CHANGE OF RADIATION FREQUENCY IN RESONANCE FLUORESCENCE

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Submitted July 24, 1968

Zh. Eksp. Teor. Fiz. 56, 362–369 (January, 1969)

We investigate resonant fluorescence in fields of sufficient intensity to cause saturation and multiphoton scattering. We calculate the scattering matrix in the resonance approximation with allowance for multiphoton processes. It is shown that in multiphoton processes a change occurs in the spectrum of the scattered radiation (a "halo" appears around the scattered line). Concrete calculations are made for two-photon scattering.

THE quantum theory of resonant fluorescence was developed by Weisskopf,[1] who regarded the scattering process as absorption followed by re-radiation of one photon. Such an analysis is valid if the number of photons colliding with the atom during the time of spontaneous emission $\Delta t = 1/\gamma$ (where γ is the Einstein coefficient) is much smaller than unity:

$$\Delta N = \sigma n c \Delta t \ll 1. \tag{1}$$

Here n is the photon flux density and σ is the scattering cross section. Expressing the average density of the photon flux in terms of the electric field intensity E and assuming that the cross section is equal to the square of the resonant emission wavelength, it is easy to verify that ΔN coincides in order of magnitude with the parameter $(DE/\hbar\gamma)^2$ (D is the dipole moment of the transition), and consequently condition (1) is equivalent to the requirement

$$(DE/\hbar\gamma)^2 \ll 1. \tag{2}$$

For laser light sources, the spectral radiation density is so large that this parameter can no longer be regarded as small, and multiphoton processes begin to play a role. This paper is devoted to the analysis of two-photon collisions in resonant fluorescence.

Multiphoton processes come into play at field intensities much smaller than the intra-atomic intensities (by a factor γ/Ω, where Ω is the transition frequency), and we can therefore eliminate from the interaction Hamiltonian V the term quadratic in the field and to assume that[2]

$$V = -\frac{e}{mc}\mathrm{pA}. \tag{3}$$

Since we are considering resonant scattering, we can confine ourselves to the two-level model of the Hamiltonian and exclude from the Hamiltonian (3), in the interaction representation, the nonresonant terms that oscillate at double the optical frequency, the contribution of which is also of the order of γ/Ω.

We consider a two-level atom with wavefunctions $\varphi_0(\mathbf{x})$ and $\varphi_1(\mathbf{x})$ of the ground and excited states, respectively. The wave function of the atom-field system is written in the form

$$\psi(t) = G_0(t)\varphi_0(\mathbf{x}) + G_1(t)\varphi_1(\mathbf{x}),$$

where $G_0(t)$ and $G_1(t)$ are the magnetic field state vectors. We use a rationalized system of units and assume $\hbar = c = 1$.

The Schrödinger equation for the vectors $G_0(t)$ and $G_1(t)$ is written in the form

$$\frac{d}{dt}\begin{pmatrix} G_1(t) \\ G_0(t) \end{pmatrix} = i\begin{pmatrix} 0 & V^-(t) \\ V^+(t) & 0 \end{pmatrix}\begin{pmatrix} G_1(t) \\ G_0(t) \end{pmatrix}, \tag{4}$$

where

$$V^-(t) = \int f_j(\mathbf{k}) a_j(\mathbf{k}) e^{i(k-\Omega)t}\, d\mathbf{k}, \quad V^+(t) = \int f_j^*(\mathbf{k}) a_j^+(\mathbf{k}) e^{i(k-\Omega)t}\, d\mathbf{k}; \tag{5}$$

$$f_j(\mathbf{k}) = \frac{1}{(2\pi)^{3/2}}\frac{e}{m\sqrt{2k}}\langle\varphi_1(\mathbf{x})\,|\,p_j e^{i\mathbf{k}\mathbf{x}}\,|\,\varphi_0(\mathbf{x})\rangle, \tag{6}$$

Ω is the transition frequency. Summation over repeated vector indices is implied. The operators $a_j^-(\mathbf{k})$ and $a_j^+(\mathbf{k})$ satisfy the following commutation relations:

$$[a_j^-(\mathbf{k}),\, a_{j'}{}^+(\mathbf{k}')] = \left(\delta_{jj'} - \frac{k_j k_{j'}}{k^2}\right)\delta(\mathbf{k}-\mathbf{k}'). \tag{7}$$

In the dipole approximation, when the radiation wavelength is much larger than the atom dimensions, we can assume that $\exp(i\mathbf{k}\cdot\mathbf{x}) \approx 1$ (for an atom situated at the origin), and (6) takes the form

$$f_j(\mathbf{k}) = iD_j\Omega / 4\pi^{3/2}\sqrt{k}, \tag{8}$$

where the dipole-moment vector D of the transition can be regarded as real for a two-level atom.

