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MULTIPOLE EXPANSION OF THE RETARDED INTERATOMIC POTENTIAL ENERGY

VI. DISPERSION AND INDUCTION ENERGY FOR RELATIVISTIC HYDROGEN ATOMS

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Synopsis

The inductive and dispersive retarded interaction energies of two ground-state hydrogen atoms described by Dirac theory are derived up to all multipole orders. The results are obtained by evaluation of Feynman diagrams and with the help of dispersion-relation methods. The non-relativistic and semi-relativistic approximations of the interaction energy are given in a form that shows explicitly the contributions of electron spin.

1. *Introduction.* In a series of papers[1,2]) the multipole expansion for the retarded interaction energy of two ground-state atoms has been obtained on the basis of non-relativistic quantum electrodynamics. Since the atoms were described by Schrödinger wave functions, electron spin contributions did not occur in the final results.

In the present paper the retarded interaction energy will be derived for a pair of ground-state hydrogen atoms that are described by relativistic electron theory. In this way spin terms are included *a priori*; moreover, the use of relativistic quantum electrodynamics leads to considerable simplifications in the calculations.

The energy shifts due to one- and two-photon exchange between the atoms are obtained from the scattering matrix in section 2. In the following section the Feynman diagrams are evaluated; the resulting interaction energy is expressed in terms of atomic one-photon vertex functions, which are subsequently expanded into cartesian multipoles in section 4. Then, in section 5 the non-relativistic and semi-relativistic approximations of the vertex functions are studied with the help of a Foldy–Wouthuysen transformation; the resulting expression for the interaction energy shows explicitly the contributions due to electron spin. The radial behaviour of the interaction energy becomes more transparent from the expansion in terms of generalized spherical multipoles, which is derived in sec-

tion 6 with the help of the Racah formalism. An alternative way to obtain this multipole expansion from the scattering matrix makes use of dispersion relation methods, as is shown in section 7. Finally, in section 8, the lowest-order multipole contributions to the interaction energy are discussed, both relativistically and in the non-relativistic approximation.

2. *The interatomic potential energy due to one- and two-photon exchange.* The interaction energy of two hydrogen atoms a and b with fixed nuclei may be obtained from the scattering matrix S_{fi} for a transition between the initial and final states i and f of the pair of atoms[3]). In fact, the scattering matrix will have the general form:

$$S_{fi} = \delta_{fi} - 2\pi i \delta (E_f - E_i) V_{fi}, \tag{1}$$

where a delta function of the difference between the initial and final energies E_i and E_f appears. The interatomic potential energy in the state i = f follows as the part of V_{fi} that depends on the internuclear separation $\boldsymbol{R} = \boldsymbol{R}_b - \boldsymbol{R}_a$.

The contribution to the S-matrix due to one- and two-photon exchange between the atomic electrons, with charges $-e$, reads*:

$$\begin{aligned} S_{fi} = \delta_{fi} &- ie^2 \int d^4R_1\, d^4R_2 \bar{\psi}_{\alpha f}(R_1)\, \gamma^{\mu} \psi_{\alpha i}(R_1)\, g_{\mu\mu'} D_F(R_1 - R_2) \\ &\times \bar{\psi}_{\beta f}(R_2)\, \gamma^{\mu'} \psi_{\beta i}(R_2) + e^4 \int d^4R_1 \cdots d^4R_4 \bar{\psi}_{\alpha f}(R_1)\, \gamma^{\mu} S_{Fa}(R_1, R_2) \\ &\times \gamma^{\nu} \psi_{\alpha i}(R_2)\, [g_{\mu\mu'} g_{\nu\nu'} D_F(R_1 - R_3)\, D_F(R_2 - R_4) \\ &+ g_{\mu\nu'} g_{\mu'\nu} D_F(R_1 - R_4)\, D_F(R_2 - R_3)]\, \bar{\psi}_{\beta f}(R_3)\, \gamma^{\mu'} S_{Fb}(R_3, R_4) \\ &\times \gamma^{\nu'} \psi_{\beta i}(R_4), \end{aligned} \tag{2}$$

where the initial and final electron states are characterized by Dirac wave functions $\psi_{\alpha i}$, $\psi_{\beta i}$ and $\psi_{\alpha f}$, $\psi_{\beta f}$, respectively. The progagator for the electron of atom a in the electrostatic field of its nucleus is given by

$$S_{Fa}(R_1, R_2) = -\frac{1}{2\pi} \sum_{\alpha} \psi_{\alpha}(R_1)\, \bar{\psi}_{\alpha}(R_2) \int_{-\infty}^{\infty} d\omega_a \frac{e^{i\omega_a (t_1 - t_2)}}{E_{\alpha}(1 - i0) + \omega_a}, \tag{3}$$

where α labels all positive- and negative-energy eigenfunctions $\psi_{\alpha}(R) = \psi_{\alpha}(\boldsymbol{R}) \times \exp(-iE_{\alpha}t)$ of the electron. The photon propagator in (2) is defined as

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

* The metric tensor $g^{\mu\nu}$ is chosen as diag $(1, -1, -1, -1)$. Rationalized gaussian units, with $\hbar$ and c put equal to unity, are used throughout.

In writing the expression (2) the overlap of the electronic wave functions of atom a and b was assumed to be negligible, so that electronic exchange needed not be taken into account.

For two hydrogen atoms in their ground states the interaction energy $V(\boldsymbol{R})$ is found, after inserting (3) and (4) into (2) and carrying out the integrations over the time variables and ω_a, ω_b. When the average is taken over the ground states of both atoms independently, $V(\boldsymbol{R})$ becomes a function of the internuclear distance R and has the form:

$$V(R) = (2\pi)^{-3} \int \mathrm{d}\boldsymbol{Q}\, \mathrm{e}^{\mathrm{i}\boldsymbol{Q}\cdot\boldsymbol{R}} [F_1(\boldsymbol{Q}) + F_2(\boldsymbol{Q})]. \tag{5}$$

The contributions, due to one- and two-photon exchange, to the Fourier transform are

$$F_1(\boldsymbol{Q}) = -\frac{1}{g_a g_b} \sum_{\alpha_0, \beta_0} \int \mathrm{d}^4k \Gamma_{\alpha_0\alpha_0,\mu}(k)\, \Gamma^{\mu}_{\beta_0\beta_0}(-k)\, \frac{\delta(k^0)\, \delta(\boldsymbol{k} - \boldsymbol{Q})}{k^2 + \mathrm{i}0}, \tag{6}$$

$$F_2(\boldsymbol{Q}) = \frac{1}{g_a g_b} \sum_{\alpha_0, \beta_0} \frac{\mathrm{i}}{2(2\pi)^4} \int \mathrm{d}^4k\, \mathrm{d}^4k' \Gamma_{\alpha_0\alpha_0,\,\mu\nu}(k, k')\, \Gamma^{\mu\nu}_{\beta_0\beta_0}(-k, -k')$$

$$\times \frac{\delta(k^0 + k'^0)\, \delta(\boldsymbol{k} + \boldsymbol{k}' - \boldsymbol{Q})}{(k^2 + \mathrm{i}0)(k'^2 + \mathrm{i}0)}, \tag{7}$$

where g_a, g_b are the orders of degeneracy of the atomic ground states labelled α_0, β_0. The one- and two-photon vertex functions are defined by

$$\Gamma^{\mu}_{\alpha'\alpha}(k) = -e \langle \alpha' | \gamma^0 \gamma^{\mu} \mathrm{e}^{-\mathrm{i}k\cdot r} - g^{0\mu} | \alpha \rangle, \tag{8}$$

$$\Gamma^{\mu\nu}_{\alpha_0\alpha_0}(k, k') = \sum_{\alpha} \left[\frac{\Gamma^{\mu}_{\alpha_0\alpha}(k)\, \Gamma^{\nu}_{\alpha\alpha_0}(k')}{E_{\alpha}(1 - \mathrm{i}0) - E_{\alpha_0} - k^0} + \frac{\Gamma^{\nu}_{\alpha_0\alpha}(k')\, \Gamma^{\mu}_{\alpha\alpha_0}(k)}{E_{\alpha}(1 - \mathrm{i}0) - E_{\alpha_0} - k'^0} \right], \tag{9}$$

with $\boldsymbol{r}$ the position of the electron relative to its nucleus. By adding the tensor $g^{0\mu}$ in the one-photon vertex function the contribution of the fixed nuclei to the interaction energy has been taken into account. When (6)–(9) are substituted into (5) the two-photon exchange term contains sums over intermediate atomic states α, β; according to the prescription for obtaining energy shifts from the S-matrix, the divergent part due to intermediate states with energies equal to that of the initial state (*i.e.*, in the present case states with $E_{\alpha} = E_{\alpha_0}$, $E_{\beta} = E_{\beta_0}$) has to be suppressed.

For the projection operator P_E on the set of atomic states with energy E time reversal invariance of the atomic hamiltonian implies

$$P_E^* = T^{-1} P_E T; \tag{10}$$

here the matrix T satisfies the relation:

$$\gamma_\mu^* = T^{-1}\gamma^\mu T. \tag{11}$$

As a consequence the space components of the one-photon vertex function, averaged over the ground states, vanish, so that in (6) only the time components of $\Gamma^\mu(k)$ remain. If the overlap of the atomic wave functions is negligible a vanishing contribution to $V(R)$ is found, however, as follows from rotation invariance arguments. Thus in the interaction energy (5) only the two-photon exchange term has to be considered.

3. *Evaluation of the two-photon exchange contribution.* Upon substitution of (9) into (7) the following two integrals show up:

$$I_{\mathrm{ncr}}(k_\alpha, k_\beta) = \int \mathrm{d}k^0 \{[E_\alpha(1 - \mathrm{i}0) - E_{\alpha_0} - k^0][E_\beta(1 - \mathrm{i}0) - E_{\beta_0} + k^0] \times (k^2 + \mathrm{i}0)(k'^2 + \mathrm{i}0)\}^{-1}, \tag{12}$$

$$I_{\mathrm{cr}}(k_\alpha, k_\beta) = \int \mathrm{d}k^0 \{[E_\alpha(1 - \mathrm{i}0) - E_{\alpha_0} - k^0][E_\beta(1 - \mathrm{i}0) - E_{\beta_0} - k^0] \times (k^2 + \mathrm{i}0)(k'^2 + \mathrm{i}0)\}^{-1} \tag{13}$$

(with $k_\alpha = E_\alpha - E_{\alpha_0}$), referring to Feynman diagrams with non-crossing and crossing photon lines. From (12) and (13) the following symmetry properties may be obtained:

$$I_{\mathrm{ncr}}(k_\alpha, k_\beta) = I_{\mathrm{ncr}}(-k_\alpha, -k_\beta) = -I_{\mathrm{cr}}(-k_\alpha, k_\beta) = -I_{\mathrm{cr}}(k_\alpha, -k_\beta), \tag{14}$$

so that the integrals have to be evaluated only for $k_\alpha \geqslant 0$, $k_\beta \geqslant 0$ (with the exclusion of $k_\alpha = k_\beta = 0$). By closing the integration paths in the complex k^0 plane one finds:

$$I_{\mathrm{ncr}}(k_\alpha, k_\beta) = \frac{\pi\mathrm{i}}{2kk'(k_\alpha + k_\beta)}\left[\frac{1}{(k_\alpha + k)(k + k')} + \frac{1}{(k_\alpha + k')(k + k')} + \frac{1}{(k_\alpha + k)(k_\alpha + k')}\right] + (\alpha \leftrightarrow \beta), \tag{15}$$

$$I_{\mathrm{cr}}(k_\alpha, k_\beta) = -\frac{\pi\mathrm{i}}{2kk'(k_\alpha - k_\beta)}\left[\frac{1}{(k_\alpha + k)(k + k')} + \frac{1}{(k_\alpha + k')(k + k')} + \frac{1}{(k_\alpha + k)(k_\alpha + k')}\right] + (\alpha \leftrightarrow \beta), \tag{16}$$

where the symbol $(\alpha \leftrightarrow \beta)$ stands for the terms obtained from the preceding ones by an interchange of α and β.

