been introduced. The space inversion invariance of the atomic hamiltonian may be invoked to show that in (27) only products of matrix elements with $L_1 + L_2$ even occur, so that the tensor $\hat{T}^{\varkappa\lambda}$ need to be considered only for parameters L_1 and L_2 satisfying that constraint.

The contracted tensor

$$\boldsymbol{\varepsilon} \cdot \hat{\mathsf{T}}_{L_1,L_2,L}(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}') \cdot \boldsymbol{\varepsilon}', \tag{29}$$

with polarization vectors satisfying (20), is, for $L_1 + L_2$ even, invariant under rotations and spatial inversion of the coordinate system; as a consequence it may be

* Formulae of the four preceding papers[1]) of this series are indicated by roman numbers I–IV.

written in the form:

$$\boldsymbol{\varepsilon} \cdot \boldsymbol{\varepsilon}' t_{\mathrm{E}}(\cos\theta) + (\boldsymbol{\varepsilon} \cdot \hat{\boldsymbol{k}}' \boldsymbol{\varepsilon}' \cdot \hat{\boldsymbol{k}} - \boldsymbol{\varepsilon} \cdot \boldsymbol{\varepsilon}' \hat{\boldsymbol{k}} \cdot \hat{\boldsymbol{k}}')\, t_{\mathrm{M}}(\cos\theta) \tag{30}$$

with as yet undetermined functions t_{E} and t_{M} depending on $\hat{\boldsymbol{k}} \cdot \hat{\boldsymbol{k}}' = \cos\theta$.

To evaluate t_{E} and t_{M} it is convenient to choose a coordinate frame in which $\hat{\boldsymbol{k}}$ and $\hat{\boldsymbol{k}}'$ have the forms (0, 0, 1) and (sin θ, 0, cos θ), respectively, so that their spherical components are $\hat{k}^0 = 1$, $\hat{k}^{\pm 1} = 0$, $\hat{k}'^0 = \cos\theta$, $\hat{k}'^{\pm 1} = \mp 2^{-\frac{1}{2}} \sin\theta$. In view of (20) the spherical polarization vector $\varepsilon^{\varkappa}$ then has a vanishing zeroth component, $\varepsilon^0 = 0$, while ε'^{λ} may be written as:

$$\varepsilon'^{\lambda} = \sum_{\mu} \varepsilon''^{\mu}\, d^{(1)}_{\mu\lambda}(\theta). \tag{31}$$

Here ε''^{μ} is a spherical vector with $\varepsilon''^0 = 0$ and $d^{(1)}_{\mu\lambda}(\theta)$ is an element of the irreducible representation matrix[5]) (with angular momentum quantum number $j = 1$) corresponding to a rotation with Euler angles $(\alpha, \beta, \gamma) = (0, \theta, 0)$. Indeed, by inserting the explicit expressions for $d^{(1)}_{\mu\lambda}$ one may check that ε'^{λ} (31) satisfies (20) for arbitrary ε''^{μ} (with $\varepsilon''^0 = 0$). Definite helicity states for photons travelling in the directions $\hat{\boldsymbol{k}}$ and $\hat{\boldsymbol{k}}'$, with helicities $\varkappa_0$ and μ_0 ($\varkappa_0, \mu_0 = \pm 1$), are characterized by polarization vectors $\varepsilon^{\varkappa}$ and ε'^{λ} obtained by putting $\varepsilon^{\varkappa} = (-)^{\varkappa} \delta_{\varkappa, -\varkappa_0}$ and, in (31), $\varepsilon''^{\mu} = (-)^{\mu} \delta_{\mu, -\mu_0}$, *v.* ref. 6. Substitution of (28) and (31) into (29) gives:

$$\boldsymbol{\varepsilon} \cdot \hat{\mathbf{T}}_{L_1, L_2, L}(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}') \cdot \boldsymbol{\varepsilon}' = \sum_{\varkappa, \lambda, M_1, M_2} (-)^{\varkappa} \{(2L_1 + 1)/4\pi\}^{\frac{1}{2}}$$

$$\times \begin{pmatrix} L_1 & 1 & L \\ 0 & \varkappa & -\varkappa \end{pmatrix} \begin{pmatrix} L_2 & 1 & L \\ M_2 & \lambda & \varkappa \end{pmatrix} \varepsilon^{-\varkappa} \varepsilon''^{-\mu}\, d^{(1)}_{-\mu, -\lambda}(\theta)\, Y^{-M_2}_{L_2}(\theta, 0), \tag{32}$$

since $Y^{M_1}_{L_1}(0, 0)$ has the form $\delta_{M_1, 0} \{(2L_1 + 1)/4\pi\}^{\frac{1}{2}}$. The Clebsch–Gordan series[5]) for the rotation matrices

$$d^{(L_1)}_{M_1', M_1}(\theta)\, d^{(L_2)}_{M_2', M_2}(\theta)$$

$$= \sum_{L} (2L + 1) \begin{pmatrix} L_1 & L_2 & L \\ M_1' & M_2' & M' \end{pmatrix} \begin{pmatrix} L_1 & L_2 & L \\ M_1 & M_2 & M \end{pmatrix} d^{(L)}_{M', M}(\theta), \tag{33}$$

with $Y^{M_2}_{L_2}(\theta, 0) = d^{(L_2)}_{0, M_2}(\theta) \{(2L_2 + 1)/4\pi\}^{\frac{1}{2}}$, may be used then to rewrite (32) in the form

$$\boldsymbol{\varepsilon} \cdot \hat{\mathbf{T}}_{L_1, L_2, L}(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}') \cdot \boldsymbol{\varepsilon}' = \sum_{\varkappa, \mu} (-)^{\varkappa} (4\pi)^{-1} \{(2L_1 + 1)(2L_2 + 1)\}^{\frac{1}{2}}$$

$$\times \begin{pmatrix} L_1 & 1 & L \\ 0 & \varkappa & -\varkappa \end{pmatrix} \begin{pmatrix} L_2 & 1 & L \\ 0 & \mu & -\mu \end{pmatrix} \varepsilon^{-\varkappa} \varepsilon''^{-\mu}\, d^{(L)}_{\mu, -\varkappa}(\theta). \tag{34}$$

On the other hand, the expression (30) becomes in the chosen coordinate frame, upon substitution of (31) and explicit expressions for $d^{(1)}_{\lambda\mu}(\theta)$:

$$\sum_{\varkappa,\mu} (-)^{\varkappa} \varepsilon^{-\varkappa}\varepsilon''^{-\mu} \tfrac{1}{2}(1 - \varkappa\mu\cos\theta)\{t_{\mathrm{E}}(\cos\theta) + \varkappa\mu t_{\mathrm{M}}(\cos\theta)\}. \tag{35}$$

When the explicit form for $d^{(L)}_{\mu,-\varkappa}(\theta)$ with $\varkappa, \mu = \pm 1$, *viz.*

$$d^{(L)}_{\mu,-\varkappa}(\theta) = \{L(L+1)\}^{-1}\tfrac{1}{2}(1 - \varkappa\mu\cos\theta)\{a^L_{\mathrm{E}}(\cos\theta) + \varkappa\mu a^L_{\mathrm{M}}(\cos\theta)\}, \tag{36}$$

with

$$\begin{aligned} a^L_{\mathrm{E}}(\cos\theta) &= P''_{L+1}(\cos\theta) + P''_{L-1}(\cos\theta) - P'_L(\cos\theta), \\ a^L_{\mathrm{M}}(\cos\theta) &= 2P''_L(\cos\theta), \end{aligned} \tag{37}$$

is inserted into (34) and the coefficients of $\varepsilon^{-\varkappa}\varepsilon''^{-\mu}$ in (34) and (35) are equated, one obtains a relation for t_{E} and t_{M}:

$$\begin{aligned} t_{\mathrm{E}}(\cos\theta) + \varkappa\mu t_{\mathrm{M}}(\cos\theta) &= \frac{[(2L_1+1)(2L_2+1)]^{\frac{1}{2}}}{4\pi L(L+1)} \\ &\times \begin{pmatrix} L_1 & 1 & L \\ 0 & \varkappa & -\varkappa \end{pmatrix}\begin{pmatrix} L_2 & 1 & L \\ 0 & \mu & -\mu \end{pmatrix}\{a^L_{\mathrm{E}}(\cos\theta) + \varkappa\mu a^L_{\mathrm{M}}(\cos\theta)\}, \end{aligned} \tag{38}$$

for $\varkappa, \mu = \pm 1$. Since $L_1 + L_2$ is even the expressions for $t_{\mathrm{E}}(\cos\theta)$ and $t_{\mathrm{M}}(\cos\theta)$ are thus found to be (for $i = E, M$):

$$\begin{aligned} t_i(\cos\theta) &= \frac{[(2L_1+1)(2L_2+1)]^{\frac{1}{2}}}{4\pi L(L+1)}\begin{pmatrix} L_1 & 1 & L \\ 0 & 1 & -1 \end{pmatrix}\begin{pmatrix} L_2 & 1 & L \\ 0 & 1 & -1 \end{pmatrix} \\ &\times \{(1 - \delta_{L,L_1}\delta_{L,L_2})\, a^L_i(\cos\theta) + \delta_{L,L_1}\delta_{L,L_2} b^L_i(\cos\theta)\}, \end{aligned} \tag{39}$$

with the abbreviations

$$b^L_{\mathrm{E}}(\cos\theta) = a^L_{\mathrm{M}}(\cos\theta), \qquad b^L_{\mathrm{M}}(\cos\theta) = a^L_{\mathrm{E}}(\cos\theta). \tag{40}$$