The time-evolution operator[4]

$$u(t) = \begin{pmatrix} u_{11}(t) & u_{10}(t) \\ u_{01}(t) & u_{00}(t) \end{pmatrix}$$

satisfies the equation (see (4))

$$\frac{d}{dt}\begin{pmatrix} u_{11}(t) & u_{10}(t) \\ u_{01}(t) & u_{00}(t) \end{pmatrix} = i\begin{pmatrix} 0 & V^-(t) \\ V^+(t) & 0 \end{pmatrix}\begin{pmatrix} u_{11}(t) & u_{10}(t) \\ u_{01}(t) & u_{00}(t) \end{pmatrix}.$$

This matrix equation breaks up into two independent systems of equations. It suffices to consider one of them:

$$\frac{d}{dt}u_{10}(t) = iV^-(t)u_{00}(t), \quad \frac{d}{dt}u_{00}(t) = iV^+(t)u_{10}(t), \tag{9}$$

with initial conditions

$$u_{00}(-\infty) = 1, \quad u_{10}(-\infty) = 0, \tag{10}$$

inasmuch as in the scattering of the wave packet before and after the scattering process the atom is in the ground state ($G_1(-\infty) = 0$). The scattering matrix here is $S = u_{00}(+\infty)$.

We seek a solution in the form

$$u_{00}(t)=1+\sum_{n=1}^{\infty}\int c_n(k_1,\ldots,k_n;\varkappa_1,\ldots,\varkappa_n)\exp\Big\{i\sum_{\lambda=1}^{n}(k_\lambda-\varkappa_\lambda)t\Big\}\times\prod_{\nu=1}^{n}f_{i_\nu}^{*}(\mathbf{k}_\nu)a_{i_\nu}^{+}(\mathbf{k}_\nu)d\mathbf{k}_\nu\prod_{\mu=1}^{n}f_{j_\mu}(\boldsymbol{\varkappa}_\mu)a_{j_\mu}^{-}(\boldsymbol{\varkappa}_\mu)d\boldsymbol{\varkappa}_\mu,$$

$$u_{10}(t)=\sum_{n=1}^{\infty}\int b_n(k_1,\ldots,k_{n-1};\varkappa_1,\ldots,\varkappa_n)\times\exp\Big\{i\Big[\sum_{\lambda=1}^{n-1}(k_\lambda-\varkappa_\lambda)+\Omega-\varkappa_n\Big]t\Big\}\times\prod_{\nu=1}^{n-1}f_{i_\nu}^{*}(\mathbf{k}_\nu)a_{i_\nu}^{+}(\mathbf{k}_\nu)d\mathbf{k}_\nu\prod_{\mu=1}^{n}f_{j_\mu}(\boldsymbol{\varkappa}_\mu)a_{j_\mu}^{-}(\boldsymbol{\varkappa}_\mu)d\boldsymbol{\varkappa}_\mu, \tag{11}$$

where the coefficients $c_n(\mathbf{k}_1,\ldots,\mathbf{k}_n;\kappa_1,\ldots,\kappa_n)$ and $b_n(\mathbf{k}_1,\ldots,\mathbf{k}_{n-1};\kappa_1,\ldots,\kappa_n)$ are to be determined (this form of the solution is suggested by perturbation theory). For a unique determination of c_n and b_n they must be regarded as symmetrical functions of the sets of variables $\mathbf{k}_\nu$ and κ_μ.