The integrations over the space components of the photon momenta may be dealt with after replacing in the expression (5) with (7)–(9) the exponential $\exp(i\boldsymbol{Q} \cdot \boldsymbol{R})$ by $\exp(i\boldsymbol{k} \cdot \boldsymbol{R}_1 + i\boldsymbol{k}' \cdot \boldsymbol{R}_2)$, and subsequently in the matrix elements (8) the momenta $\boldsymbol{k}$ and $\boldsymbol{k}'$ by the operator $-i\nabla_1$ and $-i\nabla_2$, respectively. (When the differentiations with respect to $\boldsymbol{R}_1$ and $\boldsymbol{R}_2$ have been performed, both vectors are to be put equal to the radius vector $\boldsymbol{R}$.) The integrals over $\boldsymbol{k}$ and $\boldsymbol{k}'$ may now be evaluated with the help of the relation (for $k_\alpha \geqslant 0$):

$$\int \frac{d\boldsymbol{k}}{4\pi} \frac{e^{i\boldsymbol{k}\cdot\boldsymbol{R}_1}}{k} \int \frac{d\boldsymbol{k}'}{4\pi} \frac{e^{i\boldsymbol{k}'\cdot\boldsymbol{R}_2}}{k'} \left[\frac{1}{(k_\alpha + k)(k + k')} + \frac{1}{(k_\alpha + k')(k + k')} + \frac{1}{(k_\alpha + k)(k_\alpha + k')} \right] = \frac{\pi P(k_\alpha R_1 + k_\alpha R_2)}{R_1 R_2}. \tag{17}$$

The function $P(x)$ is defined as:

$$P(x) = \int_0^\infty dt \frac{\sin t}{x + t}; \tag{18}$$

it obeys the differentiation formulae:

$$dP/dx = -Q(x), \qquad d^2P/dx^2 = -P(x) + x^{-1}, \tag{19}$$

where $Q(x)$, on a par with $P(x)$, is given by

$$Q(x) = \int_0^\infty dt \frac{\cos t}{x + t}. \tag{20}$$

As a result the interatomic potential energy is obtained as a sum of two terms, arising from the diagrams with non-crossing and crossing photon lines:

$$V(R) = -(g_a g_b)^{-1} \sum_{\alpha_0, \beta_0} \sum_{\alpha, \beta\,(k_\alpha, k_\beta \neq 0, 0)} \tfrac{1}{2}(2\pi)^{-3} \operatorname{Sgn}(E_\alpha) [(k_\alpha + k_\beta)^{-1} \times \Gamma_{\alpha 1} \cdot \Gamma_{\beta 1} \Gamma^*_{\alpha 2} \cdot \Gamma^*_{\beta 2} - (k_\alpha - k_\beta)^{-1} \Gamma_{\alpha 1} \cdot \Gamma^*_{\beta 1} \Gamma^*_{\alpha 2} \cdot \Gamma_{\beta 2}] \times P(|k_\alpha R_1 + k_\alpha R_2|)/R_1 R_2 + (\alpha \leftrightarrow \beta), \tag{21}$$

where $\operatorname{Sgn}(E_\alpha) = E_\alpha/|E_\alpha|$. The symbols $\Gamma^\mu_{\alpha i}$ and $\Gamma^\mu_{\beta i}$ stand for the matrix elements $\Gamma^\mu_{\alpha_0 \alpha}(-i\nabla_i)$ and $\Gamma^\mu_{\beta_0 \beta}(i\nabla_i)$, with $i = 1, 2$. From the time-reversal relations (10)

and (11) it may be shown that these matrix elements have the following property with respect to complex conjugation:

$$\sum_{\alpha_0} \sum_{\alpha(k_\alpha \text{ const})} (\Gamma^{\mu}_{\alpha 1} \Gamma^{\nu *}_{\alpha 2} - \Gamma^{*}_{\alpha 1,\mu} \Gamma_{\alpha 2,\nu}) = 0, \tag{22}$$

which permits to rewrite (21) in the form:

$$\begin{aligned} V(R) = (g_a g_b)^{-1} \sum_{\alpha_0, \beta_0} \sum_{\alpha, \beta (k_\alpha, k_\beta \neq 0, 0)} (2\pi)^{-3} \operatorname{Sgn}(E_\alpha) (k_\alpha^2 - k_\beta^2)^{-1} \\ \times [k_\beta (\Gamma^0_{\alpha 1} \Gamma^0_{\beta 1} \Gamma^{0*}_{\alpha 2} \Gamma^{0*}_{\beta 2} + \boldsymbol{\Gamma}_{\alpha 1} \cdot \boldsymbol{\Gamma}_{\beta 1} \boldsymbol{\Gamma}^{*}_{\alpha 2} \cdot \boldsymbol{\Gamma}^{*}_{\beta 2}) + k_\alpha (\Gamma^0_{\alpha 1} \Gamma^0_{\beta 1} \Gamma^{*}_{\alpha 2} \cdot \boldsymbol{\Gamma}^{*}_{\beta 2} \\ + \boldsymbol{\Gamma}_{\alpha 1} \cdot \boldsymbol{\Gamma}_{\beta 1} \Gamma^{0*}_{\alpha 2} \Gamma^{0*}_{\beta 2})] P(|k_\alpha R_1 + k_\alpha R_2|)/R_1 R_2 + (\alpha \leftrightarrow \beta). \end{aligned} \tag{23}$$

As in the non-relativistic case (*v.* papers I and IV of this series) the expression for the interatomic potential energy may be split up into two terms, with both atoms and with only one atom in excited intermediate states; these contributions, which are the dispersive and the inductive part of the interaction energy, respectively, will be evaluated separately in the following section.

4. *Cartesian multipole expansion of the dispersion and the induction energy.* The dispersive part of the interaction energy follows by restricting the sum over α, β in (23) to intermediate states with $k_\alpha \neq 0$, $k_\beta \neq 0$. Then one may express $V_{\text{disp}}(R)$ completely in terms of the space components of the one-photon vertex functions with the help of the relation

$$\Gamma^0_{\alpha i} = -ik_\alpha^{-1} \nabla_i \cdot \boldsymbol{\Gamma}_{\alpha i}, \tag{24}$$

which follows by commuting the operator in $\Gamma^0_{\alpha i}$ [*v.* (8)] with the atomic hamiltonian. In fact, one gets with the help of (19):

$$\begin{aligned} V_{\text{disp}}(R) = (g_a g_b)^{-1} \sum_{\alpha_0, \beta_0} \sum_{\alpha, \beta (k_\alpha \neq 0, k_\beta \neq 0)} (2\pi)^{-3} (k_\alpha k_\beta)^{-1} \\ \times \{|k_\alpha|^{-1} (k_\alpha^2 - k_\beta^2)^{-1} \boldsymbol{\Gamma}_{\alpha 1} \boldsymbol{\Gamma}_{\beta 1} : (\mathsf{U} \Delta_1 - \nabla_1 \nabla_1) \\ \times \boldsymbol{\Gamma}^{*}_{\alpha 2} \boldsymbol{\Gamma}^{*}_{\beta 2} : (\mathsf{U} \Delta_2 - \nabla_2 \nabla_2) P(|k_\alpha R_1 + k_\alpha R_2|)/R_1 R_2 \\ - |k_\alpha| \boldsymbol{\Gamma}_{\alpha 1} \cdot \boldsymbol{\Gamma}_{\beta 1} \boldsymbol{\Gamma}^{*}_{\alpha 2} \cdot \boldsymbol{\Gamma}^{*}_{\beta 2} [P(|k_\alpha R_1 + k_\alpha R_2|) \\ - \tfrac{1}{2} |k_\alpha R_1 + k_\alpha R_2|^{-1}]/R_1 R_2\} + (\alpha \leftrightarrow \beta). \end{aligned} \tag{25}$$

As a consequence of the occurrence of the function $P(x)$ the dependence of the dispersion energy on powers of R is different for small and large values of $k_\alpha R$ (with k_α a characteristic reciprocal atomic wavelength) due to retardation effects for large separations.

From the general expression (23) the induction energy is obtained by putting $k_x = 0$, $k_\beta \neq 0$ and *vice versa*. Substituting the value $\frac{1}{2}\pi$ for $P(0)$ one finds for the induction energy:

$$V_{\text{ind}}(R) = -(g_a g_b)^{-1} \sum_{\alpha_0, \beta_0} \sum_{\alpha, \beta\,(k_\alpha = 0, k_\beta \neq 0)} (4\pi)^{-2} k_\beta^{-1} \times (\Gamma^0_{\alpha 1}\Gamma^0_{\beta 1}\Gamma^{0*}_{\alpha 2}\Gamma^{0*}_{\beta 2} + \boldsymbol{\Gamma}_{\alpha 1} \cdot \boldsymbol{\Gamma}_{\beta 1}\boldsymbol{\Gamma}^*_{\alpha 2} \cdot \boldsymbol{\Gamma}^*_{\beta 2})\, 1/R_1 R_2 + (\alpha \leftrightarrow \beta). \quad (26)$$

As in paper IV the induction energy may be understood as the sum of the energy shifts for each atom in the static electric and magnetic fields generated by the other, which can be calculated from second-order perturbation theory with the interaction hamiltonian $H_{\text{int}} = -e\gamma^0\gamma_\mu A^\mu$. Indeed, the expression (26) has non-retarded character, in contrast to (25).

The expressions for the dispersion and induction energies may be expanded in terms of cartesian multipole-moment matrix elements, which are defined as

$$\boldsymbol{\mu}^{(n)}_\alpha = -e \langle \alpha_0 |\, (1/n!)\, \boldsymbol{r}^n \,| \alpha \rangle, \quad (27)$$

$$\mathbf{v}^{(n)}_\alpha = -e \langle \alpha_0 |\, [n/(n+1)!]\, \boldsymbol{r}^{n-1}\boldsymbol{r} \wedge \gamma^0\boldsymbol{\gamma} \,| \alpha \rangle \quad (28)$$

(*v.* also appendix A). The one-photon vertex functions $\Gamma^\mu_{\alpha i}$, which are in fact charge and current matrix elements, are expanded in terms of the multipoles as follows:

$$\Gamma^0_{\alpha i} = \sum_{n=1}^{\infty} (-\nabla_i)^n \vdots \boldsymbol{\mu}^{(n)}_\alpha, \quad (29)$$

$$\boldsymbol{\Gamma}_{\alpha i} = \sum_{n=1}^{\infty} (-\nabla_i)^{n-1} \vdots (-\mathrm{i}k_\alpha \boldsymbol{\mu}^{(n)}_\alpha - \mathbf{v}^{(n)}_\alpha \wedge \nabla_i). \quad (30)$$

(An analogous expression is used to derive the Maxwell equations for the fields due to composite particles from those for the fields generated by a set of charged point particles[4]).) Upon substitution of (29) and (30) into (25) and (26) the complete cartesian multipole expansions of the interatomic dispersion and induction energies may be obtained. In particular it may be seen then which multipoles contribute to the long-range asymptotic interaction energy.

The long-range behaviour of the dispersion energy follows from the power-series expansion $P(x) = x^{-1} - 2x^{-3} + \cdots$. In the first term of (25) the leading contribution then contains electric dipoles for both atoms, while in the second term the sum rule (A17) implies the leading contribution to couple the magnetic dipoles of one of the atoms with both electric and magnetic dipoles of the other. Since due to the rotation symmetry of the atomic hamiltonian only the invariant part of the product of dipole matrix elements for each atom comes into play, the

long-range dispersion energy gets the form:

$$V^{L}_{disp}(R) = -(g_a g_b)^{-1} \sum_{\alpha_0,\beta_0} \sum_{\alpha,\beta\,(k_\alpha \neq 0,\, k_\beta \neq 0)} (23\,|\boldsymbol{\mu}^{(1)}_\alpha|^2\,|\boldsymbol{\mu}^{(1)}_\beta|^2 - 7\,|\boldsymbol{\mu}^{(1)}_\alpha|^2\,|\mathbf{v}^{(1)}_\beta|^2$$

$$- 7\,|\mathbf{v}^{(1)}_\alpha|^2\,|\boldsymbol{\mu}^{(1)}_\beta|^2 + 23\,|\mathbf{v}^{(1)}_\alpha|^2\,|\mathbf{v}^{(1)}_\beta|^2)/144\pi^3\,k_\alpha k_\beta R^7. \tag{31}$$

For the electric dipole part an expression of the same form was obtained already by Casimir and Polder[5]) in the framework of non-relativistic quantum electrodynamics. The extension to magnetic dipole moments (again in the non-relativistic approximation) was given by Mavroyannis and Stephen[6]), and Condiff[7]). Employing dispersion-relation methods (*v.* also section 7) Feinberg and Sucher[8]) derived the long-range form of the dispersion energy for neutral spinless particles in terms of static electric and magnetic polarizabilities.

Just as the long-range dispersion energy, the induction energy (26) for large separations contains only contributions of electric and magnetic dipoles:

$$V^{L}_{ind}(R) = -(g_a g_b)^{-1} \sum_{\alpha_0,\beta_0} \sum_{\alpha,\beta\,(k_\alpha = 0,\, k_\beta \neq 0)} (|\boldsymbol{\mu}^{(1)}_\alpha|^2\,|\boldsymbol{\mu}^{(1)}_\beta|^2$$

$$+ |\mathbf{v}^{(1)}_\alpha|^2\,|\mathbf{v}^{(1)}_\beta|^2)/24\,\pi^2 k_\beta R^6 + (\alpha \leftrightarrow \beta). \tag{32}$$

To extract the behaviour at arbitrary separation for dipole or higher-order multipole moments a large number of vectorial differentiations has to be carried out, as is obvious from (25) and (26) with (29) and (30) inserted. A more convenient form for the dispersion and induction energy is obtained by passing over to the spherical-tensor formalism. Before doing so the non-relativistic and semi-relativistic limits of (25) and (26) will be studied.