When (39) is substituted into the expression (30) for the contracted tensor (29) and the result into (23) with (12), (26), and (27), one arrives at the following result for the vertex function contracted with polarization vectors:

$$\begin{aligned} &\boldsymbol{\varepsilon}\cdot\overline{\boldsymbol{\Gamma}}^{+}(\boldsymbol{k},\boldsymbol{k}')\cdot\boldsymbol{\varepsilon}' = \boldsymbol{\varepsilon}\cdot\overline{\boldsymbol{\Gamma}}^{-}(\boldsymbol{k},\boldsymbol{k}')\cdot\boldsymbol{\varepsilon}' \\ &\quad = k^2\boldsymbol{\varepsilon}\cdot\boldsymbol{\varepsilon}' F_{\mathrm{E}}(k,\cos\theta) + (\boldsymbol{\varepsilon}\cdot\boldsymbol{k}'\boldsymbol{\varepsilon}'\cdot\boldsymbol{k} - \boldsymbol{\varepsilon}\cdot\boldsymbol{\varepsilon}'\boldsymbol{k}\cdot\boldsymbol{k}')\, F_{\mathrm{M}}(k,\cos\theta) \end{aligned} \tag{41}$$

with the electromagnetic form factors F_i ($i = E, M$):

$$F_i(k, \cos\theta) = \frac{2e^2}{m^2k^2} \sum_{N(\neq 0)} \sum_{L, L_1, L_2, s_1, s_2} \frac{k_N}{k_N^2 - k^2 - \mathrm{i}0}$$
$$\times (-k^2)^{\frac{1}{2}(L_1+L_2)+s_1+s_2} \frac{(2L_1+1)(2L_2+1)}{L(L+1)}$$
$$\times \begin{pmatrix} L_1 & 1 & L \\ 0 & 1 & -1 \end{pmatrix} \begin{pmatrix} L_2 & 1 & L \\ 0 & 1 & -1 \end{pmatrix}$$
$$\times \{(1 - \delta_{L,L_1}\delta_{L,L_2})\, a_i^L(\cos\theta) - \delta_{L,L_1}\delta_{L,L_2} b_i^L(\cos\theta)\}$$
$$\times \Omega_{N,L}(L_1, s_1)\, \Omega^*_{N,L}(L_2, s_2) - \delta_{i,\mathrm{E}} \frac{e^2}{mk^2} \langle 0 | \mathrm{e}^{-\mathrm{i}(k+k')\cdot r} | 0 \rangle. \tag{42}$$

The symmetry between the two form factors can be pushed further by evaluating the last term containing the "direct transition" matrix element. In fact, by employing the plane-wave expansion, for both $\exp(-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r})$ and $\exp(-\mathrm{i}\boldsymbol{k}'\cdot\boldsymbol{r})$, and coupling the resulting spherical harmonics depending on $\hat{\boldsymbol{r}}$ one finds, using moreover the rotation invariance of the ground state,

$$\langle 0 | \mathrm{e}^{-\mathrm{i}(k+k')\cdot r} | 0 \rangle = \sum_{L, s_1, s_2} (-k^2)^{L+s_1+s_2} (2L+1)\, M(L, s_1, s_2)\, P_L(\cos\theta), \tag{43}$$

with the matrix element:

$$M(L, s_1, s_2) = \{(2s_1)!!\,(2s_2)!!\,(2L+2s_1+1)!!\,(2L+2s_2+1)!!\}^{-1}$$
$$\times \langle 0 | r^{2L+2s_1+2s_2} | 0 \rangle. \tag{44}$$

With the help of the sum rules (B5) and (B6) for the atomic matrix elements the form factors may be brought then into the form:

$$F_i(k, \cos\theta) = \sum_{n=0}^{\infty} (-k^2)^n \left\{ \sum_{N(\neq 0)} \frac{f_i^{(N,n)}(\cos\theta)}{k_N^2 - k^2 - \mathrm{i}0} + g_i^{(n+1)}(\cos\theta) \right\}. \tag{45}$$

Here the angular functions containing the atomic matrix elements are defined as:

$$f_i^{(N,n)}(\cos\theta) = \frac{2e^2}{m^2} \sum_{L, L_1, L_2, s_1, s_2}^{(n)} \frac{(2L_1+1)(2L_2+1)}{L(L+1)}$$
$$\times \begin{pmatrix} L_1 & 1 & L \\ 0 & 1 & -1 \end{pmatrix} \begin{pmatrix} L_2 & 1 & L \\ 0 & 1 & -1 \end{pmatrix}$$
$$\times \{(1 - \delta_{L,L_1}\delta_{L,L_2})\, a_i^L(\cos\theta) - \delta_{L,L_1}\delta_{L,L_2} b_i^L(\cos\theta)\}$$
$$\times k_N^{-1} \Omega_{N,L}(L_1, s_1)\, \Omega^*_{N,L}(L_2, s_2), \tag{46}$$

$$g_i^{(n)}(\cos\theta) = \frac{2e^2}{m^2} \sum_{L,L_1,L_2,s_1,s_2}^{(n)} \frac{(2L_1+1)(2L_2+1)}{L(L+1)}$$

$$\times \begin{pmatrix} L_1 & 1 & L \\ 0 & 1 & -1 \end{pmatrix} \begin{pmatrix} L_2 & 1 & L \\ 0 & 1 & -1 \end{pmatrix} \Bigg[-\delta_{L+1,L_1}\delta_{L-1,L_2} \frac{(2L+1)^2}{L(L+1)}$$

$$\times a_i^L(\cos\theta) \sum_{N(\neq 0)} k_N^{-1} \Omega_{N,L}(L_1, s_1)\, \Omega^*_{N,L}(L_2, s_2)$$

$$+ \delta_{L,L_1}\delta_{L,L_2} b_i^L(\cos\theta)$$

$$\times \Big\{ \sum_{N(\neq 0)} k_N^{-1} \Omega_{N,L}(L_1, s_1)\, \Omega^*_{N,L}(L_2, s_2) - \tfrac{1}{2} m M(L, s_1, s_2) \Big\} \Bigg], \quad (47)$$

where the symbols (n) at the summation signs stand for the subsidiary condition $\frac{1}{2}(L_1 + L_2) + s_1 + s_2 = n$. The advantage of the representation (45)–(47) for the form factors as compared to that of (42) lies, apart from the greater symmetry between the two form factors, in the fact that in (45) only positive powers of k^2 occur, while (42) contains apparently terms with k^{-2}. In fact, for $i = \mathrm{E}$ the parameter values $L_1 = L_2 = s_1 = s_2 = 0$ yield a contribution with k^{-2} to the first term of (42); by employing (43) and the sum rule (B6) it may be shown, however, that this term cancels indeed the k^{-2} contribution of the last term in (42).

The expressions (45)–(47) for the form factors are the main results of this section. They describe the electromagnetic structure of a neutral atom by means of the matrix elements Ω and M defined by (25) and (44). According to (41) the form factors determine the two-photon vertex function contracted with a pair of polarization vectors. Hence knowledge of the form factors is enough to be able to calculate the cross section for scattering of photons by neutral atoms. Usually the electric dipole approximation is made in an early stage of the evaluation of that cross section. The present results may be used to generalize these calculations so as to include the contribution of higher multipole transitions.

The nomenclature of the form factors becomes evident when studying them for vanishing photon momenta: $k = 0$. The expression (45) reduces in that case to:

$$F_i(0) = \sum_{N(\neq 0)} k_N^{-2} f_i^{(N,0)} + g_i^{(1)}, \quad (48)$$

where we omitted the argument $\cos\theta$ on both sides since the form factors are now in fact independent of it, as follows from (46) and (47) with (37) and (40). By using the sum rule

$$\sum_{N(\neq 0)} k_N^{-1} \Omega_{N,1}(0,0)\, \Omega^*_{N,1}(2,0) = 0, \quad (49)$$

following from (B5) for $L = 2$, $s_1 = -1$, $s_2 = 0$, one finds that in $F_E(0)$ only the term with $f_E^{(N,0)}$ contributes and in $F_M(0)$ only that with $g_M^{(1)}$; the result is:

$$\begin{aligned} F_E(0) &= \tfrac{2}{3}e^2 \sum_{N(\neq 0)} k_N^{-1} |r_N|^2, \\ F_M(0) &= \tfrac{2}{3}e^2 \sum_{N(\neq 0)} k_N^{-1} |(r \wedge p)_N/2m|^2 - (e^2/6m) \langle 0 | r^2 | 0 \rangle. \end{aligned} \tag{50}$$

Here the reduced matrix elements $r_N = \langle 0 \| r \| N, 1 \rangle$ and $(r \wedge p)_N = \langle 0 \| r \wedge p \| N, 1 \rangle$ have been introduced; they characterize the electric and magnetic dipole moments of the atom and are proportional to particular Ω-matrix elements [*v.* (III.5) and (III.9)]:

$$\begin{aligned} \Omega_{N,1}(0, 0) &= -imk_N r_N, \\ \Omega_{N,1}(1, 0) &= -i(\sqrt{2}/6)(r \wedge p)_N. \end{aligned} \tag{51}$$

The expressions (50) show that the electric and magnetic form factors contain in an analogous way the electric and magnetic dipole moment matrix elements; the magnetic form factor contains moreover a matrix element that describes the diamagnetic properties of the atom. Due to the rotation invariance of the atomic hamiltonian the reduced matrix elements $(r \wedge p)_N$ vanish in the present case for $N \neq 0$, so that in fact only the diamagnetic term contributes to $F_M(0)$.

The form factors (50) may be interpreted as the static polarizabilities of the atom in its ground state. In fact, starting from the interaction hamiltonian

$$H_{\text{int}} = er \cdot E + (e/2m)(r \wedge p) \cdot B + (e^2/8m)(r \wedge B)^2 \tag{52}$$

of a neutral one-electron atom in homogeneous static electric and magnetic fields E and B [*v.* (IV.31)] and applying perturbation theory in first and second order one finds for the energy shift ΔE of the (rotation-invariant) ground state:

$$\Delta E = -\tfrac{1}{2}F_E(0) E^2 - \tfrac{1}{2}F_M(0) B^2, \tag{53}$$

up to second order in the electromagnetic fields.