Substituting (11) in (9) and using the commutation relation (7), we obtain an equation for the coefficients

$$\Big[\sum_{\nu=1}^{n}(k_\nu-\varkappa_\nu)\Big]c_n(k_1,\ldots,k_n;\varkappa_1,\ldots,\varkappa_n)=\frac{1}{n}\sum_{\nu=1}^{n}b_n(k_1,\ldots,k_{\nu-1},k_{\nu+1},\ldots,k_n;\varkappa_1,\ldots,\varkappa_n),$$

$$\Big[\sum_{\nu=1}^{n-1}(k_\nu-\varkappa_\nu)+\Omega-\varkappa_n\Big]b_n(k_1,\ldots,k_{n-1};\varkappa_1,\ldots,\varkappa_n)=n\int\frac{\gamma(k_n)}{2\pi}c_n(k_1,\ldots,k_n;\varkappa_1,\ldots,\varkappa_n)dk_n+\frac{1}{n}\sum_{\mu=1}^{n}c_{n-1}(k_1,\ldots,k_{n-1};\varkappa_1,\ldots,\varkappa_{\mu-1},\varkappa_{\mu+1},\ldots,\varkappa_n). \tag{12}$$

Here

$$\gamma(k)=\frac{e^2}{(2\pi)^2m^2}\int\frac{1}{2\varkappa}\langle\varphi_1(\mathbf{x})\,|\,p_je^{i\boldsymbol{\varkappa}\mathbf{x}}\,|\,\varphi_0(\mathbf{x})\rangle\langle\varphi_0(\mathbf{x})\,|\,p_{j'}e^{-i\boldsymbol{\varkappa}\mathbf{x}}\,|\,\varphi_1(\mathbf{x})\rangle\times\Big(\delta_{jj'}-\frac{\varkappa_j\varkappa_{j'}}{\varkappa^2}\Big)\delta(k-\varkappa)\,d\mathbf{x}. \tag{13}$$

It is easy to verify that in the dipole approximation and for frequencies away from resonance by an amount of the order of its width, we have

$$\gamma(k)=\gamma$$

accurate to γ/Ω, where γ is the Einstein coefficient of the $1\to0$ transition (in the ordinary nonrationalized units $\gamma=4D_j^2\Omega^3/3\hbar c^3$).

The solution of the system (12) and the determination of the scattering matrix are given in the Appendix. We shall henceforth consider two-photon scattering, so that it suffices to retain in the scattering-matrix series the terms that describe the simultaneous scattering of not more than two photons. After a simple transformation (see the Appendix), the scattering matrix takes in the approximation needed by us the form

$$S=1+2\pi i\int\frac{f_i^{*}(\mathbf{k})f_j(\boldsymbol{\varkappa})}{\Omega-k-i\gamma/2}a_i^{+}(\mathbf{k})a_j^{-}(\boldsymbol{\varkappa})\delta(k-\varkappa)d\mathbf{k}\,d\boldsymbol{\varkappa}+2\pi i\sum_{P(k_\nu),\,P(\varkappa_\mu)}\int\frac{d\mathbf{k}_1\,d\mathbf{k}_2\,d\boldsymbol{\varkappa}_1\,d\boldsymbol{\varkappa}_2}{(\Omega-k_1-i\gamma/2)(\Omega-k_2-i\gamma/2)}\Big[\pi i\delta(k_1-\varkappa_1)-\frac{1}{\Omega-\varkappa_1-i\gamma/2}\Big]f_{i_1}^{*}(\mathbf{k}_1)f_{i_2}^{*}(\mathbf{k}_2)f_{j_1}(\boldsymbol{\varkappa}_1)f_{j_2}(\boldsymbol{\varkappa}_2)\times\frac{1}{(2!)^2}a_{i_1}^{+}(\mathbf{k}_1)a_{i_2}^{+}(\mathbf{k}_2)a_{j_1}^{-}(\boldsymbol{\varkappa}_1)a_{j_2}^{-}(\boldsymbol{\varkappa}_2)\delta(k_1+k_2-\varkappa_1-\varkappa_2), \tag{14}$$

where the symbol $P(\mathbf{k}_\nu)$ denotes that the summation is carried out over all the permutations of $\mathbf{k}_\nu$.