5. *The non-relativistic and semi-relativistic limit.* The atomic hamiltonian, *viz.*

$$H = \gamma^0 m + \gamma^0 \boldsymbol{\gamma} \cdot \boldsymbol{p} - e\varphi, \tag{33}$$

with $\varphi = e/4\pi r$ the nuclear potential, may be transformed in such a way that the positive- and negative-energy solutions are decoupled. In the Pauli representation for the γ-matrices the unitary Pryce–Foldy–Wouthuysen transformation for the free hamiltonian ($\varphi = 0$) reads:

$$U = [(E + m)/2E]^{\frac{1}{2}} + \boldsymbol{\gamma} \cdot \boldsymbol{p}/[2E\,(E + m)]^{\frac{1}{2}} \tag{34}$$

with $E = (p^2 + m^2)^{\frac{1}{2}}$. When this transformation is applied to (33) the hamiltonian still contains "odd" parts. It may be brought into even form up to any desired order in m^{-n}, or, more generally, in $m^{-n} f(\varphi m)$, by carrying out subsequently successive Foldy–Wouthuysen transformations. Up to order $m^{-3} f(\varphi m)$ this is achieved

by employing the transformation

$$U' = 1 - \mathrm{i}e\gamma^0\boldsymbol{\gamma}\cdot\boldsymbol{E}/4m^2 \tag{35}$$

(with $\boldsymbol{E} = -\nabla\varphi$), which is indeed unitary in this order of approximation. The resulting hamiltonian then becomes:

$$\hat{H} = \gamma^0\,(m + \tfrac{1}{2}m^{-1}p^2 - \tfrac{1}{8}m^{-3}p^4) - e\varphi - \tfrac{1}{4}em^{-2}\,(\boldsymbol{\sigma}\wedge\boldsymbol{p})\cdot\boldsymbol{E} + \tfrac{1}{8}em^{-2}\,\nabla\cdot\boldsymbol{E}. \tag{36}$$

Likewise, the charge and current operators appearing in the one-photon vertex function (8) are to be transformed with the help of $U'U$. The calculations, during which it is convenient to make use of the Weyl correspondence (see *e.g.* ref. 4), lead to the following expressions for the atomic matrix elements occurring in the interaction energy:

$$\Gamma^0_{\alpha_+} = -e\,\langle\alpha_0|\,\tfrac{1}{2}\,\{1 - \tfrac{1}{4}m^{-2}\,(\boldsymbol{p}\wedge\boldsymbol{\sigma})\cdot\nabla + \tfrac{1}{8}m^{-2}\Delta, \mathrm{e}^{-r\cdot\nabla}\} - 1\,|\alpha_+\rangle, \tag{37}$$

$$\Gamma^0_{\alpha_-} = -e\,\langle\alpha_0|\,\tfrac{1}{2}\,\{\tfrac{1}{2}\mathrm{i}m^{-1}\,\boldsymbol{\sigma}\cdot\nabla - \tfrac{1}{4}\,\mathrm{i}m^{-3}p^2\boldsymbol{\sigma}\cdot\nabla - \tfrac{1}{4}\,\mathrm{i}m^{-3}\boldsymbol{\sigma}\cdot\boldsymbol{p}\boldsymbol{p}\cdot\nabla - \tfrac{1}{16}\,\mathrm{i}m^{-3}\boldsymbol{\sigma}\cdot\nabla\Delta, \mathrm{e}^{-r\cdot\nabla}\}\,|\alpha_-\rangle, \tag{38}$$

$$\boldsymbol{\Gamma}_{\alpha_+} = -e\,\langle\alpha_0|\,\tfrac{1}{2}\,\{m^{-1}\boldsymbol{p} - \tfrac{1}{2}m^{-1}\boldsymbol{\sigma}\wedge\nabla - \tfrac{1}{2}m^{-3}p^2\boldsymbol{p} - \tfrac{1}{8}m^{-3}\boldsymbol{p}\cdot\nabla\nabla + \tfrac{1}{4}m^{-3}p^2\boldsymbol{\sigma}\wedge\nabla + \tfrac{1}{4}m^{-3}\boldsymbol{\sigma}\wedge\boldsymbol{p}\boldsymbol{p}\cdot\nabla + \tfrac{1}{16}\,m^{-3}\boldsymbol{\sigma}\wedge\nabla\Delta + \tfrac{1}{2}em^{-2}\boldsymbol{\sigma}\wedge\boldsymbol{E}, \mathrm{e}^{-r\cdot\nabla}\}\,|\alpha_+\rangle, \tag{39}$$

$$\boldsymbol{\Gamma}_{\alpha_-} = -e\,\langle\alpha_0|\,\tfrac{1}{2}\,\{\boldsymbol{\sigma} - \tfrac{1}{2}m^{-2}\boldsymbol{\sigma}\cdot\boldsymbol{p}\boldsymbol{p} + \tfrac{1}{4}m^{-2}\boldsymbol{p}\wedge\nabla - \tfrac{1}{4}m^{-2}\boldsymbol{\sigma}\cdot\nabla\nabla + \tfrac{1}{8}m^{-2}\boldsymbol{\sigma}\Delta, \mathrm{e}^{-r\cdot\nabla}\}\,|\alpha_-\rangle, \tag{40}$$

up to order $m^{-3}f(\varphi m)$. For brevity the suffix i at the nabla operator has been suppressed here; states with positive and negative energy have been labelled by α_+ and α_-, respectively. The results (37) and (39) are different from those given in ref. 9, the reason being that in the (non-unitary) transformation employed there the approximations up to the desired order have not been made correctly. With the help of (37)–(40) the interatomic dispersion and induction energies (25) and (26) may be obtained in the "semi-relativistic" approximation, which includes, beyond the non-relativistic terms, for instance spin–orbit coupling effects.

The non-relativistic limit of the interatomic potential energy, *i.e.*, that following from the Schrödinger–Pauli hamiltonian, may be derived from (25) and (26) by

retaining in (37) and (39) the leading terms:

$$\bar{\Gamma}^0_{\alpha_+} = -e \langle \alpha_0 | \, e^{-r\cdot\nabla} - 1 \, | \dot{\alpha}_+ \rangle, \tag{41}$$

$$\bar{\Gamma}_{\alpha_+} = -e \langle \alpha_0 | \, \tfrac{1}{2} \{ m^{-1} p - \tfrac{1}{2} m^{-1} \sigma \wedge \nabla, e^{-r\cdot\nabla} \} \, | \alpha_+ \rangle, \tag{42}$$

while the leading term of the negative-energy contribution (40) gives rise to a direct matrix element in the interaction energy:

$$g_a^{-1} \sum_{\alpha_0} \sum_{\alpha_-} f(k_{\alpha_-}) \bar{\Gamma}_{\alpha_- 1} \bar{\Gamma}^*_{\alpha_- 2} = g_a^{-1} \sum_{\alpha_0} f(-2m) \, e^2 \langle \alpha_0 | \, e^{-r\cdot(\nabla_1 + \nabla_2)} \, | \alpha_0 \rangle \, \mathsf{U}. \tag{43}$$

Indeed in the Schrödinger–Pauli hamiltonian a term linear in the scalar potential occurs, while both linear and quadratic terms in the vector potential are present; these three terms give rise to the matrix elements (41), (42) and (43). Inserting these matrix elements into (25) and (26) one recovers the results of papers I and IV, if moreover the Pauli spin terms are suppressed.

6. *Spherical-multipole expansion.* In this section the expressions (25) and (26) for the interatomic dispersion and induction energies will be evaluated in the framework of the spherical-tensor formalism[10]). Correspondingly the atomic states will be chosen as simultaneous eigenstates of the free-atomic hamiltonian, the total atomic angular momentum and its third component: $|\alpha\rangle = |N_a, J_a, M_a\rangle$, with eigenvalues E_{N_a}, $J_a(J_a + 1)$ and M_a, respectively. The ground states $|\alpha_0\rangle$ will be denoted by $|0, J_a^0, M_a^0\rangle$.

In the dispersion energy (25) the first term only contains transverse one-photon vertex functions $\Gamma_\perp = \Gamma \cdot (\mathsf{U} - \nabla\nabla/\Delta)$ (*v.* appendix B). In fact these occur in the combination:

$$X_{ab}(\nabla_1, \nabla_2) f(R_1 + R_2)/R_1 R_2 = (2J_a^0 + 1)^{-1} (2J_b^0 + 1)^{-1} \sum_{\substack{M_a^0, M_a \\ M_b^0, M_b}} \Gamma_{a1} \Gamma_{b1} :$$
$$\times (\mathsf{U}\Delta_1 - \nabla_1\nabla_1) \, \Gamma^*_{a2} \Gamma^*_{b2} : (\mathsf{U}\Delta_2 - \nabla_2\nabla_2) f(R_1 + R_2)/R_1 R_2. \tag{44}$$

The second term of (25) may be rewritten so as to contain this combination as well, if use is made of the sum rule (A17). In this way the dispersion energy may be cast into the form:

$$V_{disp}(R) = \sum_{\substack{N_a(\neq 0), J_a \\ N_b(\neq 0), J_b}} \frac{1}{8\pi^3 k_a k_b} X_{ab}(\nabla_1, \nabla_2) \frac{1}{R_1 R_2}$$
$$\times \{ (k_a^2 - k_b^2)^{-1} |k_a|^{-1} P(|k_a R_1 + k_a R_2|) - |k_a|^{-3}$$
$$\times [P(|k_a R_1 + k_a R_2|) - |k_a R_1 + k_a R_2| \log |k_a R_1 + k_a R_2|$$
$$+ \tfrac{1}{12} |k_a R_1 + k_a R_2|^3 \log |k_a R_1 + k_a R_2|] \} + (a \leftrightarrow b), \tag{45}$$

where the differentiation formulae (19) have been used.

Expression (44) can be evaluated with the methods of paper II. In particular the Rayleigh expansion may be used for the exponentials occurring in the matrix elements. Subsequently the resulting spherical harmonics $Y_L^M(\hat{r})$ are to be coupled with the matrix $\gamma^0\gamma = \alpha$ so as to yield the spherical tensor operator

$$\Omega_L^M(L', s) = -e \sum_{\lambda, M'} (-1)^{1-\lambda+L'-M'} (2L+1)^{\frac{1}{2}} \begin{pmatrix} L' & 1 & L \\ -M' & -\lambda & M \end{pmatrix} \times \frac{1}{(2s)!!\,(2L'+2s+1)!!} \left(\frac{4\pi}{2L'+1}\right)^{\frac{1}{2}} \alpha^\lambda r^{L'+2s} Y_{L'}^{M'}(\hat{r}), \quad (46)$$

the reduced matrix element of which is written as:

$$\Omega_{NJ}(L, L', s) = (2J^0+1)^{-\frac{1}{2}} \langle 0, J^0 \| \Omega_L(L', s) \| N, J \rangle. \quad (47)$$

Employing now the graphical techniques of paper II (sections 3, 4) to separate the radial and angular dependence [*v.* in particular formulae (28), (31)–(34), (36), (39)–(41) of paper II] one arrives at the following result for (44):

$$\sum_{\substack{L_a, L_{a1}, L_{a2}, L \\ L_b, L_{b1}, L_{b2}}} \sum_{\substack{s_{a1}, s_{a2} \\ s_{b1}, s_{b2}}} (2L+1)\, C^{ab} \Omega_{N_a J_a}(L_a, L_{a1}, s_{a1})\, \Omega_{N_b J_b}(L_b, L_{b1}, s_{b1}) \times \Omega^*_{N_a J_a}(L_a, L_{a2}, s_{a2})\, \Omega^*_{N_b J_b}(L_b, L_{b2}, s_{b2})\, D(L_{a1} + L_{b1} + 2s_{a1} + 2s_{b1} + 2, L_{a2} + L_{b2} + 2s_{a2} + 2s_{b2} + 2, L) f(R_1 + R_2). \quad (48)$$

Here the symbol D is defined as:

$$D(N_1, N_2, L) = R_1^L \left(\frac{1}{R_1}\frac{\mathrm{d}}{\mathrm{d}R_1}\right)^L \frac{1}{R_1} \left(\frac{\mathrm{d}}{\mathrm{d}R_1}\right)^{N_1-L} \times R_2^L \left(\frac{1}{R_2}\frac{\mathrm{d}}{\mathrm{d}R_2}\right)^L \frac{1}{R_2} \left(\frac{\mathrm{d}}{\mathrm{d}R_2}\right)^{N_2-L}. \quad (49)$$