In the following it will be convenient to write the form factors as functions of the absolute values of the photon momenta and the momentum transfer $t = -(k+k')^2 = -2k^2(1 + \cos\theta)$. To that end the Legendre polynomials $P_L(\cos\theta)$ occurring in (37) are written as:

$$P_L(\cos\theta) = \sum_m [P_L(\cos\theta)]_m \{\tfrac{1}{2}(1 + \cos\theta)\}^m, \tag{54}$$

where the coefficient $[P_L]_m$ follows by making use of the representation of the Legendre polynomial as a hypergeometric function[7]):

$$P_L(\cos\theta) = (-)^L\, {}_2F_1\{-L, L+1; 1; \tfrac{1}{2}(1+\cos\theta)\}. \tag{55}$$

Series expansion of the right-hand side leads to

$$[P_L(\cos\theta)]_m = \frac{(-)^{L+m}(L+m)!}{(L-m)!\,(m!)^2}, \tag{56}$$

while similar expressions, *viz.*

$$[P_L^{(j)}(\cos\theta)]_m = \frac{(-)^{L+m+j}(L+m+j)!}{2^j(L-m-j)!\,(m+j)!\,m!}, \tag{57}$$

are found for the coefficients of the derivatives $P_L^{(j)}(\cos\theta) = \mathrm{d}^j P_L(\cos\theta)/\mathrm{d}(\cos\theta)^j$ of the Legendre polynomials. Evidently these coefficients are different from zero only when $0 \leqslant m \leqslant L - j$.

Introducing these expansions into (45) one finds for the form factors:

$$F_i(k,t) = \sum_{n=0}^{\infty} \sum_m (-k^2)^{n-m} \left(\frac{t}{4}\right)^m \left\{ \sum_{N(\neq 0)} \frac{f_i^{(N,n,m)}}{k_N^2 - k^2 - \mathrm{i}0} + g_i^{(n+1,m)} \right\}, \tag{58}$$

with $i = \mathrm{E}, \mathrm{M}$. The coefficients, defined as

$$f_i^{(N,n,m)} = [f_i^{(N,n)}(\cos\theta)]_m, \qquad g_i^{(n+1,m)} = [g_i^{(n+1)}(\cos\theta)]_m, \tag{59}$$

may be evaluated by using the explicit expressions (46) and (47) with (37) and (40). From these it follows that the coefficients are nonvanishing only when $0 \leqslant m \leqslant n$ so that indeed no negative power of k^2 occurs in the form factor (58).

In ref. 2 the dependence of the form factors on the momentum transfer is neglected in the course of the calculation of the interaction energy of neutral particles. Since explicit expressions have been obtained now for the atomic form factors the consequences of such an approximation may be discussed. In (58) the approximation amounts to the suppression of all terms with m different from zero, so that effectively, according to (59), the functions f_i and g_i are considered to be independent of $\cos\theta$. The expressions (46) and (47) with (37) and (40) then show that only the terms with $L = 1$ are treated correctly in both F_E and F_M. Since in particular the electric dipole terms in the form factors are of this type, this approximation may be considered as an extension of the well-known "dipole approximation". In the following we shall avoid all approximations of this type by retaining the full dependence of the form factors on the momentum transfer, so that the complete multipole expansion of the dispersion energy will be derived.

4. *Evaluation of the retarded dispersion energy.* In this section the interatomic dispersion energy will be derived from the general integral expression (16) by evaluating the weight function $\varrho(t)$ given in (17) with (14). First of all the contracted product of two-photon vertex functions occurring in (14) has to be calculated. From (22) and (41) one finds, using the identity [*v.* (21)]:

$$\sum_{\lambda=\pm} \varepsilon_\lambda(\boldsymbol{k})\, \varepsilon_\lambda(\boldsymbol{k}) = \mathsf{U} - \boldsymbol{k}\boldsymbol{k}/k^2, \tag{60}$$

the following expression for the product of vertex functions:

$$\bar{\Gamma}^{+}_{a,\mu\nu}(\boldsymbol{k}, \boldsymbol{k}')\, \bar{\Gamma}^{-\mu\nu}_{b}(-\boldsymbol{k}, -\boldsymbol{k}') = \{k^4 + (\boldsymbol{k}\cdot\boldsymbol{k}')^2\}\,(F_{a\mathrm{E}}F_{b\mathrm{E}} + F_{a\mathrm{M}}F_{b\mathrm{M}}) - 2k^2\boldsymbol{k}\cdot\boldsymbol{k}'\,(F_{a\mathrm{E}}F_{b\mathrm{M}} + F_{a\mathrm{M}}F_{b\mathrm{E}}) \tag{61}$$

with form factors depending on k and $\hat{\boldsymbol{k}}\cdot\hat{\boldsymbol{k}}' = \cos\theta$ or on k and t, as given in (58). Inserting now $\boldsymbol{k} = \frac{1}{2}\boldsymbol{Q} + \boldsymbol{\varkappa}_\perp$, $\boldsymbol{k}' = \frac{1}{2}\boldsymbol{Q} - \boldsymbol{\varkappa}_\perp$, so that $k^2 = k'^2 = -\frac{1}{4}t + \varkappa_\perp^2$ and $\boldsymbol{k}\cdot\boldsymbol{k}' = -\frac{1}{4}t - \varkappa_\perp^2$ with $t = -Q^2$, one finds for the right-hand side of (61) a function of t and $\varkappa_\perp^2$. After analytical continuation of that function to positive values of t (which is an altogether trivial manipulation in the present case, since the power series expansion in t is known) it has to be substituted into (14) .Then one gets, with (17), for the weight function:

$$\varrho(t) = -\frac{1}{8\pi^2}\int_0^{\frac{1}{2}\sqrt{t}} \mathrm{d}\varkappa_\perp \frac{\varkappa_\perp}{[t(\frac{1}{4}t - \varkappa_\perp^2)]^{\frac{1}{2}}}\{(\tfrac{1}{16}t^2 + \varkappa_\perp^4)\,(F_{a\mathrm{E}}F_{b\mathrm{E}} + F_{a\mathrm{M}}F_{b\mathrm{M}}) - (\tfrac{1}{16}t^2 - \varkappa_\perp^4)\,(F_{a\mathrm{E}}F_{b\mathrm{M}} + F_{a\mathrm{M}}F_{b\mathrm{E}})\}, \tag{62}$$

or, with the new integration variable x defined by $\{\frac{1}{4}t - \varkappa_\perp^2\}^{\frac{1}{2}} = \frac{1}{2}t^{\frac{1}{2}}x$,

$$\varrho(t) = -\frac{t^2}{(16\pi)^2}\sum_{i,j=\mathrm{E},\mathrm{M}}\int_0^1 \mathrm{d}x\, \varphi_{ij}(x)\, F_{a,i}(k, t)\, F_{b,j}(k, t), \tag{63}$$

where the abbreviations

$$\varphi_{\mathrm{EE}}(x) = \varphi_{\mathrm{MM}}(x) = 2 - 2x^2 + x^4,$$
$$\varphi_{\mathrm{EM}}(x) = \varphi_{\mathrm{ME}}(x) = -2x^2 + x^4 \tag{64}$$

have been employed. In (63) the form factors (58) have to be substituted, with the replacement of k^2 by $-\frac{1}{4}t + \varkappa_\perp^2$, or by $-\frac{1}{4}tx^2$; the result is

$$\varrho(t) = -\frac{1}{16\pi^2} \sum_{n_a, n_b, m_a, m_b} \sum_{i,j=\mathrm{E},\mathrm{M}} \left(\frac{t}{4}\right)^{n_a+n_b+2}$$

$$\times \int_0^1 \mathrm{d}x\, \varphi_{ij}(x)\, x^{2(n_a+n_b-m_a-m_b)} \left\{ \sum_{N_a(\neq 0)} \frac{f_{a,i}^{(N_a, n_a, m_a)}}{k_{N_a}^2 + \frac{1}{4}tx^2} + g_{a,i}^{(n_a+1, m_a)} \right\}$$

$$\times \left\{ \sum_{N_b(\neq 0)} \frac{f_{b,j}^{(N_b, n_b, m_b)}}{k_{N_b}^2 + \frac{1}{4}tx^2} + g_{b,j}^{(n_b+1, m_b)} \right\}. \tag{65}$$

From this weight function the interatomic dispersion energy is obtained by making use of (16). With the new integration variable $z = \frac{1}{2}t^{\frac{1}{2}}$ one arrives at the expression:

$$V(R) = -\frac{1}{8\pi^3 R} \sum_{n_a, n_b, m_a, m_b} \sum_{i,j=\mathrm{E},\mathrm{M}} \int_0^\infty \mathrm{d}z \int_0^1 \mathrm{d}x\, \mathrm{e}^{-2zR}$$

$$\times z^{2(n_a+n_b+2)+1} \varphi_{ij}(x)\, x^{2(n_a+n_b-m_a-m_b)}$$

$$\left\{ \sum_{N_a(\neq 0)} \frac{f_{a,i}^{(N_a, n_a, m_a)}}{k_{N_a}^2 + z^2x^2} + g_{a,i}^{(n_a+1, m_a)} \right\} \left\{ \sum_{N_b(\neq 0)} \frac{f_{b,j}^{(N_b, n_b, m_b)}}{k_{N_b}^2 + z^2x^2} + g_{b,j}^{(n_b+1, m_b)} \right\}. \tag{66}$$