The energy scattered in a solid-angle element in the direction of the wave vector $\mathbf{k}$ is given by the expression[3]

$$W(\mathbf{k})=\langle G\,|\,S^{+}k^3a_i^{+}(\mathbf{k})a_i^{-}(\mathbf{k})S\,|\,G\rangle, \tag{15}$$

where G is the vector of the initial state of the electromagnetic field. It is easy to verify that the first two terms in (14) describe the usual resonant fluorescence. The cross section of the resonant fluorescence is

$$\sigma(k,\psi\ \theta)=\frac{D^4\Omega^4\sin^2\psi\cos^2\theta}{16\pi^2[(\Omega-k)^2+\gamma^2/4]},$$

or, in the usual nonrationalized units,

$$\sigma(\omega,\psi,\theta)=\frac{D^4\Omega^4\sin^2\psi\cos^2\theta}{[(\Omega-\omega)^2+\gamma^2/4]\hbar^2c^4},$$

where θ is the angle between the vector D and the polarization vector of the incident radiation, ψ is the angle between D and the direction of observation of the scattered light, and ω is the radiation frequency.

The last term in (14) is responsible for the two-photon scattering, and the first term in the square bracket describes, as can be readily verified, the scattering without a change of frequency, while the second leads to a change of the spectral composition of the scattered radiation (owing to the redistribution of the energy between the two scattered photons). It follows from (15) that the scattered energy is expressed in terms of the Fourier components of the correlation functions of the field[5]. The general expression for $W(\mathbf{k})$ is quite complicated, and we present the result for a monochromatic fully-coherent[5] light field, which apparently describes sufficiently well the laser radiation. In this case, the energy scattered without a change of the frequency is

$$W'(\omega,\theta,\psi)=\sigma(\omega,\theta,\psi)\Big[1-\frac{(\mathbf{ED})^2}{\hbar^2}\frac{1}{(\Omega-\omega)^2+\gamma^2/4}\Big]I, \tag{16}$$

where I is the energy flux of the incident radiation and E is the amplitude of the electric field intensity of the light wave. The second term in the square brackets describes the saturation effect[6].

In addition, there is scattered radiation with a continuous spectrum the intensity of which is given by the expression

$$\frac{d}{d\omega'}W''(\omega,\omega';\theta,\psi)=\sigma(\omega,\theta\ \psi)\frac{(\mathbf{ED})^2}{2\hbar^2[(\Omega-\omega)^2+\gamma^2/4]}IF(\omega,\omega'), \tag{17}$$

where

$$F(\omega,\omega')=\frac{\gamma}{\pi}\frac{(\Omega-\omega)^2+\gamma^2/4}{[(\Omega-\omega')^2+\gamma^2/4][(\Omega+\omega'-2\omega)^2+\gamma^2/4]}$$

determines the form of the contour of the scattered radiation, and

$$\int F(\omega,\omega')d\omega'=1.$$

Here ω, as before, is the frequency of the incident radiation and ω' is the frequency of the scattered radiation. At exact resonance $\omega=\Omega$ the spectrum of the scattered radiation has the same form as the square of the dispersion contour. It follows from (16) and (17) that the saturation parameter, determined from the total scattered energy, is equal to

$$\frac{1}{2}\frac{(DE)^2}{\hbar^2}\Big[(\Omega-\omega)^2+\frac{\gamma^2}{4}\Big]^{-1}.$$

In the case of scattering in a gas, it is necessary to take into account the motion of the atoms. For an atom moving with velocity $\mathbf{v}$, the formula for the spectrum of the scattered radiation is obtained from (17) by replacing ω and ω' respectively by $\omega - \mathbf{k}\cdot\mathbf{v}$ and $\omega' - \mathbf{k}'\cdot\mathbf{v}$. The obtained expression must be averaged over the Maxwellian velocity distribution

$$(\sqrt{\pi}\,\bar v)^{-3}\exp\{-(\mathbf{v})^2/\bar v^2\},\qquad \bar v=\sqrt{2kT/M}.$$

We present the result of averaging only for forward scattering, for under ordinary conditions the Doppler line width greatly exceeds the radiative line width, and at large scattering angles the Doppler effect masks the described broadening. Assuming that $\gamma \ll k\bar v$, we obtain

$$\frac{dW''}{d\omega'}=\frac{3}{\sqrt{\pi}}\frac{\gamma}{k_0\bar v}\exp\Big\{-\frac{(\Omega-\omega)^2}{(k_0\bar v)^2}\Big\}\frac{\Omega^4D^4\sin^2\psi\cos^2\theta}{\hbar^2c^4[(\omega'-\omega)^2+\gamma^2]}\times\frac{(ED)^2}{2\hbar^2[(\omega'-\omega)^2+\gamma^2/4]}I\frac{1}{\gamma},$$

where $k_0 = \Omega/c$.