When the differentiations have been carried out both R_1 and R_2 are to be put equal to the interatomic separation R. Then (49) may be rewritten in the form (II. 45):

$$D(N_1 + N_2, L) = \sum_{k_1, k_2=0}^{L} \frac{4(-1)^{k_1+k_2}(L+k_1)!\,(L+k_2)!}{k_1!k_2!\,(L-k_1)!\,(L-k_2)!} \times \left(\frac{1}{2R}\right)^{k_1+k_2+2} \left(\frac{\mathrm{d}}{\mathrm{d}2R}\right)^{N_1+N_2-k_1-k_2} \quad (50)$$

[*v.* also (V. 86)]. The symbol C^{ab} in (48) stands for the angular coefficient:

$$C^{ab} = \prod_{i=1,2} (2L_{ai} + 1)(2L_{bi} + 1) \left[(-1)^{L_a+L_b} \begin{pmatrix} L_{ai} & L_{bi} & L \\ 0 & 0 & 0 \end{pmatrix} \begin{Bmatrix} L_a & L_b & L \\ L_{bi} & L_{ai} & 1 \end{Bmatrix} \right.$$
$$\left. + \begin{pmatrix} 1 & L_a & L_{ai} \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} 1 & L_b & L_{bi} \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} L_a & L_b & L \\ 0 & 0 & 0 \end{pmatrix} \right], \tag{51}$$

which may be written in an alternative form by using the definition of the 6*j*-symbol:

$$C^{ab} = \prod_{i=1,2} (2L_{ai} + 1)(2L_{bi} + 1) \sum_{\lambda_i = \pm 1} \begin{pmatrix} 1 & L_a & L_{ai} \\ \lambda_i & -\lambda_i & 0 \end{pmatrix}$$
$$\times \begin{pmatrix} 1 & L_b & L_{bi} \\ \lambda_i & -\lambda_i & 0 \end{pmatrix} \begin{pmatrix} L_a & L_b & L \\ \lambda_i & -\lambda_i & 0 \end{pmatrix}. \tag{52}$$

From this form for C^{ab} it follows that in (48) the atomic matrix elements occur in the combinations (B13) and (B15) which contain the transverse electric and magnetic multipole matrix elements $\varkappa$ and ν. Writing (48) in terms of these matrix elements one gets:

$$\sum_{\substack{L_a, S_a, L \\ L_b, S_b}} (2L + 1) \begin{pmatrix} L_a & L_b & L \\ 1 & -1 & 0 \end{pmatrix}^2 \{ \delta_{L_a+L_b+L, \text{ even}} [\mathscr{K}_{N_aJ_a}(L_a, S_a)\, \mathscr{K}_{N_bJ_b}(L_b, S_b)$$
$$\times D(N, L) + \mathscr{N}_{N_aJ_a}(L_a, S_a) \mathscr{N}_{N_bJ_b}(L_b, S_b)\, D(N + 4, L)]$$
$$+ \delta_{L_a+L_b+L, \text{ odd}} [\mathscr{K}_{N_aJ_a}(L_a, S_a) \mathscr{N}_{N_bJ_b}(L_b, S_b)\, D(N + 2, L)$$
$$+ \mathscr{N}_{N_aJ_a}(L_a, S_a)\, \mathscr{K}_{N_bJ_b}(L_b, S_b)\, D(N + 2, L)]\} f(2R), \tag{53}$$

with $N = 2(L_a + L_b + S_a + S_b)$.

Upon insertion of the form (53) for the expression (44), the dispersion energy (45) is obtained as a sum of three terms, representing interactions between the electric (both static and transverse) and magnetic multipole moments, given in (B1), (B10) and (B11):

$$V_{\text{disp, e-e}}(R) = (2\pi)^{-3} \sum_{\substack{N_a(\neq 0), J_a \\ N_b(\neq 0), J_b}} \sum_{\substack{L_a, S_a, L \\ L_b, S_b}} \delta_{L_a+L_b+L, \text{ even}} (2L + 1) \begin{pmatrix} L_a & L_b & L \\ 1 & -1 & 0 \end{pmatrix}^2$$
$$\times \{ \delta_{S_a,0} \delta_{S_b,0} \mathscr{M}_{N_aJ_a}(L_a, S_a) \mathscr{M}_{N_bJ_b}(L_b, S_b)\, D(N, L)$$
$$- [\delta_{S_a,0}(1 - \delta_{S_b,0}) \mathscr{M}_{N_aJ_a}(L_a, S_a)\, \mathscr{K}_{N_bJ_b}(L_b, S_b)$$
$$+ (a \leftrightarrow b)]\, D(N - 2, L) + (1 - \delta_{S_a,0})(1 - \delta_{S_b,0})$$
$$\times \mathscr{K}_{N_aJ_a}(L_a, S_a)\, \mathscr{K}_{N_bJ_b}(L_b, S_b)\, D(N - 4, L)\}$$
$$\times \frac{k_a k_b}{k_a^2 - k_b^2} \left[\frac{P(2|k_a|R)}{|k_a|} - \frac{P(2|k_b|R)}{|k_b|} \right], \tag{54}$$

$$V_{\text{disp, e-m}}(R) = (2\pi)^{-3} \sum_{\substack{N_a(\neq 0), J_a \\ N_b(\neq 0), J_b}} \sum_{\substack{L_a, S_a, L \\ L_b, S_b}} \delta_{L_a+L_b+L, \text{odd}} (2L+1) \begin{pmatrix} L_a & L_b & L \\ 1 & -1 & 0 \end{pmatrix}^2$$

$$\times [-\delta_{S_a,0} \mathscr{M}_{N_aJ_a}(L_a, S_a) \mathscr{N}_{N_bJ_b}(L_b, S_b) D(N, L)$$

$$+ (1 - \delta_{S_a,0}) \mathscr{K}_{N_aJ_a}(L_a, S_a) \mathscr{N}_{N_bJ_b}(L_b, S_b) D(N-2, L)$$

$$+ (a \leftrightarrow b)] \frac{k_a k_b}{k_a^2 - k_b^2} \left[\frac{P(2|k_a|R)}{|k_a|} - \frac{P(2|k_b|R)}{|k_b|} \right], \tag{55}$$

$$V_{\text{disp, m-m}}(R) = (2\pi)^{-3} \sum_{\substack{N_a(\neq 0), J_a \\ N_b(\neq 0), J_b}} \sum_{\substack{L_a, S_a, L \\ L_b, S_b}} \delta_{L_a+L_b+L, \text{even}} (2L+1) \begin{pmatrix} L_a & L_b & L \\ 1 & -1 & 0 \end{pmatrix}^2$$

$$\times \mathscr{N}_{N_aJ_a}(L_a, S_a) \mathscr{N}_{N_bJ_b}(L_b, S_b) D(N, L)$$

$$\times \frac{k_a k_b}{k_a^2 - k_b^2} \left[\frac{P(2|k_a|R)}{|k_a|} - \frac{P(2|k_b|R)}{|k_b|} \right]. \tag{56}$$

In the terms with parameter value $S = 0$ the product $\mathscr{M}_{NJ}(L, S)$ (B14) of electrostatic multipole matrix elements could be introduced with the help of (B12). Furthermore, the sum rule (B20) and the differentiation relations (19) have been employed in order to arrive at (54)–(56).

In the expression (26) for the induction energy the radial function $1/R_1R_2$ occurs, for which the two-centre expansion gets a simple form (*v.* paper II, appendix A). The first part of (26) may then be expressed in terms of the longitudinal electric multipole moments μ (B1) with $s = 0$, which are the static electric multipole moments. In the second part both the transverse electric and magnetic multipole matrix elements $\varkappa$ and ν [(B10) and (B11)] show up, again only with $s = 0$; employing the fact that for one of the atoms only ground level intermediate states occur and making use of the sum rule (B20), with (B12), one finds that the transverse electric multipole moments $\varkappa$ do not contribute, so that only the static magnetic multipole moments ν come into play. As a result the induction energy may be cast into the form:

$$V_{\text{ind}}(R) = -\sum_{N_b(\neq 0), J_b} \sum_{L_a, L_b} \frac{1}{16\pi^2 k_b} \frac{(2L_a + 2L_b)!}{4^{L_a+L_b}} \binom{2L_a}{L_a - 1} \binom{2L_b}{L_b - 1}$$

$$\times [\mathscr{M}_{0J_a0}(L_a, 0) \mathscr{M}_{N_bJ_b}(L_b, 0) + \mathscr{N}_{0J_a0}(L_a, 0) \mathscr{N}_{N_bJ_b}(L_b, 0)]$$

$$\times \frac{1}{R^{2L_a+2L_b+2}} + (a \leftrightarrow b), \tag{57}$$

where the abbreviations (B13)–(B15) have been used.

In this section the complete multipole expansion of the retarded interatomic potential energy has been derived. It contains the matrix elements of the irreducible tensor operators for the atomic multipole moments that follow from the longitudinal and transversal part of the one-photon vertex function.

7. *Derivation from dispersion relation methods.* An alternative way to derive the interaction energy due to two-photon exchange makes use of dispersion-relation methods. For non-relativistic atoms the multipole expansion of the dispersion energy has been obtained along such lines in paper V of this series, on the basis of Feinberg and Sucher's derivation[8]) in which a generalized dipole approximation was employed. In this section both the interatomic dispersion and induction energy for a pair of relativistic hydrogen atoms will be found by considering the analytical behaviour of the Fourier transform F_2, given in (7), as a function of complex $Q^2 = -t$ (with t the momentum transfer).

As in paper V, appendix A, the dispersive part $F_{\text{disp}}(t)$ of $F_2(t)$ may be shown to be an analytic function in the complex t plane apart from a cut along the positive axis, a branchpoint being present at $t = 0$. The dispersion energy may then be written as:

$$V_{\text{disp}}(R) = -\frac{\mathrm{i}}{8\pi^2 R}\int_0^\infty \mathrm{d}t\, \mathrm{e}^{-\sqrt{t}R}\,[F_{\text{disp}}(t + \mathrm{i}0) - F_{\text{disp}}(t - \mathrm{i}0)]. \tag{58}$$

Here the discontinuity across the cut is given by

$$F_{\text{disp}}(t + \mathrm{i}0) - F_{\text{disp}}(t - \mathrm{i}0) = -\frac{\mathrm{i}}{8\pi}\,\frac{1}{(2J_a^0 + 1)(2J_b^0 + 1)}$$
$$\times \sum_{M_a^0, M_b^0} \int_0^{\frac{1}{2}\sqrt{t}} \mathrm{d}\varkappa_\perp \varkappa_\perp \tilde{\Gamma}_a^{+,\mu\nu}(k, k')\, g_{\mu\mu'} g_{\nu\nu'} \tilde{\Gamma}_b^{-,\mu'\nu'}(-k, -k') \tag{59}$$

[*v.* (V.14)–(V.17)], with $\tilde{\Gamma}_a^{\pm,\mu\nu}(k, k')$ equal to the dispersive part of $\Gamma_a^{\mu\nu}(k, k')$ for $k^0 = -k'^0$, $k^{02} = k^2 = k'^2$, $k^0 \gtrless 0$, corresponding to mass-shell photons. By inserting $\boldsymbol{k} = \frac{1}{2}\boldsymbol{Q} + \boldsymbol{\varkappa}_\perp$, $\boldsymbol{k}' = \frac{1}{2}\boldsymbol{Q} - \boldsymbol{\varkappa}_\perp$ (with $\boldsymbol{Q} \cdot \boldsymbol{\varkappa}_\perp = 0$) the product of vertex functions (averaged over the atomic ground states) will depend only on $\varkappa_\perp^2$ and $t < 0$; in the integrand of (59) the analytical continuation of this product towards positive t, with fixed $\varkappa_\perp^2$, is to be substituted.