The coefficients $f_i^{(N,n,m)}$ and $g_i^{(n,m)}$ follow from (59) with (37), (40), (46), (47) and (57), as expansions involving the atomic matrix elements $\Omega_{N,L}(L_1, s_1)$ and $M(L, s_1, s_2)$, defined in (25) and (44), respectively. To evaluate the integral occurring in (66) it is convenient to split $V(R)$ into three parts by working out the product of the curly bracket expressions. The first term becomes, upon using the definitions (59), (64) and shifting the summation variable $m = m_a + m_b$ in an appropriate way:

$$V_{\mathrm{I}}(R) = -\frac{1}{8\pi^3 R} \sum_{N_a(\neq 0), N_b(\neq 0)} \sum_{n_a, n_b=0}^{\infty} \sum_{m=0}^{n_a+n_b+2} \Big[\sum_{\lambda=\pm 1} (f_{a,\mathrm{E}}^{(N_a, n_a)} + \lambda f_{a,\mathrm{M}}^{(N_a, n_a)})$$

$$\times (f_{b,\mathrm{E}}^{(N_b, n_b)} + \lambda f_{b,\mathrm{M}}^{(N_b, n_b)}) \tfrac{1}{4}(1 - \lambda\cos\theta)^2 \Big]_m$$

$$\times \int_0^\infty \mathrm{d}z \int_0^1 \mathrm{d}x\, \mathrm{e}^{-2zR} \frac{z^{2(n_a+n_b+2)+1} x^{2(n_a+n_b-m+2)}}{(k_{N_a}^2 + z^2x^2)(k_{N_b}^2 + z^2x^2)}. \tag{67}$$

The double integral may be evaluated by using the method of partial fractions and employing the relation (for $p \geqslant 0$, $q \geqslant 0$)

$$\int_0^\infty \mathrm{d}z \int_0^1 \mathrm{d}x\, \mathrm{e}^{-2zR} \frac{z^{2(p+q)+1}x^{2p}}{k^2 + z^2x^2} = \frac{\mathrm{d}^{2q}}{\mathrm{d}(2R)^{2q}} \frac{1}{2kR} \frac{\mathrm{d}^{2p}}{\mathrm{d}(2R)^{2p}} P(2kR), \tag{68}$$

which may be proved by induction with respect to p. The function $P(x)$ occurring here is connected to the sine and cosine integrals[7]):

$$P(x) = \int_0^\infty \mathrm{d}z \frac{\mathrm{e}^{-zx}}{1 + z^2} = \mathrm{Ci}(x) \sin x - \left\{\mathrm{Si}(x) - \frac{\pi}{2}\right\} \cos x; \tag{69}$$

it satisfies the differential equation $\mathrm{d}^2P(x)/\mathrm{d}x^2 = -P(x) + x^{-1}$. By using (68) in (67) the following result is found for $V_{\mathrm{I}}(R)$:

$$\begin{aligned} V_{\mathrm{I}}(R) = {} & \frac{1}{16\pi^3} \sum_{N_a(\neq 0), N_b(\neq 0)} \sum_{n_a, n_b=0}^{\infty} \sum_{m=0}^{n_a+n_b+2} \frac{1}{k_{N_a}^2 - k_{N_b}^2} \\ & \times \left[\sum_{\lambda=\pm 1} (f_{a,\mathrm{E}}^{(N_a, n_a)} + \lambda f_{a,\mathrm{M}}^{(N_a, n_a)})(f_{b,\mathrm{E}}^{(N_b, n_b)} + \lambda f_{b,\mathrm{M}}^{(N_b, n_b)}) \tfrac{1}{4}(1 - \lambda\cos\theta)^2\right]_m \\ & \times \frac{1}{R} \frac{\mathrm{d}^{2m}}{\mathrm{d}(2R)^{2m}} \frac{1}{R} \frac{\mathrm{d}^{2(n_a+n_b-m+2)}}{\mathrm{d}(2R)^{2(n_a+n_b-m+2)}} \left\{\frac{P(2k_{N_a}R)}{k_{N_a}} - \frac{P(2k_{N_b}R)}{k_{N_b}}\right\}. \end{aligned} \tag{70}$$

Likewise the second and third contributions to the dispersion energy become:

$$\begin{aligned} V_{\mathrm{II}}(R) = {} & -\frac{1}{16\pi^3} \sum_{N_a(\neq 0)} \sum_{n_a, n_b=0}^{\infty} \sum_{m=0}^{n_a+n_b+2} \\ & \times \left[\sum_{\lambda=\pm 1} (f_{a,\mathrm{E}}^{(N_a, n_a)} + \lambda f_{a,\mathrm{M}}^{(N_a, n_a)})(g_{b,\mathrm{E}}^{(n_b+1)} + \lambda g_{b,\mathrm{M}}^{(n_b+1)}) \tfrac{1}{4}(1 - \lambda\cos\theta)^2\right]_m \\ & \times \frac{1}{R} \frac{\mathrm{d}^{2m}}{\mathrm{d}(2R)^{2m}} \frac{1}{R} \frac{\mathrm{d}^{2(n_a+n_b-m+2)}}{\mathrm{d}(2R)^{2(n_a+n_b-m+2)}} \frac{P(2k_{N_a}R)}{k_{N_a}} + (a \leftrightarrow b), \end{aligned} \tag{71}$$

$$\begin{aligned} V_{\mathrm{III}}(R) = {} & -\frac{1}{16\pi^3} \sum_{n_a, n_b=0}^{\infty} \sum_{m=0}^{n_a+n_b+2} \left[\sum_{\lambda=\pm 1} (g_{a,\mathrm{E}}^{(n_a+1)} + \lambda g_{a,\mathrm{M}}^{(n_a+1)})\right. \\ & \left. \times (g_{b,\mathrm{E}}^{(n_b+1)} + \lambda g_{b,\mathrm{M}}^{(n_b+1)}) \tfrac{1}{4}(1 - \lambda\cos\theta)^2\right]_m \\ & \times \frac{1}{R} \frac{\mathrm{d}^{2m}}{\mathrm{d}(2R)^{2m}} \frac{1}{R} \frac{\mathrm{d}^{2(n_a+n_b-m+2)}}{\mathrm{d}(2R)^{2(n_a+n_b-m+2)}} \frac{1}{2R}, \end{aligned} \tag{72}$$

where $(a \leftrightarrow b)$ stands for the preceding terms with a and b interchanged. The square bracket expressions in (70)–(72) may be worked out by substituting (46), (47) for f, g and using for $a^L_{E,M}$ and $b^L_{E,M}$ the relations (36) and (40). In that way these bracket expressions become, respectively:

$$\sum_{\{L,s\}}^{(n_a,n_b)} [C^{ab}(\cos\theta)]_m \, f^a f^b, \tag{73}$$

$$\sum_{\{L,s\}}^{(n_a,n_b+1)} [C^{ab}(\cos\theta)]_m \, f^a g^b, \tag{74}$$

$$\sum_{\{L,s\}}^{(n_a+1,n_b+1)} [C^{ab}(\cos\theta)]_m \, g^a g^b, \tag{75}$$

with sums extending over $\{L,s\} \equiv \{L_a, L_{a1}, L_{a2}, s_{a1}, s_{a2}, L_b, L_{b1}, L_{b2}, s_{b1}, s_{b2}\}$. The brackets at the summation signs have an analogous interpretation as in (46), (47); for instance, the subsidiary conditions in (75) are $\frac{1}{2}(L_{a1} + L_{a2}) + s_{a1} + s_{a2} = n_a + 1$ and $\frac{1}{2}(L_{b1} + L_{b2}) + s_{b1} + s_{b2} = n_b + 1$. The atomic matrix elements are contained in the factors f and g defined as:

$$f(N; L, L_1, L_2, s_1, s_2) = (2e^2/m^2)\, k_N^{-1} \Omega_{N,L}(L_1, s_1)\, \Omega^*_{N,L}(L_2, s_2), \tag{76}$$

$$\begin{aligned} g(L, L_1, L_2, s_1, s_2) = {} & -(2e^2/m^2)\, \delta_{L+1,L}\, \delta_{L-1,L_2} \{(2L+1)^2/L(L+1)\} \\ & \times \sum_{N(\neq 0)} k_N^{-1} \Omega_{N,L}(L_1, s_1)\, \Omega^*_{N,L}(L_2, s_2) - (2e^2/m^2) \\ & \times \delta_{L,L_1} \delta_{L,L_2} \{ \sum_{N(\neq 0)} k_N^{-1} \Omega_{N,L}(L_1, s_1)\, \Omega^*_{N,L}(L_2, s_2) \\ & - \tfrac{1}{2} mM(L, s_1, s_2)\}, \end{aligned} \tag{77}$$

while the coefficients follow from the function:

$$\begin{aligned} & C^{ab}(L_a, L_{a1}, L_{a2}, L_b, L_{b1}, L_{b2}; \cos\theta) \\ & = \tfrac{1}{2} \sum_{M_1, M_2 = \pm 1} \prod_{i=1}^{2} \left\{ (2L_{ai}+1)(2L_{bi}+1) \begin{pmatrix} L_{ai} & 1 & L_a \\ 0 & M_i & -M_i \end{pmatrix} \right. \\ & \left. \times \begin{pmatrix} L_{bi} & 1 & L_b \\ 0 & M_i & -M_i \end{pmatrix} \right\} d^{(L_a)}_{M_1, M_2}(\theta)\, d^{(L_b)}_{-M_1, -M_2}(\theta). \end{aligned} \tag{78}$$

By employing the Clebsch–Gordan series (33) for the d-matrices these coefficients may be written as:

$$[C^{ab}(\cos\theta)]_M = \sum_L (2L+1)\, \bar{C}^{ab}\, [P_L(\cos\theta)]_m, \tag{79}$$

since $d^{(L)}_{00}(\theta) = P_L(\cos\theta)$. Here $\bar{C}^{ab}$ stands for the expression

$$\tfrac{1}{2}\prod_{i=1}^{2}\Big\{(2L_{ai}+1)(2L_{bi}+1) \times \sum_{M_i=\pm 1}\begin{pmatrix} L_{ai} & 1 & L_a \\ 0 & M_i & -M_i \end{pmatrix}\begin{pmatrix} L_{bi} & 1 & L_b \\ 0 & M_i & -M_i \end{pmatrix}\begin{pmatrix} L_a & L_b & L \\ M_i & -M_i & 0 \end{pmatrix}\Big\}, \tag{80}$$

which may be evaluated by extending the sum to the value $M_i = 0$ and subtracting subsequently that extra term; the result is:

$$\bar{C}^{ab} = \tfrac{1}{2}\prod_{i=1}^{2}\{(2L_{ai}+1)(2L_{bi}+1)\Big[(-)^{L_a+L_b}\begin{pmatrix} L_{ai} & L_{bi} & L \\ 0 & 0 & 0 \end{pmatrix}\begin{Bmatrix} L_a & L_b & L \\ L_{bi} & L_{ai} & 1 \end{Bmatrix} + \begin{pmatrix} L_{ai} & 1 & L_a \\ 0 & 0 & 0 \end{pmatrix}\begin{pmatrix} L_{bi} & 1 & L_b \\ 0 & 0 & 0 \end{pmatrix}\begin{pmatrix} L_a & L_b & L \\ 0 & 0 & 0 \end{pmatrix}\Big]\Big\}. \tag{81}$$