We did not take into account here the collisions, and therefore our results are valid so long as the collision line width is smaller than the radiative line width. Allowance for collisions calls for an additional analysis, and we hope to deal with this question in another paper.

In conclusion, the authors thank S. G. Rautian for a discussion of the work and for valuable remarks.

APPENDIX

To solve Eqs. (12) it is necessary to establish the role for circuiting around the poles that arise when both halves of the equations are divided by

$$\sum_{\lambda=1}^{n}(k_\lambda-\varkappa_\lambda)\ \text{and}\ \sum_{\lambda=1}^{n-1}(k_\lambda-\varkappa_\lambda)+\Omega-\varkappa_n.$$

We shall show later that the initial conditions (10) correspond to the following circuiting rule:

$$\Big[\sum_{\nu=1}^{n}(k_\nu-\varkappa_\nu)-i\varepsilon\Big]c_n(k_1,\dots,k_n;\varkappa_1,\dots,\varkappa_n)=\frac{1}{n}\sum_{\nu=1}^{n}b_n(k_1,\dots,k_{\nu-1},k_{\nu+1},\dots,k_n;\varkappa_1,\dots,\varkappa_n),\tag{A.1}$$

$$\Big[\sum_{\nu=1}^{n-1}(k_\nu-\varkappa_\nu)+\Omega-\varkappa_n-i\varepsilon\Big]b_n(k_1,\dots,k_{n-1};\varkappa_1,\dots,\varkappa_n)=n\int\frac{\gamma(k_n)}{2\pi}c_n(k_1,\dots,k_n;\varkappa_1,\dots,\varkappa_n)dk_n+\frac{1}{n}\sum_{\mu=1}^{n}c_{n-1}(k_1,\dots,k_{n-1};\varkappa_1,\dots,\varkappa_{\mu-1},\varkappa_{\mu+1},\dots,\varkappa_n).\tag{A.2}$$

We express c_n in terms of b_n from (A.1) and substitute into the integral term of (A.2):

$$n\int_0^\infty\frac{\gamma(k_n)}{2\pi}c_n(k_1,\dots,k_n;\varkappa_1,\dots,\varkappa_n)dk_n=b_n(k_1,\dots,k_{n-1};\varkappa_1,\dots,\varkappa_n)\int_0^\infty dk_n\frac{\gamma(k_n)}{2\pi}\Big[\sum_{\lambda=1}^{n}(k_\lambda-\varkappa_\lambda)-i\varepsilon\Big]^{-1}+\int_0^\infty dk_n\frac{\gamma(k_n)}{2\pi}\Big[\sum_{\lambda=1}^{n}(k_\lambda-\varkappa_\lambda)-i\varepsilon\Big]^{-1}\times\sum_{\nu=1}^{n-1}b_n(k_1,\dots,k_{\nu-1},k_{\nu+1},\dots,k_n;\varkappa_1,\dots,\varkappa_n).\tag{A.3}$$

Using the relation

$$\int\frac{\varphi(x)}{x-i\varepsilon}dx=i\pi\varphi(0)+\int\frac{\varphi(x)}{x}dx,$$

where the integral on the right hand side is taken in the sense of the principal value, we obtain

$$\frac{1}{2\pi}\int_0^\infty dk_n\,\gamma(k_n)\Big[\sum_{\lambda=1}^{n}(k_\lambda-\varkappa_\lambda)-i\varepsilon\Big]^{-1}=\frac{i}{2}\gamma\Big[\sum_{\lambda=1}^{n-1}(\varkappa_\lambda-k_\lambda)+\varkappa_n\Big]+\frac{1}{2\pi}\int_0^\infty\gamma(k_n)\Big[\sum_{\lambda=1}^{n}(k_\lambda-\varkappa_\lambda)\Big]^{-1}dk_n\approx i\frac{\gamma}{2}+\Delta.$$