In the same way as in section V.3 the combination of two-photon vertex functions occurring in (59) may be written as the product of two vertex functions, each contracted with a pair of polarization vectors $\varepsilon_\lambda^\mu(\boldsymbol{k}) = (0, \boldsymbol{\varepsilon}_\lambda(\boldsymbol{k}))$, with $\boldsymbol{\varepsilon}_\lambda(\boldsymbol{k}) \cdot \boldsymbol{k} = 0$ ($\lambda = \pm$). In fact, for mass-shell photons ($k^2 = 0$) the polarization vectors fulfil the identity:

$$g^{\mu\mu'} = -\sum_{\lambda=\pm} \varepsilon_\lambda^\mu(\boldsymbol{k})\,\varepsilon_\lambda^{\mu'}(\boldsymbol{k}) - (k \cdot \eta)^{-2} k^\mu k^{\mu'} + (k \cdot \eta)^{-1}(k^\mu \eta^{\mu'} + \eta^\mu k^{\mu'}), \tag{60}$$

with $\eta^\mu = (1, 0)$. The two-photon vertex function satisfies the orthogonality relation $k_\mu \Gamma^{\mu\nu}(k, k') = 0$, as follows from current conservation $k_\mu \Gamma^\mu(k) = (k^0 - k_N)\, \Gamma^0(k)$. Hence, after contraction of $\tilde{\Gamma}_a^{\mu\nu}$ and $\tilde{\Gamma}_b^{\mu'\nu'}$ with the right-hand side of (60) only the first term remains, so that, in order to evaluate (59), the contracted two-photon vertex function $\varepsilon \cdot \tilde{\boldsymbol{\Gamma}}(k, k') \cdot \varepsilon'$ is to be studied. It contains one-photon vertex functions for which one may derive the following multipole expansion:

$$(2J_a^0 + 1)^{-1} \sum_{M_a^0} \sum_{N_a(k_a \text{ const}),\, J_a M_a} \varepsilon \cdot \boldsymbol{\Gamma}_a(k)\, \boldsymbol{\Gamma}_a^*(-k') \cdot \varepsilon'$$

$$= \sum_{N_a(k_a \text{ const}),\, J_a} \sum_{n_a=0}^{\infty} \sum_{m_a=0}^{n_a} (-k^2)^{n_a - m_a} (\tfrac{1}{4}t)^{m_a} [k^2 \varepsilon \cdot \varepsilon' \varphi_E^{(N_a, J_a, n_a, m_a)}(k)$$

$$- (\varepsilon \wedge \boldsymbol{k}) \cdot (\varepsilon' \wedge \boldsymbol{k}')\, \varphi_M^{(N_a, J_a, n_a, m_a)}(k)], \tag{61}$$

for $k^2 = k'^2$ and $(\boldsymbol{k} + \boldsymbol{k}')^2 = -t$. Here φ_E and φ_M are defined in terms of the products (B13) and (B15) of the transverse electric and magnetic multipole matrix elements (B10) and (B11) as:

$$\varphi_i^{(N, J, n, m)}(k) = \sum_{L, S}^{(n)} \tfrac{1}{2} [L(L+1)]^{-1} \{[a_i^L]_m\, k^{-2} \mathcal{K}_{NJ}(L, S)$$

$$+ [b_i^L]_m \mathcal{N}_{NJ}(L, S)\}, \tag{62}$$

with (n) denoting the restriction $L + S = n + 1$. Furthermore a_i^L and b_i^L are linear combinations of derivatives of Legendre polynomials:

$$a_E^L = b_M^L = P''_{L+1} + P''_{L-1} - P'_L, \tag{63}$$

$$a_M^L = b_E^L = 2P''_L. \tag{64}$$

The bracket notation $[\ldots]_m$ is defined by:

$$f(\cos\theta) = \sum_m [f]_m (\tfrac{1}{2} + \tfrac{1}{2}\cos\theta)^m; \tag{65}$$

in particular one finds for the Legendre polynomials and their derivatives

$$[P_L^{(j)}]_m = \frac{(-1)^{L+m+j}(L+m+j)!}{2^j (L-m-j)!\,(m+j)!\,m!}. \tag{66}$$

Upon insertion of (61) into (9) the following expression for the contracted (mass-shell) two-photon vertex function is obtained:

$$(2J_a^0 + 1)^{-1} \sum_{M_a^0} \varepsilon \cdot \tilde{\boldsymbol{\Gamma}}_a(k, k') \cdot \varepsilon'$$

$$= k^2 \varepsilon \cdot \varepsilon' F_{aE}(k, t) - (\varepsilon \wedge \boldsymbol{k}) \cdot (\varepsilon' \wedge \boldsymbol{k}')\, F_{aM}(k, t) \tag{67}$$

with form factors:

$$F_i(k, t) = \sum_{N(\neq 0), J} \sum_{n=0}^{\infty} \sum_{m=0}^{n} (-k^2)^{n-m} (\tfrac{1}{4}t)^m \, 2k_N (k_N^2 - k^2 - \mathrm{i}0)^{-1} \varphi_i^{(N, J, n, m)}(k). \tag{68}$$

For soft photons ($k = 0$) the form factors reduce to the static polarizabilities:

$$F_{\mathrm{E}}(0) = \tfrac{2}{3} \sum_{N(\neq 0), J} k_N^{-1} |\mu_{NJ}(1, 0)|^2, \tag{69}$$

$$F_{\mathrm{M}}(0) = \tfrac{2}{3} \sum_{N(\neq 0), J} k_N^{-1} |\nu_{NJ}(1, 0)|^2, \tag{70}$$

as may be shown by employing the sum rule (B20), with (B12).

The dispersion energy may now be evaluated by substitution of the expression (67) for the contracted two-photon vertex function (after analytical continuation to positive t) in (58) with (59). In this way one may recover the results (54)–(56). In particular the long-range part of the dispersion energy is determined by the soft-photon form factors (69) and (70), as may be seen directly from (31).

In a recent paper[11]) Au and Feinberg extended the work by Feinberg and Sucher[8]) so as to include higher-order multipole contributions in the dispersion energy of two neutral spinless particles. The structure of their results, which contain the properties of the particles in terms of unspecified spectral functions, is different from that of the expressions given here in terms of the atomic multipole matrix elements. Indeed these authors assume integral representations for the electromagnetic form factors in closed form, which do not hold for each term separately in the multipole expansion (68) with (62) of the form factors.

The inductive part of the interaction energy is the Fourier transform of F_{ind}, occurring in (7). As a consequence of time-reversal invariance the time–time and space–space components of the two-photon vertex functions, of which one contains only ground-level intermediate states and the other excited intermediate states, may be written as:

$$\Gamma^{\mu\nu}_{\mathrm{a}(k_{\mathrm{a}}=0)}(k, k') = 2\pi\mathrm{i}\delta(k^0) \sum_{(N_{\mathrm{a}}=0), J_{\mathrm{a}}, M_{\mathrm{a}}} \Gamma^{\mu}_{\mathrm{a}}(k) \, \Gamma^{\nu *}_{\mathrm{a}}(-k') \tag{71}$$

and

$$\Gamma^{\mu\nu}_{\mathrm{b}(k_{\mathrm{b}}\neq 0)}(k, k') = 2 \sum_{N_{\mathrm{b}}(\neq 0), J_{\mathrm{b}}, M_{\mathrm{b}}} k_{\mathrm{b}}^{-1} \Gamma^{\mu}_{\mathrm{b}}(k) \, \Gamma^{\nu *}_{\mathrm{b}}(-k'), \tag{72}$$

respectively (in the latter k^0 has been put equal to zero); the mixed time–space components drop out. The analytical properties of the function $F_{\mathrm{Ind}}(t)$, with $t = -Q^2$, are similar to those of $F_{\mathrm{disp}}(t)$, so that V_{ind} may be evaluated from a formula analogous to (58). The discontinuity across the cut along the positive

t axis is in the present case given by:

$$F_{\text{ind}}(t + \text{i}0) - F_{\text{ind}}(t - \text{i}0)$$
$$= -\tfrac{1}{16}\pi^{-1}\, t^{-\frac{1}{2}}\,(2J_a^0 + 1)^{-1}\,(2J_b^0 + 1)^{-1} \sum_{M_a^0, M_b^0} \int \mathrm{d}k^0\, \hat{\Gamma}^{\mu\nu}_{a(k_a=0)}(k, k')$$
$$\times\, g_{\mu\mu'} g_{\nu\nu'}\, \hat{\Gamma}^{\mu'\nu'}_{b(k_b\neq 0)}(-k, -k') + (a \leftrightarrow b), \tag{73}$$

(*v.* appendix C) where $\hat{\Gamma}^{\mu\nu}$ is defined as $\Gamma^{\mu\nu}$ with $k^0 = -k'^0$, $k^2 = k'^2$ inserted. By writing again $\boldsymbol{k} = \tfrac{1}{2}\boldsymbol{Q} + \boldsymbol{\kappa}_\perp$, $\boldsymbol{k}' = \tfrac{1}{2}\boldsymbol{Q} - \boldsymbol{\kappa}_\perp$ the product of vertex functions will depend on k^0, $\varkappa_\perp^2$ and t. It has to be continued to positive values of t, whereupon $\varkappa_\perp^2$ is put equal to $\frac{1}{4}t$, before insertion into (73).

As was the case for the dispersion energy the metric tensors contracted with the two-photon vertex functions in (73) may now be replaced by expressions containing polarization vectors ε_λ^μ (with $\lambda = \pm$) for photons which in the present case are off the mass-shell ($k^2 \neq 0$), but have vanishing time components ($k^0 = 0$):

$$g^{\mu\mu'} = -\sum_{\lambda=\pm} \varepsilon_\lambda^\mu(k)\, \varepsilon_\lambda^{\mu'}(k) + \eta^\mu \eta^{\mu'} + k^{-2} k^\mu k^{\mu'}. \tag{74}$$

Upon substitution into (73) the term $k^{-2}k^\mu k^{\mu'}$ drops out due to the orthogonality relation $k_\mu \Gamma^{\mu\nu}(k, k') = 0$. The term arising from contraction of the two-photon vertex functions with $\varepsilon^\mu \varepsilon^{\mu'}$ may be expressed in multipoles with the help of the expansion (61). Similarly the expansion

$$(2J_a^0 + 1)^{-1} \sum_{M_a^0} \sum_{N_a(k_a\,\text{const}),\, J_a,\, M_a} \Gamma_a^0(k)\, \Gamma_a^{0*}(-k')$$
$$= \sum_{N_a(k_a\text{const}),\, J_a} \sum_{n_b=0}^{\infty} \sum_{m_b=0}^{n_a+1} (-k^2)^{n_a - m_a + 1}\, (\tfrac{1}{4}t)^{m_a}\, \psi^{(N_a, J_a, n_a, m_a)} \tag{75}$$

[*v.* (B2)] may be used to rewrite the contractions with $\eta^\mu \eta^{\mu'}$ in terms of multipole matrix elements. Here ψ contains longitudinal electric multipole matrix elements (B1) only:

$$\psi^{(N, J, n, m)} = \sum_{L, S}^{(n)} L\,(L + 1)^{-1}\, [P_L]_m\, \mathscr{M}_{NJ}(L, S) \tag{76}$$

with (n) standing for the condition $L + S = n + 1$.

Upon substituting the multipole expansions for $\varepsilon \cdot \tilde{\Gamma} \cdot \varepsilon'$ and $\hat{\Gamma}^{00}$ into (73) the resulting integrand, which is a function of k^0, $\varkappa_\perp^2 = \frac{1}{4}t + k^2$ and t, has to be continued analytically towards positive t; then $\varkappa_\perp^2$ is to be put equal to $\frac{1}{4}t$, so that k^2 becomes zero. As a result only the multipoles with $s = 0$ in (62) and (76) contribute; the transverse electric multipoles contained in $\mathscr{M}_{NJ}(L, 0)$ may be shown to drop out altogether, due to (B12) and (B20). The resulting multipole series for (73) leads upon evaluation of the dispersion integral to the induction energy (57).

Both the dispersive and inductive part of the interatomic potential energy have now been derived from dispersion-relation methods, with results identical to those obtained in the preceding sections by evaluation of the Feynman diagrams. In the following section special cases of the general formulae will be studied.

8. *The lowest-order multipole contributions to the dispersion energy.* In this section the formulae (54)–(56) for the complete multipole expansion of the interatomic dispersion energy will be used to evaluate the explicit expressions for electric and magnetic dipole and quadrupole interactions. To that end the values $s_{ai} = 0$, $s_{bi} = 0$, $L_a = 1, 2$ and $L_b = 1, 2$ are to be inserted in the multipole matrix elements, leading to the lowest-order static multipole matrix elements $\mu_{NJ}(L, 0)$ and $\nu_{NJ}(L, 0)$. The radial differentiations in (54)–(56) may be carried out with the help of (19) and (50). Substituting moreover the numerical values for the $3j$-symbols[12]) one gets for the dipole dispersion energy:

$$
\begin{aligned}
V_{\text{disp, d-d}}(R) = \sum_{\substack{N_a(\neq 0), J_a \\ N_b(\neq 0), J_b}} & \frac{k_a k_b}{36\pi^3 (k_a^2 - k_b^2) R^5} \{[|\mu_a(1)|^2 |\mu_b(1)|^2 + (\mu \leftrightarrow \nu)] \\
& \times [(\varrho_a^3 - 5\varrho_a + 3\varrho_a^{-1}) P(2\varrho_a) + (-2\varrho_a^2 + 6) Q(2\varrho_a) - \tfrac{1}{2}\varrho_a^2] \\
& + [|\mu_a(1)|^2 |\nu_b(1)|^2 + (\mu \leftrightarrow \nu)] [(-\varrho_a^3 + \varrho_a) P(2\varrho_a) \\
& + 2\varrho_a^2 Q(2\varrho_a)]\} + (a \leftrightarrow b), \qquad (77)
\end{aligned}
$$

where the notation $\varrho_a = |k_a| R$ has been introduced. Furthermore the parameters N, J and $s(=0)$ in the matrix elements have been suppressed for brevity.