Substituting (79) into (73)–(75) and the results into (70)–(72) one obtains finally the following expressions for the interatomic dispersion energy:

$$V(R) = \sum_{\sigma=\mathrm{I},\mathrm{II},\mathrm{III}} V_\sigma(R), \tag{82}$$

$$V_{\mathrm{I}}(R) = \frac{1}{16\pi^3}\sum_{N_a(\neq 0),N_b(\neq 0)}\sum_{\{L,s\},L}\frac{2L+1}{k_{N_a}^2 - k_{N_b}^2}\bar{C}^{ab} \times f^a f^b \sum_{m=0}^{\frac{1}{2}N}[P_L]_m \frac{1}{R}\frac{\mathrm{d}^{2m}}{\mathrm{d}(2R)^{2m}}\frac{1}{R}\frac{\mathrm{d}^{N-2m}}{\mathrm{d}(2R)^{N-2m}}\left\{\frac{P(2k_{N_a}R)}{k_{N_a}} - \frac{P(2k_{N_b}R)}{k_{N_b}}\right\}, \tag{83}$$

$$V_{\mathrm{II}}(R) = -\frac{1}{16\pi^3}\sum_{N_a(\neq 0)}\sum_{\{L,s\},L}(2L+1)\bar{C}^{ab} \times f^a g^b \sum_{m=0}^{\frac{1}{2}N-1}[P_L]_m \frac{1}{R}\frac{\mathrm{d}^{2m}}{\mathrm{d}(2R)^{2m}}\frac{1}{R}\frac{\mathrm{d}^{N-2m-2}}{\mathrm{d}(2R)^{N-2m-2}}\frac{P(2k_{N_a}R)}{k_{N_a}} + (a \leftrightarrow b), \tag{84}$$

$$V_{\mathrm{III}}(R) = -\frac{1}{16\pi^3}\sum_{\{L,s\},L}(2L+1)\bar{C}^{ab} \times g^a g^b \sum_{m=0}^{\frac{1}{2}N-2}[P_L]_m \frac{1}{R}\frac{\mathrm{d}^{2m}}{\mathrm{d}(2R)^{2m}}\frac{1}{R}\frac{\mathrm{d}^{N-2m-4}}{\mathrm{d}(2R)^{N-2m-4}}\frac{1}{2R}, \tag{85}$$

with coefficients $[P_L]_m$ and $\bar{C}^{ab}$ given in (56) and (81), respectively, atomic matrix element functions $f^{a,b}$ and $g^{a,b}$ defined in (76) and (77) (with (25) and (44)) and the abbreviation $N = \sum_{i=1}^{2} (L_{ai} + L_{bi} + 2s_{ai} + 2s_{bi} + 2)$.

The present results may be compared with those obtained in paper II of this series. When using, for an arbitrary function $f(2R)$, the identity, proved in appendix C,

$$\sum_{m=0}^{L} [P_L]_m \frac{1}{R} \frac{\mathrm{d}^{2m}}{\mathrm{d}(2R)^{2m}} \frac{1}{R} \frac{\mathrm{d}^{N_1+N_2-2m}}{\mathrm{d}(2R)^{N_1+N_2-2m}} f(2R)$$

$$= R_1^L \left(\frac{1}{R_1} \frac{\mathrm{d}}{\mathrm{d}R_1}\right)^L \frac{1}{R_1} \frac{\mathrm{d}^{N_1-L}}{\mathrm{d}R_1^{N_1-L}} R_2^L \left(\frac{1}{R_2} \frac{\mathrm{d}}{\mathrm{d}R_2}\right)^L$$

$$\times \frac{1}{R_2} \frac{\mathrm{d}^{N_2-L}}{\mathrm{d}R_2^{N_2-L}} f(R_1 + R_2), \tag{86}$$

$(0 \leqslant L \leqslant \min(N_1, N_2))$, where R_1 and R_2 are to be put equal to R when the differentiations have been performed, one recovers directly from (83) the expression (II.44) with (II.36) and (II.39–41) for $V_{\mathrm{I}}(R)$. Likewise one may derive from (84) and (85) the expressions for V_{II} and V_{III} found in paper II, by employing the sum rules (B5) and (B6) in a judicious way.

5. *The interatomic dispersion energy for large separations.* The asymptotic form of the dispersion energy for large interatomic separations may be obtained from the behaviour of the weight function $\varrho(t)$ for small t[2]). In fact, the expression (65) shows that one may write

$$\varrho(t) = \sum_{n=2}^{\infty} \varrho_n t^n, \tag{87}$$

so that $\varrho(t)$ is proportional to t^2 for small t. From (16) one finds then for the dispersion energy an expansion in powers of R^{-1}:

$$V(R) = \sum_{n=2}^{\infty} \frac{(2n+1)!\,\varrho_n}{2\pi R^{2n+3}}. \tag{88}$$

For large interatomic separations the dominant term is determined by ϱ_2. It has the form:

$$\varrho_2 = -\frac{1}{(16\pi)^2} \sum_{i,j=\mathrm{E,M}} \left\{ \sum_{N_a(\neq 0)} k_{N_a}^{-2} f_{a,i}^{(N_a,0,0)} + g_{a,i}^{(1,0)} \right\}$$

$$\times \left\{ \sum_{N_b(\neq 0)} k_{N_b}^{-2} f_{b,j}^{(N_b,0,0)} + g_{b,j}^{(1,0)} \right\} \int_0^1 \mathrm{d}x\, \varphi_{ij}(x) \tag{89}$$

[from (59) with (37), (40), (46) and (47) it follows that $n_a = 0$, $n_b = 0$ implies $m_a = 0$, $m_b = 0$ in (65)]. The definitions (64) imply:

$$\begin{aligned} \int_0^1 \mathrm{d}x\, \varphi_{ij}(x) &= 23/15 \qquad \text{for} \qquad i,j = \mathrm{EE}, \mathrm{MM}, \\ &= -7/15 \qquad \text{for} \qquad i,j = \mathrm{EM}, \mathrm{ME}, \end{aligned} \tag{90}$$

while the expressions between curly brackets in (89) follow from (59) with (48) and (50). Thus the dominant term in the dispersion energy for large interatomic separations is found to be:

$$\begin{aligned} V^{\mathrm{L}}(R) = -\frac{e^4}{(4\pi)^3 R^7} \Big\{ & 23 \Big(\sum_{N_a(\neq 0)} \tfrac{2}{3} k_{N_a}^{-1} |r_{N_a}|^2 \Big) \Big(\sum_{N_b(\neq 0)} \tfrac{2}{3} k_{N_b}^{-1} |r_{N_b}|^2 \Big) \\ & + 23\, (\tfrac{1}{6} m^{-1} \langle 0 |r_a^2| 0 \rangle)\, (\tfrac{1}{6} m^{-1} \langle 0 |r_b^2| 0 \rangle \\ & + 7 \Big(\sum_{N_a(\neq 0)} \tfrac{2}{3} k_{N_a}^{-1} |r_{N_a}|^2 \Big) (\tfrac{1}{6} m^{-1} \langle 0 |r_b^2| 0 \rangle) \\ & + 7\, (\tfrac{1}{6} m^{-1} \langle 0| r_a^2 |0 \rangle) \Big(\sum_{N_b(\neq 0)} \tfrac{2}{3} k_{N_b}^{-1} |r_{N_b}|^2 \Big) \Big\}, \end{aligned} \tag{91}$$

where the fact has been used that in the magnetic form factor only the diamagnetic term contributes in the present case of spherically symmetric ground-state atoms. By employing the sum rule

$$m^{-1} \langle 0| r^2 |0 \rangle = \tfrac{1}{5} \sum_{N(\neq 0)} k_N |\mathsf{q}_N|^2, \tag{92}$$

which follows from (B6) with $L = 1$ and $s_1 = s_2 = 0$, (44) and the relation

$$\Omega_{N,2}(1, 0) = -(im/3\sqrt{6})\, k_N \mathsf{q}_N, \tag{93}$$

[*v.* (III.10)], with q_N the reduced matrix element of the electric quadrupole moment, $(4\pi/5)^{\frac{1}{2}} \langle 0 \| r^2 Y_2(\hat{r}) \| N, 2 \rangle$, one recovers the expressions (III.1), (III.15) and (III.18) derived in paper III of this series. In contrast to those expressions the present result (91) shows clearly the symmetry of the asymptotic form of the dispersion energy with respect to electric and (dia)magnetic phenomena, as could be expected on the basis of the expressions (50) for the form factors characterizing the emission of a pair of soft photons.

APPENDIX A

Analytical properties of the Fourier transform of the interatomic dispersion energy. In this appendix the expression (6), with (11) inserted, for the Fourier transform $F(\boldsymbol{Q})$ of the interatomic interaction energy will be studied. Since this energy is

purely dispersive for atoms in nondegenerate ground states only the dispersive part of $F(\boldsymbol{Q})$ will be considered. Due to the rotation invariance of the atomic hamiltonians $F(\boldsymbol{Q})$ depends for such atoms on the momentum transfer $t = -Q^2$ only.

From (11) it follows that the product of vertex functions occurring in the dispersive contribution to (6) may be written in the following form (with $k^0 = -k'^0$):

$$
\begin{aligned}
&\Gamma_{a,\mu\nu}(k, k')\,\Gamma_b^{\mu\nu}(-k, -k') \\
&= \sum_{k_\alpha(\neq 0),\, k_\beta(\neq 0)} \frac{f_{ab}^{(1)}(k^2, k'^2, \boldsymbol{k}\cdot\boldsymbol{k'}; k^{02})}{(k_\alpha^2 - k^{02} - \mathrm{i}0)\,(k_\beta^2 - k^{02} - \mathrm{i}0)} \\
&\quad + \sum_{k_\alpha(\neq 0)} \frac{f_{ab}^{(2)}(k^2, k'^2, \boldsymbol{k}\cdot\boldsymbol{k'})}{k_\alpha^2 - k^{02} - \mathrm{i}0} + \sum_{k_\beta(\neq 0)} \frac{f_{ba}^{(2)}(k^2, k'^2, \boldsymbol{k}\cdot\boldsymbol{k'})}{k_\beta^2 - k^{02} - \mathrm{i}0} \\
&\quad + f_{ab}^{(3)}(k^2, k'^2, \boldsymbol{k}\cdot\boldsymbol{k'}).
\end{aligned}
\tag{A1}
$$

Here the summations over α and β, at fixed k_α and k_β, are performed already. The functions $f_{ab}^{(i)}$ contain the matrix elements (12) and are thus regular in their arguments; in view of the property (9) they are symmetric in k^2 and k'^2. The general form (A1), which shows explicitly the nature and position of the poles of the products of vertex functions in the complex k^0 plane, will be useful in the discussion of the analytical properties of the function $F(t)$.

For the first contribution $F^{(1)}(t)$ to $F(t)$, obtained by inserting the first term of (A1) into (6), one finds upon introduction of the variable $\boldsymbol{\kappa}$ according to $\boldsymbol{k} = \frac{1}{2}\boldsymbol{Q} + \boldsymbol{\kappa}$, $\boldsymbol{k'} = \frac{1}{2}\boldsymbol{Q} - \boldsymbol{\kappa}$:

$$
\begin{aligned}
F^{(1)}(t) = \frac{\mathrm{i}}{2\,(2\pi)^3} \sum_{k_\alpha(\neq 0),\, k_\beta(\neq 0)} \int \frac{\mathrm{d}k^0\,\mathrm{d}\kappa_\parallel\,\mathrm{d}\kappa_\perp \kappa_\perp f_{ab}^{(1)}(Q^2, \kappa_\perp^2, \kappa_\parallel^2; k^{02})}{(k_\alpha^2 - k^{02} - \mathrm{i}0)\,(k_\beta^2 - k^{02} - \mathrm{i}0)} \\
\times [\{k^{02} - (\kappa_\parallel + \tfrac{1}{2}Q)^2 - \kappa_\perp^2 + \mathrm{i}0\} \\
\times \{k^{02} - (\kappa_\parallel - \tfrac{1}{2}Q)^2 - \kappa_\perp^2 + \mathrm{i}0\}]^{-1}
\end{aligned}
\tag{A2}
$$

with $Q^2 = -t$. Here we introduced cylindrical coordinates for $\boldsymbol{\kappa}$ with $\kappa_\parallel = Q^{-1}\boldsymbol{Q}\cdot\boldsymbol{\kappa}$ and $\kappa_\perp = (\kappa^2 - \kappa_\parallel^2)^{\frac{1}{2}}$. Since $f_{ab}^{(1)}$ is symmetric under an interchange of k^2 and k'^2 it is an even function of $\kappa_\parallel$.

The integral over k^0 in (A2) may be performed by closing the contour in the lower half of the complex k^0 plane. Since $f_{ab}^{(1)}$ is of second degree in k^0, the large semicircle gives no contribution. The poles below the real axis are situated at the positions $k^0 = R_\pm \equiv \{(\kappa_\parallel^2 \pm \frac{1}{2}Q)^2 + \kappa_\perp^2 - \mathrm{i}0\}^{\frac{1}{2}}$, $k^0 = k_\alpha - \mathrm{i}0$ and $k^0 = k_\beta - \mathrm{i}0$, where the square roots lying just below the positive real axis are meant. Evaluating the residues we find for $F^{(1)}(t)$ a sum of two terms, resulting from the first and second pair of poles, respectively:

$$
F^{(1)}(t) = (4\pi)^{-2} \sum_{k_\alpha(\neq 0),\, k_\beta(\neq 0)} \int \mathrm{d}\kappa_\parallel\,\mathrm{d}\kappa_\perp\,\kappa_\perp \sum_{i=1}^{2} \varphi_i(Q^2, \kappa_\perp^2, \kappa_\parallel^2),
\tag{A3}
$$

with the abbreviations:

$$\varphi_1 = \frac{1}{(R_+^2 - R_-^2)} \left\{ \frac{f_{ab}^{(1)}(Q^2, \varkappa_\perp^2, \varkappa_\parallel^2; R_+^2)}{R_+ (R_+^2 - k_\alpha^2)(R_+^2 - k_\beta^2)} - (R_+ \leftrightarrow R_-) \right\}, \tag{A4}$$

$$\varphi_2 = \frac{1}{k_\alpha^2 - k_\beta^2} \left\{ \frac{f_{ab}^{(1)}(Q^2, \varkappa_\perp^2, \varkappa_\parallel^2; k_\alpha^2)}{k_\alpha (R_+^2 - k_\alpha^2)(R_-^2 - k_\alpha^2)} - (k_\alpha \leftrightarrow k_\beta) \right\}. \tag{A5}$$

The symbol $(R_+ \leftrightarrow R_-)$ stands for the preceding terms with R_+ and R_- interchanged; $(k_\alpha \leftrightarrow k_\beta)$ has an analogous interpretation. If $\varkappa_\parallel$ tends to zero the roots R_+ and R_- coincide; the function φ_1 remains finite, however, in this limit. Likewise φ_2 remains finite if k_α and k_β tend to the same value.

In the complex $\varkappa_\parallel$ plane the integrand of (A3) has various singularities: branchpoints are present at the positions $\varkappa_\parallel = -\frac{1}{2}Q \pm \mathrm{i}(\varkappa_\perp^2 - \mathrm{i}0)^{\frac{1}{2}}$ and $\varkappa_\parallel = \frac{1}{2}Q \pm \mathrm{i}(\varkappa_\perp^2 - \mathrm{i}0)^{\frac{1}{2}}$, corresponding to the square roots R_+ and R_-, respectively. As a consequence the integrand in (A3) is defined, for $\varkappa_\perp \neq 0$, $Q \neq 0$, on four Riemann sheets, as a function of complex $\varkappa_\parallel$; these sheets (labelled by roman numbers) are connected across cuts in the way indicated in fig. 1. To identify the various sheets the signs of the roots R_+ and R_- along the real $\varkappa_\parallel$ axis have been indicated. The integration path C_0 that must be followed in evaluating (A3) extends over sheets I, II and IV. For $\varkappa_\perp$ tending to zero it is pinched by branchpoints which are then collapsing pairwise. Indeed, the integral over $\varkappa_\parallel$ is logarithmically divergent for

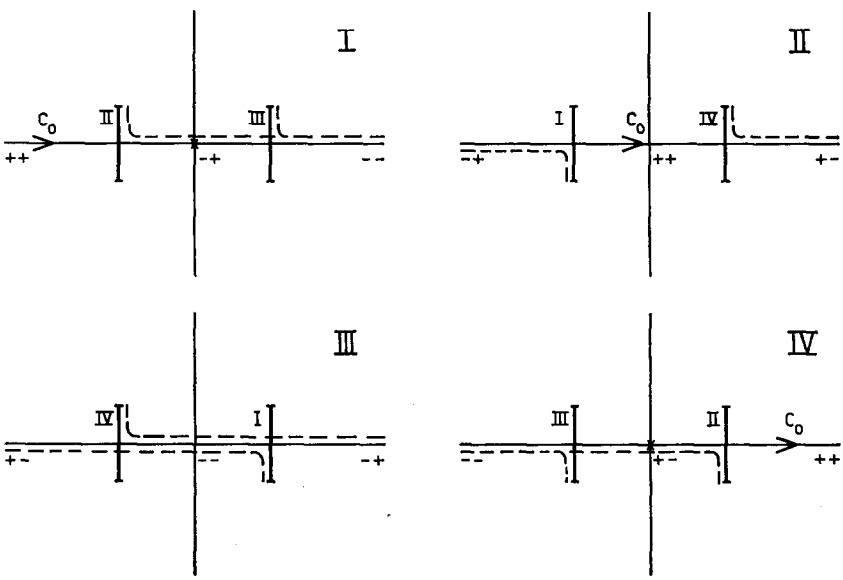

Fig. 1. Riemann sheets of the integrand of $F^{(1)}(t)$ as a function of complex $\varkappa_\parallel$.

$\varkappa_\perp = 0$. On multiplication by the explicitly present factor $\varkappa_\perp$ this end-point singularity for the $\varkappa_\perp$-integration disappears, however, leading to a finite result for $F^{(1)}(t)$.

Apart from the branchpoints the integrand of $F^{(1)}(t)$ (A3) has a number of poles. First of all, poles occur for $\varkappa_\parallel = 0$ on those sheets where R_+ and R_- have opposite sign, *i.e.*, on sheets I and IV (on sheet II, where the integration path C_0 passes $\varkappa_\parallel = 0$, no pole is present at this value); they have been indicated by crosses in fig. 1. Furthermore, series of poles occur for those $\varkappa_\parallel$ that satisfy one of the four equations $R_\pm = -k_\alpha$, $R_\pm = -k_\beta$ [for all k_α and k_β appearing in the sums in (A3)]. The first of these equations, for instance, with the upper sign, has solutions $\varkappa_\parallel = -\frac{1}{2}Q \pm (-\varkappa_\perp^2 + k_\alpha^2 + \mathrm{i}0)^{\frac{1}{2}}$, leading to poles on those sheets where R_+ is negative for these values of $\varkappa_\parallel$. The location of the series of poles corresponding to all four equations have been indicated in fig. 1 by dashed lines (since k_α and k_β are positive the lines are bounded in one direction). Inspection of the figure shows that the integration path C_0 is in fact not hindered by any of these poles: they are located on sheets different from those on which C_0 is situated.