Similarly the mixed dipole–quadrupole contribution to the dispersion energy reads:

$$
\begin{aligned}
V_{\text{disp, d-q}}(R) = \sum_{\substack{N_a(\neq 0), J_a \\ N_b(\neq 0), J_b}} & \frac{k_a k_b}{80\pi^3 (k_a^2 - k_b^2) R^7} \{[|\mu_a(1)|^2 |\mu_b(2)|^2 \\
& + |\mu_a(2)|^2 |\mu_b(1)|^2 + (\mu \leftrightarrow \nu)] [(-\varrho_a^5 + 27\varrho_a^3 - 162\varrho_a \\
& + 90\varrho_a^{-1}) P(2\varrho_a) + (6\varrho_a^4 - 84\varrho_a^2 + 180) Q(2\varrho_a) \\
& + \tfrac{1}{2}\varrho_a^4 - \tfrac{61}{4}\varrho_a^2] + [|\mu_a(1)|^2 |\nu_b(1)|^2 + |\mu_a(2)|^2 |\nu_b(1)|^2 \\
& + (\mu \leftrightarrow \nu)] [(\varrho_a^5 - 15\varrho_a^3 + 9\varrho_a) P(2\varrho_a) \\
& + (-6\varrho_a^4 + 18\varrho_a^2) Q(2\varrho_a) - \tfrac{1}{2}\varrho_a^4 + \tfrac{37}{4}\varrho_a^2]\} + (a \leftrightarrow b). \qquad (78)
\end{aligned}
$$

Finally the quadrupole–quadrupole part of the dispersion energy is found as:

$$V_{\text{disp, q–q}}(R) = \sum_{\substack{N_a(\neq 0), J_a \\ N_b(\neq 0), J_b}} \frac{9k_a k_b}{1600\pi^3 (k_a^2 - k_b^2) R^9} \{[|\mu_a(2)|^2 |\mu_b(2)|^2 + (\mu \leftrightarrow \nu)]$$

$$\times [(\varrho_a^7 - 89\varrho_a^5 + 1983\varrho_a^3 - 9360\varrho_a + 5040\varrho_a^{-1}) P(2\varrho_a)$$

$$+ (-10\varrho_a^6 + 510\varrho_a^4 - 5280\varrho_a^2 + 10080) Q(2\varrho_a) - \tfrac{1}{2}\varrho_a^6$$

$$+ \tfrac{189}{4}\varrho_a^4 - \tfrac{4583}{4}\varrho_a^2] + [|\mu_a(2)|^2 |\nu_b(2)|^2 + (\mu \leftrightarrow \nu)]$$

$$\times [(-\varrho_a^7 + 53\varrho_a^5 - 324\varrho_a^3 + 180\varrho_a) P(2\varrho_a) + (10\varrho_a^6 - 168\varrho_a^4$$

$$+ 360\varrho_a^2) Q(2\varrho_a) + \tfrac{1}{2}\varrho_a^6 - \tfrac{117}{4}\varrho_a^4 + \tfrac{887}{4}\varrho_a^2]\} + (\text{a} \leftrightarrow \text{b}). \quad (79)$$

An alternative form for the dipole and quadrupole dispersion energies may be obtained by employing for $P(x)$ and $Q(x)$ the integral representations:

$$P(x) = \int_0^\infty \mathrm{d}t \frac{e^{-xt}}{1 + t^2}, \qquad Q(x) = \int_0^\infty \mathrm{d}t \frac{te^{-xt}}{1 + t^2}; \quad (80)$$

in this form the non-relativistic dipole approximation was given in refs. 5 and 7.

The long-range expressions for $V_{\text{disp}}(R)$ up to quadrupoles follow from (77)–(79) by inserting the asymptotic expansions of $P(x)$ and $Q(x)$, or, more directly, by putting $P(x) = x^{-1}$ in the general formulae (54)–(56). For $V_{\text{disp, d–d}}$ this leads to (31) (with cartesian dipole matrix elements replaced by the corresponding spherical ones); the asymptotic expressions for the higher-order multipoles are:

$$V^{\text{L}}_{\text{disp, d–q}}(R) = -\sum_{\substack{N_a(\neq 0), J_a \\ N_b(\neq 0), J_b}} \frac{1}{640\pi^3 k_a k_b R^9}$$

$$\times \{1593 [|\mu_a(1)|^2 |\mu_b(2)|^2 + |\mu_a(2)|^2 |\mu_b(1)|^2 + (\mu \leftrightarrow \nu)]$$

$$- 297 [|\mu_a(1)|^2 |\nu_b(2)|^2 + |\mu_a(2)|^2 |\nu_b(1)|^2 + (\mu \leftrightarrow \nu)]\}, \quad (81)$$

$$V^{\text{L}}_{\text{disp, q–q}}(R) = -\sum_{\substack{N_a(\neq 0), J_a \\ N_b(\neq 0), J_b}} \frac{1}{6400\pi^3 k_a k_b R^{11}} \{5591 [|\mu_a(2)|^2 |\mu_b(2)|^2 + (\mu \leftrightarrow \nu)]$$

$$- 47223 [|\mu_a(2)|^2 |\nu_b(2)|^2 + (\mu \leftrightarrow \nu)]\}. \quad (82)$$

From the formulae (77)–(79) the non-relativistic limit for the dipole and quadrupole dispersion energies may now be derived (*v.* appendix B). In the sums over

positive-energy intermediate states the multipole matrix elements μ and ν are then to be replaced by their non-relativistic counterparts, while the negative-energy intermediate states give rise [*v.* (B26), (B29)] to direct matrix elements $M(L + s_1 + s_2)$ of the form (B24). The direct matrix element which originates in this way from the magnetic dipole moments is in fact the diamagnetic matrix element $M(1)$. Thus the long-range dipole dispersion energy (31), (which is in fact the complete asymptotic dispersion energy), leads in the non-relativistic limit to an interaction energy containing, apart from electric and magnetic dipole contributions, terms with these diamagnetic matrix elements. The latter may be transformed with the help of (B30) into a sum, over positive-energy intermediate states, of squared quadrupole matrix elements. In such a form, with dipoles and electric quadrupoles, the non-relativistic limit of the asymptotic dispersion energy has been given in paper III, *viz.* in (III.1), (III.15) and (III.18) (in that paper spin terms were not considered; then in addition the orbital magnetic-dipole matrix elements may be shown to vanish, as a consequence of rotation invariance arguments). In the same way the non-relativistic limit of the general expression (77) for $V_{\text{disp, d-d}}$ may be rewritten as the sum of the terms in (III.19), (III.23) and (III.25) due to V_{I}, V_{II} and V_{III}, respectively (in the notation of papers I–III). The remaining terms in (III.23) and (III.25) follow from the non-relativistic limit of the $\mu(1)$–$\mu(2)$, $\nu(1)$–$\mu(2)$ and $\mu(2)$–$\mu(2)$ interactions in (78) and (79).

APPENDIX A

Cartesian multipole matrix elements and sum rules. The operator occurring in the one-photon vertex function (8), *viz.*

$$\Gamma^{\mu} = -e\,(\gamma^0\gamma^{\mu}\,\mathrm{e}^{-\boldsymbol{r}\cdot\nabla} - g^{0\mu}) \tag{A1}$$

(with ∇ instead of $i\boldsymbol{k}$) may be expanded into cartesian multipole operators:

$$\boldsymbol{\mu}^{(n)} = -e\,(1/n!)\,\boldsymbol{r}^n, \tag{A2}$$

$$\boldsymbol{\nu}^{(n)} = -e\,[n/(n+1)!]\,\boldsymbol{r}^{n-1}\boldsymbol{r} \wedge \gamma^0\boldsymbol{\gamma}. \tag{A3}$$

In fact these expansions are:

$$\Gamma^0 = \sum_{n=1}^{\infty} (-\nabla)^n \vdots\, \boldsymbol{\mu}^{(n)}, \tag{A4}$$

$$\boldsymbol{\Gamma} = \sum_{n=1}^{\infty} (-\nabla)^{n-1} \vdots\, (\mathrm{i}\,[H, \boldsymbol{\mu}^{(n)}] - \boldsymbol{\nu}^{(n)} \wedge \nabla), \tag{A5}$$

from which the current conservation relation (24) is obvious.

Time-reversal invariance of the atomic hamiltonian leads [*v.* formulae (10)–(11)] for the matrix elements of the operators (A2) and (A3) between states $|\alpha_0\rangle$ and $|\alpha\rangle$ to the following properties with respect to complex conjugation:

$$\mathscr{I}m\, \mathsf{S}_\alpha\, \boldsymbol{\mu}_\alpha^{(n_1)} \boldsymbol{\mu}_\alpha^{(n_2)*} = 0, \tag{A6}$$

$$\mathscr{R}e\, \mathsf{S}_\alpha\, \boldsymbol{\mu}_\alpha^{(n_1)} \mathbf{v}_\alpha^{(n_2)*} = 0, \tag{A7}$$

$$\mathscr{I}m\, \mathsf{S}_\alpha\, \mathbf{v}_\alpha^{(n_1)} \mathbf{v}_\alpha^{(n_2)*} = 0, \tag{A8}$$

where the abbreviation $\mathsf{S}_\alpha = \sum_{\alpha_0} \sum_{\alpha(k_\alpha \text{ const})}$ has been introduced. The corresponding property (22), for the one-photon vertex function (A1), may be obtained now by employing (A4) and (A5). Space-inversion invariance of the atomic hamiltonian implies the parity selection rules

$$\mathsf{S}_\alpha\, \boldsymbol{\mu}_\alpha^{(n_1)} \boldsymbol{\mu}_\alpha^{(n_2)*} = 0 \qquad (n_1 - n_2 \text{ odd}), \tag{A9}$$

$$\mathsf{S}_\alpha\, \boldsymbol{\mu}_\alpha^{(n_1)} \mathbf{v}_\alpha^{(n_2)*} = 0 \qquad (n_1 - n_2 \text{ even}), \tag{A10}$$

$$\mathsf{S}_\alpha\, \mathbf{v}_\alpha^{(n_1)} \mathbf{v}_\alpha^{(n_2)*} = 0 \qquad (n_1 - n_2 \text{ odd}), \tag{A11}$$

while rotational invariance entails moreover the relation:

$$\mathsf{S}_\alpha\, \boldsymbol{\mu}_\alpha^{(n_1)} \mathbf{v}_\alpha^{(1)*} = 0 \qquad (n_1 \text{ even}). \tag{A12}$$

From the vanishing of the repeated commutator $[[H, \boldsymbol{r}^{n_1}], \boldsymbol{r}^{n_2}]$ one gets, with the help of (A6), the sum rule:

$$\mathsf{S}\, k_\alpha \boldsymbol{\mu}_\alpha^{(n_1)} \boldsymbol{\mu}_\alpha^{(n_2)*} = 0, \tag{A13}$$

with $\mathsf{S} = \sum_{k_\alpha(\neq 0)} \mathsf{S}_\alpha$. Likewise a second sum rule is proved, *viz.*:

$$\mathsf{S}\, \boldsymbol{\mu}_\alpha^{(n_1)} \mathbf{v}_\alpha^{(n_2)*} = 0. \tag{A14}$$

In view of (A4) and (A5), the ensuing sum rules for the one-photon vertex functions read:

$$\mathsf{S}\, k_\alpha \Gamma_{\alpha 1}^0 \Gamma_{\alpha 2}^{0*} = 0, \tag{A15}$$

$$\mathsf{S}\, \Gamma_{\alpha 1}^0 \boldsymbol{\Gamma}_{\alpha 2}^* = 0, \tag{A16}$$

$$\mathsf{S}\, k_\alpha^{-1} \boldsymbol{\Gamma}_{\alpha 1} \boldsymbol{\Gamma}_{\alpha 2}^* = \mathsf{S}\, k_\alpha^{-1} \sum_{n_1, n_2 = 1}^{\infty} (-\boldsymbol{\nabla}_1)^{n_1 - 1} : \mathbf{v}_\alpha^{(n_1)} \wedge \boldsymbol{\nabla}_1 (-\boldsymbol{\nabla}_2)^{n_2 - 1} : \mathbf{v}_\alpha^{(n_2)*} \wedge \boldsymbol{\nabla}_2 . \tag{A17}$$

These sum rules are not independent; in fact, current conservation (24) may be used to derive (A15) and (A16) from (A17).

In the non-relativistic limit the time and space components of the one-photon vertex function for positive energy states α_+ reduce to:

$$\bar{\Gamma}^0_{\alpha_+ i} = -e \langle \alpha_0 | \, \mathrm{e}^{-r \cdot \nabla_i} - 1 \, | \alpha_+ \rangle, \tag{A18}$$

$$\bar{\boldsymbol{\Gamma}}_{\alpha_+ i} = -(e/m) \langle \alpha_0 | \, \tfrac{1}{2} \{ \boldsymbol{p} - \tfrac{1}{2} \boldsymbol{\sigma} \wedge \nabla_i, \mathrm{e}^{-r \cdot \nabla_i} \} \, | \alpha_+ \rangle, \tag{A19}$$

respectively. Multipole expansions of the same form as (A4) and (A5) may be found if the non-relativistic multipole moments are defined as:

$$\bar{\boldsymbol{\mu}}^{(n)}_{\alpha_+} = -e \langle \alpha_0 | \, (1/n!) \, \boldsymbol{r}^n \, | \alpha_+ \rangle, \tag{A20}$$

$$\bar{\mathbf{v}}^{(n)}_{\alpha_+} = -(e/m) \langle \alpha_0 | \, \tfrac{1}{2} \{ \boldsymbol{r}^{n-1}, [n/(n+1)!] \, \boldsymbol{r} \wedge \boldsymbol{p} + [1/2 \, (n-1)!] \, \boldsymbol{\sigma} \} \, | \alpha_+ \rangle. \tag{A21}$$

The non-relativistic magnetic-multipole matrix element $\bar{\mathbf{v}}^{(n)}$ differs from the non-relativistic limit of the corresponding relativistic matrix element (A2) by terms that drop out in the multipole expansion for (A19).

The sum rule (A17) and hence (A15) and (A16) lead in the non-relativistic limit to the sum rules given in paper I when, moreover, the spin terms are suppressed. In fact, in (A17) the negative-energy intermediate states give rise, according to (43), to a direct matrix element in the left-hand side, and in the right-hand side to the matrix element (I.B6). In this way (A17) may be seen to entail the non-relativistic sum rule (I.B9), which in turn implies (I.53), as follows when the inner product with ∇_i is taken.

APPENDIX B

Multipole matrix elements and sum rules in the spherical-tensor formalism. In this appendix the spherical-multipole expansion of the one-photon vertex function is derived. The resulting multipoles and their sum rules will be discussed both relativistically and in the non-relativistic limit.

The time component $\Gamma^0(\boldsymbol{k})$ of the operator in the one-photon vertex function (8) may be expressed, with the help of the Rayleigh expansion, in terms of the electric multipoles

$$\mu^M_L(s) = -e \, \frac{[4\pi \, (2L+1)]^{\frac{1}{2}}}{(2s)!! \, (2L+2s+1)!!} \, r^{L+2s} Y^M_L(\hat{\boldsymbol{r}}), \tag{B1}$$

with parity eigenvalues $(-1)^L$; in fact one has:

$$\Gamma^0(\boldsymbol{k}) = \sum_{L, M, s (L+2s>0)} (-1)^{L-M} \, [4\pi/(2L+1)]^{\frac{1}{2}} \, (\mathrm{i}k)^{L+2s} \, Y^{-M}_L(\hat{\boldsymbol{k}}) \, \mu^M_L(s). \tag{B2}$$

The spherical components $\Gamma^\lambda(\boldsymbol{k})$ ($\lambda = 0, \pm 1$) of the space part of the operator in (8) may be expressed in terms of the operators $\Omega_L^M(L', s)$, defined in (46), as:

$$\Gamma^\lambda(\boldsymbol{k}) = \sum_{\substack{L, L', s \\ M, M'}} (-1)^{L-M+L'-M'} [4\pi(2L+1)(2L'+1)]^{\frac{1}{2}} \begin{pmatrix} 1 & L & L' \\ \lambda & -M & -M' \end{pmatrix} \times (\mathrm{i}k)^{L'+2s} Y_{L'}^{M'}(\hat{\boldsymbol{k}}) \Omega_L^M(L', s). \tag{B3}$$

By applying the addition theorem for spherical harmonics

$$\prod_{i=1}^{n} [4\pi/(2L_i+1)]^{\frac{1}{2}} Y_{L_i}^{M_i}(\hat{\boldsymbol{k}}) = \sum_{L, M} (-1)^M [4\pi(2L+1)]^{\frac{1}{2}} \begin{pmatrix} L_1 \cdots L_n & L \\ M_1 \cdots M_n & -M \end{pmatrix} Y_L^M(\hat{\boldsymbol{k}}) \tag{B4}$$

[that follows from the definition (II.17) for the Gaunt coefficient], the longitudinal part $\boldsymbol{\Gamma}_\parallel = \boldsymbol{k}\boldsymbol{k} \cdot \boldsymbol{\Gamma}/k^2$ of (B3) may be cast into the form:

$$\Gamma_\parallel^\lambda(\boldsymbol{k}) = \sum_{\substack{L, L', n \\ M, M'}} (-1)^{L-M+L'-M'} (\mathrm{i}k)^n \begin{pmatrix} 1 & L & L' \\ \lambda & -M & -M' \end{pmatrix} T_{\parallel, L}^M(L', n) Y_{L'}^{M'}(\hat{\boldsymbol{k}}). \tag{B5}$$

Here the tensor operator $T_\parallel$ is defined as:

$$T_{\parallel, L}^M(L', n) = \sum_{L''}{}' [4\pi(2L+1)(2L'+1)]^{\frac{1}{2}} (2L''+1) \begin{pmatrix} 1 & L & L' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} 1 & L & L'' \\ 0 & 0 & 0 \end{pmatrix} \times \Omega_L^M\left(L'', \frac{n-L''}{2}\right) \tag{B6}$$

(the prime at the summation sign stands for the restriction $n - L''$ even and non-negative); it is non-vanishing only for $L' = L \pm 1$ and has the parity eigenvalue $(-1)^{L'+1} = (-1)^L$. Since $\boldsymbol{\Gamma}_\parallel$ may be written as $-[H, \boldsymbol{\Gamma}^0]\,\boldsymbol{k}/k^2$, the right-hand sides of (B2) and (B5) are closely related. As a matter of fact the gradient formula yields:

$$\mathrm{i}\,[H, \mu_L^M(s)] = \sum_{L''} (-1)^L (2L+1)^{\frac{1}{2}} (2L''+1) \begin{pmatrix} 1 & L & L'' \\ 0 & 0 & 0 \end{pmatrix} \times \Omega_L^M\left(L'', s + \frac{L-L''-1}{2}\right), \tag{B7}$$

which is precisely the combination of operators Ω occurring in (B5) with (B6).

The transversal part $\boldsymbol{\Gamma}_\perp = \boldsymbol{\Gamma} - \boldsymbol{\Gamma}_\parallel$ follows by subtracting (B5), with (B6), from (B3). Then an expression analogous to (B5) results for $\boldsymbol{\Gamma}_\perp$, with $T_\perp$ given

by:

$$T^M_{\perp,L}(L',n) = \sum_{L''}{}' [4\pi(2L+1)(2L'+1)]^{\frac{1}{2}} \times \left[\delta_{L'L''} - (2L''+1)\begin{pmatrix}1 & L & L'\\ 0 & 0 & 0\end{pmatrix}\begin{pmatrix}1 & L & L''\\ 0 & 0 & 0\end{pmatrix}\right] \Omega^M_L\left(L'', \frac{n-L''}{2}\right). \tag{B8}$$

With the help of the orthogonality relation of the $3j$-symbols an alternative form for $T_\perp$ is obtained:

$$T^M_{\perp,L}(L',n) = \sum_{L''}{}' [4\pi(2L+1)(2L'+1)]^{\frac{1}{2}}(2L''+1) \times \sum_{\lambda=\pm 1}\begin{pmatrix}1 & L & L'\\ \lambda & -\lambda & 0\end{pmatrix}\begin{pmatrix}1 & L & L''\\ \lambda & -\lambda & 0\end{pmatrix}\Omega^M_L\left(L'', \frac{n-L''}{2}\right). \tag{B9}$$

Here the cases $L = L' \pm 1$ and $L = L'$ may be considered separately. In the former case (B9) contains the following combination of operators Ω:

$$\varkappa^M_L(s) = \sum_{L''=L\pm 1}(-1)^{L+1}\left[\frac{2L(2L+1)}{L+1}\right]^{\frac{1}{2}}(2L''+1)\begin{pmatrix}1 & L & L''\\ 1 & -1 & 0\end{pmatrix} \times \Omega^M_L\left(L'', s + \frac{L-L''-1}{2}\right), \tag{B10}$$

with parity eigenvalue $(-1)^L$; in the latter case one is led to the operator:

$$\nu^M_L(s) = \mathrm{i}(-1)^{L+1}\left[\frac{2L(2L+1)}{L+1}\right]^{\frac{1}{2}}(2L+1)\begin{pmatrix}1 & L & L\\ 1 & -1 & 0\end{pmatrix}\Omega^M_L(L,s), \tag{B11}$$

with parity eigenvalue $(-1)^{L+1}$. These combinations of operators also occur in the theory of multipole radiation from atomic systems. In fact the amplitudes for the electric and magnetic 2^L-pole radiation may be expressed in terms of (B10) and (B11), respectively. For that reason the operators $\varkappa^M_L(s)$ and $\nu^M_L(s)$ may be called the transversal electric and magnetic multipole operators. From (B10) it follows that $\varkappa^M_L(0)$ is related to the electrostatic multipole moment $\mu^M_L(0)$:

$$\varkappa^M_L(0) = \mathrm{i}\,[H, \mu^M_L(0)]. \tag{B12}$$

The two-photon vertex functions (9) lead, upon evaluation in the spherical-tensor formalism, to the following products of reduced matrix elements of the multipole moment operators (B1), (B10) and (B11):

$$\mathscr{K}_{NJ}(L,S) = \frac{L+1}{L(2L+1)}\sum_s \varkappa_{NJ}(L,s)\,\varkappa^*_{NJ}(L,S-s), \tag{B13}$$

$$\mathcal{M}_{NJ}(L, S) = \frac{L+1}{L(2L+1)} \sum_s \mu_{NJ}(L, s)\, \mu^*_{NJ}(L, S-s), \tag{B14}$$

$$\mathcal{N}_{NJ}(L, S) = \frac{L+1}{L(2L+1)} \sum_s \nu_{NJ}(L, s)\, \nu^*_{NJ}(L, S-s). \tag{B15}$$

[The dipole matrix elements are obtained from (B1), (B10) and (B11) by putting $L = 1, s = 0$:

$$\varkappa_1^M(0) = -e\alpha^M, \tag{B16}$$

$$\mu_1^M(0) = -er^M, \tag{B17}$$

$$\nu_1^M(0) = -\tfrac{1}{2}e\,(\boldsymbol{r} \wedge \alpha)^M. \tag{B18}$$

Sum rules for the reduced matrix elements of the spherical multipole operators may be derived from those for the one-photon vertex functions, given in appendix A (with $-i\nabla_1$ and $-i\nabla_2$ replaced by $\boldsymbol{k}$ and $\boldsymbol{k}'$, respectively). Inserting (B2) and (B5) into (A16) and using the orthogonality of the $3j$-symbols one gets:

$$\sum_{N(\neq 0), J} \langle 0, J^0 || \mu_L(s_1) || N, J\rangle \langle N, J || T_{\parallel, L}(L_2', n_2) || 0, J^0\rangle = 0, \tag{B19}$$

which leads with (B6) and (B7) to the sum rule:

$$\sum_{N(\neq 0), J} k_N \mu_{NJ}(L, s_1)\, \mu^*_{NJ}(L, s_2) = 0. \tag{B20}$$

A formula similar to (B19) is found for $T_\perp$, giving the sum rule:

$$\sum_{N(\neq 0), J} \mu_{NJ}(L, s_1)\, \varkappa^*_{NJ}(L, s_2) = 0. \tag{B21}$$

The analogous sum rule with $\varkappa$ replaced by ν is trivial since then all terms vanish separately, as may be seen from parity considerations.