The function $F^{(1)}(t)$ (A3) may be continued analytically in the complex t plane, by continuing the integrand for complex values of Q, starting from $Q > 0$ (corresponding to $t = -Q^2 < 0$), and deforming the integration contours continually to avoid singularities in the integrand.

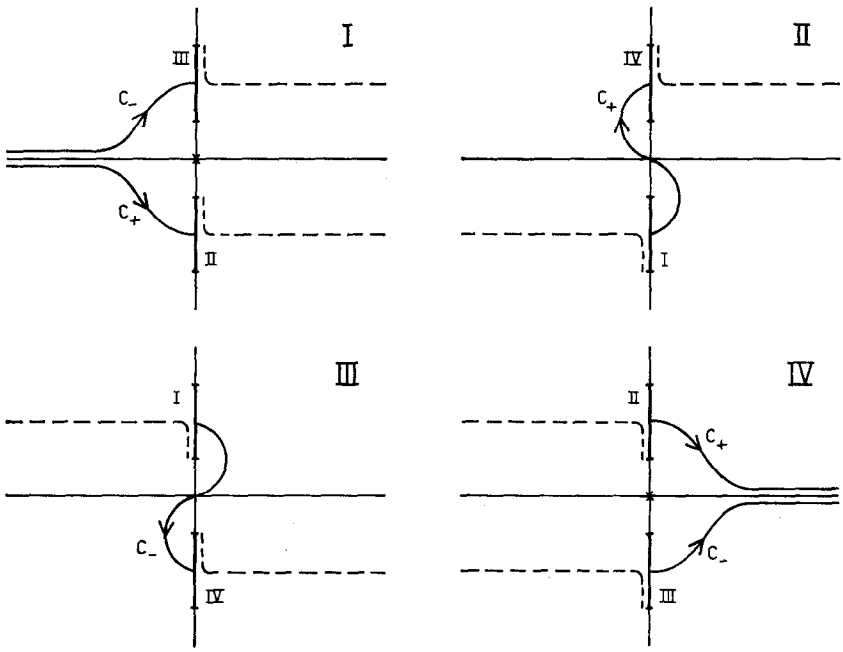

Fig. 2. Riemann sheets of the integrand of $F^{(1)}(t)$ as a function of complex $\varkappa_\parallel$, after analytical continuation in t, for $0 < \varkappa_\perp < \frac{1}{2}|Q|$.

When continuing analytically into the lower half t plane by writing $Q = |Q| e^{i\psi}$, with ψ running from 0 to $\frac{1}{2}\pi$, one finds that the cuts in the complex $\varkappa_{\|}$ plane corresponding to the branchpoints of the integrand in (A3) are rotating counter-clockwise over ψ. The $\varkappa_{\|}$-integration path has to be deformed to pass in the same order as before through the cuts and to avoid the poles. In the course of this analytical continuation (*i.e.*, for $0 \leqq \psi < \frac{1}{2}\pi$) no singularities are encountered that may pinch the contour in the $\varkappa_{\|}$ plane (except for the harmless one connected with $\varkappa_{\perp} = 0$), so that a function $F^{(1)}(t)$ regular in the lower half t plane is obtained. For the final value $\frac{1}{2}\pi$ of ψ the position of the cuts and the poles on the Riemann sheets of the integrand $F^{(1)}(t)$ as a function of $\varkappa_{\|}$, for $0 < \varkappa_{\perp} < \frac{1}{2}|Q|$, have been drawn in fig. 2; the integration path to be followed for the evaluation of $F^{(1)}(t)$ has been labelled there by C_+.

When the function $F^{(1)}(t)$ is analytically continued from the negative t axis into the upper half plane, according to $Q = |Q| e^{i\psi}$ with ψ running from 0 to $-\frac{1}{2}\pi$, the cuts in the Riemann sheets of the integrand are rotating clockwise. For $\psi = -\frac{1}{2}\pi$ one arrives at the same integrand as for $\psi = \frac{1}{2}\pi$ discussed above, as inspection of (A3)–(A5) shows. Thus, the Riemann sheets of the integrand at $\psi = -\frac{1}{2}\pi$ are again represented by fig. 2, where I, II, III and IV result now from I, III, II and IV in fig. 1, respectively. However, the path of integration for $\varkappa_{\|}$ is different in the present case. As a consequence of the continuous path deformation one ends up with the contour labelled C_- in fig. 2. Thus $F^{(1)}(t)$ can be continued analytically from the negative t axis into the complex t plane, leading to a function regular in the whole plane with the exception of the positive t axis. Depending on the way the continuation is carried out different values are found along the positive t axis: the function has a cut along that axis, starting from $t = 0$. The discontinuity over the cut may be evaluated by taking the difference between the integrals over $\varkappa_{\|}$ along the contours C_+ and C_-. In calculating this difference we may use the invariance properties of the integrand of $F^{(1)}(t)$ in going from one sheet to another. In fact one may prove, from the change of sign of $R_{\pm}$ when passing the corresponding cuts, that the following relations connect the functions φ_1 and φ_2 on the various sheets in fig. 2:

$$\begin{aligned} &\varphi_1(\mathrm{I}) = -\varphi_1(\mathrm{IV}), \qquad \varphi_1(\mathrm{II}) = -\varphi_1(\mathrm{III}), \\ &\varphi_2(\mathrm{I}) = \varphi_2(\mathrm{II}) = \varphi_2(\mathrm{III}) = \varphi_2(\mathrm{IV}). \end{aligned} \tag{A6}$$

As a consequence it follows that the contour $C_+ - C_-$ may be reduced to a small circular path in counter-clockwise direction around the pole at $\varkappa_{\|} = 0$ on sheet I in fig. 2. In deriving this result we used the fact that the branchpoints correspond to square roots so that integration paths going twice around them give a vanishing contribution to the integral.

The above discussion was valid only when $0 < \varkappa_{\perp} < \frac{1}{2}|Q|$. For $\varkappa_{\perp} > \frac{1}{2}|Q|$ the cuts are no longer separated after rotation over $\pm\frac{1}{2}\pi$. Both the contours C_+ and C_- can then be chosen to follow the real axis on the Riemann sheets I and IV. As a

consequence no contribution to the discontinuity of $F^{(1)}(t)$ across the cut along the positive t axis arises from the integration range $\varkappa_\perp > \frac{1}{2}|Q|$.

The discontinuity of $F^{(1)}(t)$ across the cut along the positive t axis is obtained now by evaluating, for $0 < \varkappa_\perp < \frac{1}{2}|Q|$ the residue of the pole at $\varkappa_\parallel = 0$ of the integrand of (A3) on the Riemann sheet I of fig. 2 and integrating the result over $\varkappa_\perp$. Since only φ_1, given in (A4), has a pole there one finds for the discontinuity across the cut:

$$F^{(1)}(t + \mathrm{i}0) - F^{(1)}(t - \mathrm{i}0)$$
$$= -\frac{\mathrm{i}}{8\pi} \sum_{k_\alpha(\neq 0),\, k_\beta(\neq 0)} \int_0^{\frac{1}{2}\sqrt{t}} \varkappa_\perp \, \mathrm{d}\varkappa_\perp \frac{f_{ab}^{(1)}(-t, \varkappa_\perp^2, 0; \varkappa_\perp^2 - \frac{1}{4}t)}{[t(\frac{1}{4}t - \varkappa_\perp^2)]^{\frac{1}{2}}}$$
$$\times \frac{1}{(k_\alpha^2 + \frac{1}{4}t - \varkappa_\perp^2)(k_\beta^2 + \frac{1}{4}t - \varkappa_\perp^2)}, \qquad (t > 0). \tag{A7}$$

Comparison of this expression with (A1) and (A2) shows that the discontinuity of $F^{(1)}(t)$ is given by an integral over $\varkappa_\perp$ of the analytical continuation [to positive values of $t = -Q^2 = -(\boldsymbol{k} + \boldsymbol{k}')^2$] of the first term of $\Gamma_{a,\mu\nu}(k, k')\, \Gamma_b^{\mu\nu}(-k, -k')$ [(A1)], for values of $k^0 = -k'^0$ and $\boldsymbol{k} = \frac{1}{2}\boldsymbol{Q} + \boldsymbol{\kappa}$, $k' = \frac{1}{2}\boldsymbol{Q} - \boldsymbol{\kappa}$ satisfying the relations $k^{02} = \frac{1}{4}Q^2 + \varkappa^2$, $\boldsymbol{Q} \cdot \boldsymbol{\kappa} = 0$ or $k^{02} = k^2 = k'^2$. In other words, to evaluate the discontinuity across the cut the vertex functions have to be known only on the photon mass shells. Such a state of affairs could be expected on the basis of Cutkosky's rules[8]) for the determination of discontinuities of covariant scattering amplitudes. The application of these rules to the present case of atoms with fixed nuclei is not altogether straightforward; therefore an independent approach was preferred.

The study of the contributions to $F(t)$, arising from the second to fourth terms in (A1) leads to similar results: again these contributions may be continued analytically to regular functions in the cut t plane. [The contribution to $F(t)$ resulting from the last term in (A1) can be made convergent by adding a cutoff factor.] As a consequence one may write the discontinuity of $F(t)$ across the cut in a form analogous to that of (A7) and given in (14).