The non-relativistic approximation of the operator (46) has the form:

$$\begin{aligned}\bar{\Omega}_L^M(L', s) = -e \sum_{\lambda, M'} (-1)^{1-\lambda+L'-M'} (2L+1)^{\frac{1}{2}} \begin{pmatrix} L' & 1 & L \\ -M' & -\lambda & M \end{pmatrix} \\ \times [(2s)!!\,(2L'+2s+1)!!]^{-1}\, [4\pi/(2L'+1)]^{\frac{1}{2}} \\ \times (\tfrac{1}{2}\{m^{-1}p^\lambda + \alpha^\lambda, r^{L'+2s} Y_{L'}^{M'}(\hat{r})\} \\ + \tfrac{1}{2}i\,[m^{-1}(\sigma \wedge p)^\lambda, r^{L'+2s} Y_{L'}^{M'}(\hat{r})])\end{aligned} \tag{B22}$$

[*v.* (39) and (40)], from which expressions for non-relativistic operators $\bar{\varkappa}$, $\bar{\mu}$ and $\bar{\nu}$ follow by relations analogous to (B7), (B10) and (B11). Sums over negative-energy intermediate states containing products of the reduced matrix element $\bar{\Omega}_{NJ}(L,L',s)$ [*v.* (47)] may be rewritten as:

$$\sum_{N(k_N<0),J} f(k_N)\,\bar{\Omega}_{NJ}(L,L_1,s_1)\,\bar{\Omega}^*_{NJ}(L,L_2,s_2)$$
$$= f(-2m)\,(2L+1)\,(2L_1+1)^{-1}\,\delta_{L_1,L_2}\,c(L_1,s_1,s_2)\,M(L+s_1+s_2), \tag{B23}$$

where the direct matrix element

$$M(L+s_1+s_2) = e^2\,(2J^0+1)^{-\frac{1}{2}}\,\langle 0,J^0||r^{2L+2s_1+2s_2}||0,J^0\rangle \tag{B24}$$

and the coefficient

$$c(L,s_1,s_2) = [(2s_1)!!\,(2s_2)!!\,(2L+2s_1+1)!!\,(2L+2s_2+1)!!]^{-1} \tag{B25}$$

have been introduced. The relation (B23) leads, with the use of (B7), (B10) and (B11) to analogous formulae for the sums of products of multipole matrix elements:

$$\sum_{N(k_N<0),J} f(k_N)\,\bar{\mu}_{NJ}(L,s_1)\,\bar{\mu}^*_{NJ}(L,s_2)$$
$$= \tfrac{1}{4}m^{-2}\,f(-2m)\,(2L+1)\,[Lc(L-1,s_1,s_2) + (L+1)$$
$$\times\, c(L+1,s_1-1,s_2-1)]\,M(L+s_1+s_2-1), \tag{B26}$$

$$\sum_{N(k_N<0),J} f(k_N)\,\bar{\mu}_{NJ}(L,s_1)\,\bar{\varkappa}^*_{NJ}(L,s_2)$$
$$= -\tfrac{1}{2}im^{-1}\,f(-2m)\,L\,(2L+1)\,[c(L-1,s_1,s_2) - c(L+1,s_1-1,s_2-1)]$$
$$\times\, M(L+s_1+s_2-1), \tag{B27}$$

$$\sum_{N(k_N<0),J} f(k_N)\,\bar{\varkappa}_{NJ}(L,s_1)\,\bar{\varkappa}^*_{NJ}(L,s_2)$$
$$= f(-2m)\,L\,(2L+1)\,(L+1)^{-1}\,[(L+1)\,c(L-1,s_1,s_2)$$
$$+ Lc(L+1,s_1-1,s_2-1)]\,M(L+s_1+s_2-1), \tag{B28}$$

$$\sum_{N(k_N<0),J} f(k_N)\,\bar{\nu}_{NJ}(L,s_1)\,\bar{\nu}^*_{NJ}(L,s_2)$$
$$= f(-2m)\,L\,(2L+1)^2\,(L+1)^{-1}\,c(L,s_1,s_2)\,M(L+s_1+s_2). \tag{B29}$$

Parity conservation implies the vanishing of similar expressions containing products of an electric and a magnetic multipole matrix element. From (B26) and (B27) the non-relativistic limits of the sum rules (B20) and (B21) follow immediately. The sum rules may then be used to rewrite the direct matrix element M as a sum, over positive-energy intermediate states, of products of non-relativistic electric-multipole matrix elements; in this way one gets:

$$M(L + s_1 + s_2)$$
$$= 2m(2L+3)^{-1}\,[(L+1)\,c(L, s_1, s_2) + (L+2)\,c(L+2, s_1-1, s_2-1)]^{-1}$$
$$\times \sum_{N(k_N>0),\,J} k_N \bar{\mu}_{NJ}(L+1, s_1)\,\bar{\mu}^*_{NJ}(L+1, s_2), \tag{B30}$$

and an analogous expression for M in terms of products of $\bar{\mu}$ and $\bar{\varkappa}$.

APPENDIX C

The analytical behaviour of the Fourier-transformed induction energy. The inductive part F_{ind} of F_2 given in (7) contains a contracted product of averaged two-photon vertex functions which has the form [*v.* (71) and (72)]

$$\frac{1}{(2J_a^0+1)(2J_b^0+1)} \sum_{\substack{(N_a=0),\,J_a,\,M_a,\,M_a^0\\ N_b(\neq 0),\,J_b,\,M_b,\,M_b^0}} \Gamma_{a,\mu\nu}(k, k')\,\Gamma_b^{\mu\nu}(-k, -k')$$
$$= \delta(k^0)\,f_{ab}(k^2, k'^2, \boldsymbol{k}\cdot\boldsymbol{k}'), \tag{C1}$$

with a regular function f_{ab} that is symmetric under an interchange of $\boldsymbol{k}$ and $\boldsymbol{k}'$. Introducing the variable $\boldsymbol{\kappa}$, by writing $\boldsymbol{k} = \frac{1}{2}\boldsymbol{Q} + \boldsymbol{\kappa}$, $\boldsymbol{k}' = \frac{1}{2}\boldsymbol{Q} - \boldsymbol{\kappa}$, the Fourier transform $F_{\text{ind}}(t)$ becomes (with $t = -Q^2$):

$$F_{\text{ind}}(t) = \frac{\mathrm{i}}{2(2\pi)^3} \int \mathrm{d}\varkappa_{\parallel}\,\mathrm{d}\varkappa_{\perp}\varkappa_{\perp} \frac{f_{ab}(Q^2, \varkappa_{\perp}^2, \varkappa_{\parallel}^2)}{[(\varkappa_{\parallel}+\frac{1}{2}Q)^2 + \varkappa_{\perp}^2]\,[(\varkappa_{\parallel}-\frac{1}{2}Q)^2 + \varkappa_{\perp}^2]}, \tag{C2}$$

where cylindrical coordinates around $\boldsymbol{Q}$ have been used for $\boldsymbol{\kappa}$.

In the complex $\varkappa_{\parallel}$-plane the integrand of (C2) has poles at $\varkappa_{\parallel} = \pm\frac{1}{2}Q \pm \mathrm{i}\varkappa_{\perp}$. Upon analytical continuation in t away from the negative t axis, counter-clockwise and clockwise by putting $Q = |Q|\exp(\mathrm{i}\varphi)$ with φ running from 0 to $\frac{1}{2}\pi$ and $-\frac{1}{2}\pi$, respectively, the integration contour in the complex $\varkappa_{\parallel}$ plane has to be deformed so as to avoid the poles. Although the contours for $\varphi = \frac{1}{2}\pi$ and $\varphi = -\frac{1}{2}\pi$ are different, the resulting integrals over $\varkappa_{\parallel}$ turn out to be equal if $\varkappa_{\perp}$ is different from $\frac{1}{2}|Q|$. When $\varkappa_{\perp}$ equals $\frac{1}{2}|Q|$ the contour gets pinched for φ approaching the values $\pm\frac{1}{2}\pi$. As a result a pole on the $\varkappa_{\perp}$-integration path shows up at $\varkappa_{\perp} = \frac{1}{2}|Q|$. The prescription for analytical continuation requires this pole to be avoided,

through the upper $\varkappa_\perp$ plane for $\varphi \to \frac{1}{2}\pi$, and through the lower $\varkappa_\perp$ plane for $\varphi \to -\frac{1}{2}\pi$ (*v.* fig 1a). As a consequence the ensuing values for F_{ind}, *viz.* $F_{\text{ind}}(t - \text{i}0)$ and $F_{\text{ind}}(t + \text{i}0)$, respectively, with $t > 0$, are different. In fact, in the former case one finds:

$$F_{\text{ind}}(t - \text{i}0) = \frac{|Q|}{4(2\pi)^3} \int_0^\pi \mathrm{d}\psi \int_{C_\psi} \mathrm{d}\varkappa_{\|} \frac{\varrho\, \mathrm{e}^{\mathrm{i}\psi} f_{\text{ab}}(-|Q|^2, \frac{1}{4}|Q|^2, \varkappa_{\|}^2)}{(\varkappa_{\|}^2 + |Q|^2)(\varkappa_{\|}^2 + \varrho^2\, \mathrm{e}^{2\mathrm{i}\psi})}, \tag{C3}$$

where the small arc around $\frac{1}{2}|Q|$ in the $\varkappa_\perp$-integration path has been parametrized by polar coordinates (ϱ, ψ). The corresponding contours in the $\varkappa_{\|}$ plane have been denoted by C_ψ and have been drawn in fig. 1b, c, d for $\psi = \pi$, $\frac{1}{2}\pi$ and 0, respectively. An expression analogous to (C3) may be found for $F_{\text{ind}}(t + \text{i}0)$.

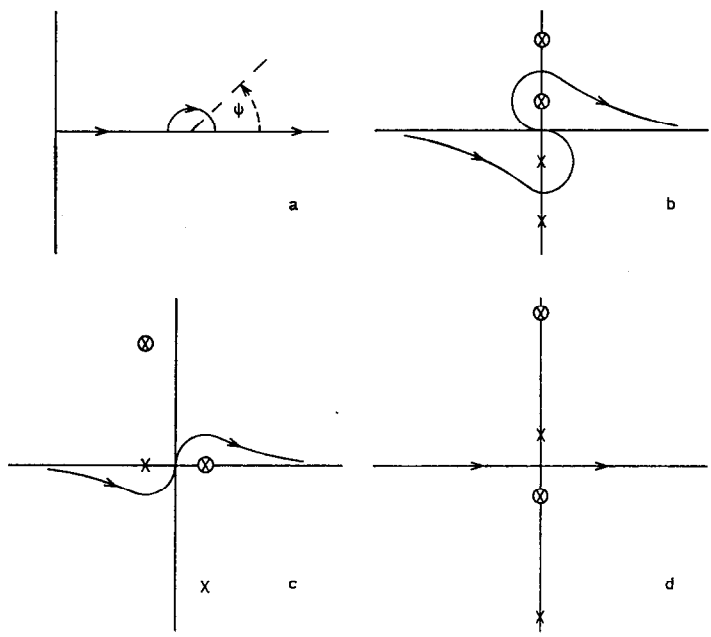

Fig. 1. Integration contours, after counter-clockwise analytical continuation towards positive t, (a) in the $\varkappa_\perp$ plane, (b, c, d) in the $\varkappa_{\|}$ plane.

The discontinuity of $F_{\text{ind}}(t)$ across the cut along the positive t axis follows by subtracting (C3) from its counterpart. Calculating the residues arising from the different circumventions of the poles in the $\varkappa_{\|}$ plane, one gets

$$F_{\text{ind}}(t + \text{i}0) - F_{\text{ind}}(t - \text{i}0) = -\tfrac{1}{16}\pi^{-1} t^{-\frac{1}{2}} f_{\text{ab}}(-t, \tfrac{1}{4}t, 0), \tag{C4}$$

which leads upon comparing with (C1) to formula (73) of the main text.

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REFERENCES

1) Michels, M.A.J. and Suttorp, L.G., Physica **59** (1972) 609, **61** (1972) 481, 506, 517; Suttorp, L.G., Physica **64** (1973) 217 (referred to as I–V, respectively).
2) Michels, M.A.J. and Suttorp, L.G., Phys. Letters **42A** (1973) 473.
3) Rodberg, L.S., Phys. Rev. **110** (1958) 277.
4) De Groot, S.R. and Suttorp, L.G., Foundations of Electrodynamics, North-Holland Publ. Co. (Amsterdam, 1972).
5) Casimir, H.B.G. and Polder, D., Phys. Rev. **73** (1948) 360.
6) Mavroyannis, C. and Stephen, M.J., Mol. Phys. **5** (1962) 629.
7) Condiff, D.W., University of Wisconsin Theoretical Chemistry Institute Report WIS-TCI-155 (1966).
8) Feinberg, G. and Sucher, J., J. chem. Phys. **48** (1968) 3333; Phys. Rev. **A2** (1970) 2395.
9) Gomberoff, L. and Power, E.A., Proc. Roy. Soc. **A295** (1966) 476.
10) See, *e.g.*, Edmonds, A.R., Angular Momentum in Quantum Mechanics, Princeton University Press (Princeton, 1957).
11) Au, C.-K.E. and Feinberg, G., Phys. Rev. **A6** (1972) 2433.
12) Rotenberg, M., Bivins, R., Metropolis, N. and Wooten, J.K., The 3*j*- and 6*j*-symbols, Technology Press, M.I.T. (Cambridge, 1959).