APPENDIX B

Sum rules. In this appendix sum rules will be derived for the Ω-matrix elements defined in (25). A convenient starting point for this derivation is the expression:

$$\sum_{N(\neq 0),\, M,\, M_2,\, \lambda} 4\pi\, \mathrm{i} m k_N^{-1} \{(2s_1)!!\,(2s_2)!!\,(2L + 2s_1 + 1)!!\,(2L + 2s_2 + 1)!!\}^{-1}$$
$$\times \begin{pmatrix} L & 1 & L_2 \\ M & \lambda & M_2 \end{pmatrix} \langle 0 |\, [H, r^{L+2s_1} Y_L^{-M}(\hat{\boldsymbol{r}})]\, | N, L, M \rangle$$
$$\times \langle N, L, M |\, \tfrac{1}{2}\{p^{-\lambda}, r^{L_2+2s_2} Y_{L_2}^{-M_2}(\hat{\boldsymbol{r}})\}\, | 0 \rangle. \tag{B1}$$

In fact, sum rules will be obtained by rewriting it in two different ways and equating the results.

The projection operators on states with the same energy are real in the coordinate representation, since the (nonrelativistic) atomic hamiltonian H is real in this representation. Since the parity invariance of the hamiltonian implies that only terms with $L + L_2 + 1$ even contribute in (B1), this expression may be shown to be real. Taking half the sum of (B1) and its complex conjugate one may write it as:

$$- \sum_{M, M_2, \lambda} 2\pi\, im \{(2s_1)!!\,(2s_2)!!\,(2L + 2s_1 + 1)!!\,(2L + 2s_2 + 1)!!\}^{-1}$$

$$\times \begin{pmatrix} L & 1 & L_2 \\ M & \lambda & M_2 \end{pmatrix} \langle 0| \,[r^{L+2s_1} Y_L^{-M}(\hat{r}),\, \tfrac{1}{2}\{p^{-\lambda},\, r^{L_2+2s_2} Y_{L_2}^{-M_2}(\hat{r})\}\,|0\rangle. \qquad \text{(B2)}$$

The commutator can be evaluated by means of the gradient formula[5]). Since the rotation-invariant part of $Y_{L_1}^{M_1}(\hat{r})\, Y_{L_2}^{M_2}(\hat{r})$ is $(4\pi)^{-1}(-)^{M_1}\delta_{L_1,L_2}\delta_{M_1,-M_2}$ one arrives then at the result:

$$\tfrac{1}{2}m\{(2L + 1)(2L_2 + 1)\}^{\frac{1}{2}}\{(2s_1)!!\,(2s_2)!!\,(2L + 2s_1 + 1)!!\,(2L + 2s_2 + 1)!!\}^{-1}$$

$$\times \begin{pmatrix} L & 1 & L_2 \\ 0 & 0 & 0 \end{pmatrix} \{\delta_{L_2, L+1}\, 2s_1 + \delta_{L_2, L-1}(2L + 2s_1 + 1)\}$$

$$\times \langle 0|\, r^{L+L_2+2s_1+2s_2-1}\,|0\rangle. \qquad \text{(B3)}$$

The expression (B1) may be rewritten in a different way by using the gradient formula to evaluate the commutator in the first matrix element. Then one obtains, with the definition (25) of the Ω-matrix elements:

$$\sum_{N(\neq 0)} k_N^{-1}\{(2L_2 + 1)(2L + 1)\}^{\frac{1}{2}}\left\{(2L + 3)\begin{pmatrix} L & 1 & L+1 \\ 0 & 0 & 0 \end{pmatrix}\right.$$

$$\times \Omega_{NL}(L + 1, s_1 - 1)\,\Omega^*_{N,L}(L_2, s_2) + (2L - 1)$$

$$\left.\times \begin{pmatrix} L & 1 & L-1 \\ 0 & 0 & 0 \end{pmatrix} \Omega_{N,L}(L - 1, s_1)\,\Omega^*_{N,L}(L_2, s_2)\right\}. \qquad \text{(B4)}$$

Equating the expressions (B3) and (B4) one finds, by putting $L_2 = L \pm 1$, two types of sum rules; upon insertion of algebraic expressions for the $3j$-symbols[5]) they get the form:

$$\tfrac{1}{2}mM(L, s_1, s_2) = \sum_{N(\neq 0)} k_N^{-1}(2L - 1)^{-1}\{(2L + 1)/L\}^{\frac{1}{2}}$$

$$\times [\{L(2L + 1)\}^{\frac{1}{2}}\Omega_{N,L-1}(L, s_1)\,\Omega^*_{N,L-1}(L, s_2)$$

$$- \{(L - 1)(2L - 3)\}^{\frac{1}{2}}\Omega_{N,L-1}(L - 2, s_1 + 1)\,\Omega^*_{N,L-1}(L, s_2)], \qquad \text{(B5)}$$

$$\tfrac{1}{2}mM(L, s_1, s_2) = \sum_{N(\neq 0)} k_N^{-1}(2L+3)^{-1}\{(2L+1)/(L+1)\}^{\frac{1}{2}}$$
$$\times [\{(L+1)(2L+1)\}^{\frac{1}{2}}\Omega_{N,L+1}(L, s_1)\Omega^*_{N,L+1}(L, s_2)$$
$$- \{(L+2)(2L+5)\}^{\frac{1}{2}}\Omega_{N,L+1}(L+2, s_1-1)\Omega^*_{N,L+1}(L, s_2)], \qquad \text{(B6)}$$

with the matrix element $M(L, s_1, s_2)$ defined in (44). When employing these sum rules the reality of the expression

$$\sum_{N, \text{fixed } k_N} \Omega_{NL}(L_1, s_1)\Omega^*_{N,L}(L_2, s_2) \qquad \text{(B7)}$$

may be used to eliminate terms $\Omega_{N,L}(L_1, s_1)\Omega^*_{N,L}(L_2, s_2)$ with $L_1 < L_2$. This property follows from the definition (25) of the Ω-matrix element together with the reality of the nonrelativistic atomic hamiltonian H and the atomic angular momentum L^2 in the coordinate representation.

The sum rules derived in paper III of this series, *viz.* (III.A5) and (III.A9) are special cases of the results (B5)–(B6) obtained here. In fact (III.A5) follows from (B5) with $L = 1$, while the information contained in (III.A9) may be derived from (B5) for $s_1 = -1$ and from (B6) for $s_1 = 0$. (It should be borne in mind that the definitions (25) and (44) imply that the Ω- and M-matrix elements vanish for negative values of the parameters s.)

APPENDIX C

An identity for differentiations. The proof of the identity (86) starts by writing the arbitrary function $f(R_1 + R_2)$ appearing there as a Fourier integral:

$$f(R_1 + R_2) = \int_0^\infty \mathrm{d}k\, \phi(k) \sin(kR_1 + kR_2)$$
$$= -\int_0^\infty \mathrm{d}k\, \phi(k) k^2 R_1 R_2 \{j_0(kR_1) y_0(kR_2) + y_0(kR_1) j_0(kR_2)\}. \qquad \text{(C1)}$$

Then the identity

$$R^L \left(\frac{1}{R}\frac{\mathrm{d}}{\mathrm{d}R}\right)^L \frac{1}{R}\frac{\mathrm{d}^{N-L}}{\mathrm{d}R^{N-L}} Rf_0(kR) = (\mathrm{i}k)^N \mathrm{i}^L f_L(kR) \qquad \text{(C2)}$$

($N \geqslant L$) valid for spherical Bessel functions f_L of the first and second type [*v.* (II.A14) with (II.A16)] may be used to write the right-hand side of (86) in the form:

$$D(N, L) f(R_1 + R_2) = 2\int_0^\infty \mathrm{d}k\, \phi(k) (\mathrm{i}k)^{N+2} (-)^L j_L(kR) y_L(kR), \qquad \text{(C3)}$$

where we employed the abbreviation:

$$D(N, L) = R_1^L \left(\frac{1}{R_1}\frac{\mathrm{d}}{\mathrm{d}R_1}\right)^L \frac{1}{R_1}\frac{\mathrm{d}^{N_1-L}}{\mathrm{d}R_1^{N_1-L}} R_2^L \left(\frac{1}{R_2}\frac{\mathrm{d}}{\mathrm{d}R_2}\right)^L \frac{1}{R_2}\frac{\mathrm{d}^{N_2-L}}{\mathrm{d}R_2^{N_2-L}} \tag{C4}$$

with $N_1 + N_2 = N$. In the left-hand side of (C3) R_1 and R_2 are to be replaced by R when the differentiations have been performed. With the help of the recurrence relations[7]) for the Bessel functions the equality

$$R^{-1}\{\mathrm{d}^2/\mathrm{d}(2R)^2\} RD(N, L) = \tfrac{1}{2}L(2L+1)^{-1} D(N+2, L-1) + \tfrac{1}{2}D(N+2, L) + \tfrac{1}{2}(L+1)(2L+1)^{-1} D(N+2, L+1) \tag{C5}$$

may be derived from (C3) so that, by successive elimination, it follows that $D(N, L)$ can be expanded in the following way:

$$D(N, L) = \sum_m c_m^L R^{-1}\{\mathrm{d}^{2m}/\mathrm{d}(2R)^{2m}\} RD(N-2m, 0), \tag{C6}$$

with coefficients c_m^L independent of N. Insertion into (C5) yields the following recursion relation for the coefficients:

$$2(2L+1)c_{m-1}^L = (L+1)c_m^{L+1} + (2L+1)c_m^L + Lc_m^{L-1}; \tag{C7}$$

it has to be solved under the initial constraint $c_m^0 = \delta_{m0}$. The generating function

$$F(x, y) = \sum_{L,m} c_m^L x^L y^m, \tag{C8}$$

with $F(0, y) = 1$, may be shown to satisfy a linear first-order differential equation in x of which the solution is:

$$F(x, y) = \{x^2 - 2x(2y-1) + 1\}^{-\frac{1}{2}} = \sum_{L=0}^{\infty} P_L(2y-1)\, x^L, \tag{C9}$$

where Legendre polynomials appear. With the use of (54) and (56) one obtains thus:

$$c_m^L = [P_L]_m = \frac{(-)^{L+m}(L+m)!}{(L-m)!\,(m!)^2}, \tag{C10}$$

so that, upon comparing (C4) and (C6), one arrives indeed at the relation (86).

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