Here Δ is the change of the line frequency owing to the interaction of the atom with the radiation field (as usual[7]), γ and Δ are assumed independent of the frequency). We do not take into account the frequency shift Δ, assuming that it is already included in Ω. We shall show subsequently that the second integral in the right side of (A.3) vanishes, so that (A.2) takes the form

$$\Big[\sum_{\lambda=1}^{n-1}(k_\lambda-\varkappa_\lambda)+\Omega-\varkappa_n-i\frac{\gamma}{2}\Big]b_n(k_1,\dots,k_{n-1};\varkappa_1,\dots,\varkappa_n)=\frac{1}{n}\sum_{\mu=1}^{n}c_{n-1}(k_1,\dots,k_{n-1};\varkappa_1,\dots,\varkappa_{\mu-1},\varkappa_{\mu+1},\dots,\varkappa_n).\tag{A.4}$$

Equations (A.1) and (A.4) form a recurrent system of algebraic equations, the solution of which is

$$c_n(k_1,\dots,k_n;\varkappa_1,\dots,\varkappa_n)=\frac{1}{(n!)^2}\sum_{P(k_\nu)P(\varkappa_\mu)}\prod_{m=1}^{n}\Big\{\Big[\sum_{\nu=1}^{m}(k_\nu-\varkappa_\nu)-i\varepsilon\Big]^{-1}\times\Big[\sum_{\mu=1}^{m-1}(k_\mu-\varkappa_\mu)+\Omega-\varkappa_m-i\frac{\gamma}{2}\Big]^{-1}\Big\},$$

$$b_{n+1}(k_1,\dots,k_n;\varkappa_1,\dots,\varkappa_{n+1})=\frac{1}{n!(n+1)!}\sum_{P(k_\nu)P(\varkappa_\mu)}\Big[\sum_{\lambda=1}^{n}(k_\lambda-\varkappa_\lambda)+\Omega-\varkappa_{n+1}-i\frac{\gamma}{2}\Big]^{-1}\prod_{m=1}^{n}\Big\{\Big[\sum_{\nu=1}^{m}(k_\nu-\varkappa_\nu)-i\varepsilon\Big]^{-1}\times\Big[\sum_{\mu=1}^{m-1}(k_\mu-\varkappa_\mu)+\Omega-\varkappa_m-i\frac{\gamma}{2}\Big]^{-1}\Big\}.\tag{A.5}$$

We can now prove that the second integral in the right side of (A.3) vanishes. We note first that $c_n(k_1,\dots,k_n;\ \varkappa_1,\dots,\varkappa_n)$ and $b_n(k_1,\dots,k_{n-1};\ \varkappa_1\dots,\varkappa_n)$, in accord with (A.5), decrease rapidly with increasing differences $|\kappa_\nu-\Omega|$ and $|\kappa_\mu-\Omega|$, and therefore we can assume in this integral the dipole approximation for $\gamma(k)$, with accuracy of order γ/Ω, and to extend the integral to the entire real axis. After doing so, we immediately can verify, on the basis of the residue theorem, that the integral vanishes, since it contains a rational-fraction function all of whose poles in k_n lie in the upper half-plane, and which decreases sufficiently rapidly at infinity to ensure the vanishing of the integral over an infinitely remote semicircle.

We now return to the operator $u(t)$. With the aid of the equality[8]

$$\frac{e^{ixt}}{x-i0}\to\begin{cases}2\pi i\delta(x) & \text{for } t\to+\infty,\\ 0 & \text{for } t\to-\infty\end{cases}$$

we verify, first, that $u_{00}(-\infty)=1$, i.e., the obtained solution satisfies the initial condition, and second, we find the scattering matrix

$$S=1+2\pi i\sum_{n=1}^{\infty}\frac{1}{(n!)^2}\int\sum_{P(k_\nu)P(\varkappa_\mu)}\left[\sum_{\lambda=1}^{n-1}(k_\lambda-\varkappa_\lambda)+\Omega-\varkappa_n-i\frac{\gamma}{2}\right]^{-1}$$
$$\times\delta\left[\sum_{\lambda=1}^{n}(k_\lambda-\varkappa_\lambda)\right]\prod_{m=1}^{n-1}\left\{\left[\sum_{\nu=1}^{m}(k_\nu-\varkappa_\nu)-i\varepsilon\right]^{-1}\left[\sum_{\mu=1}^{m-1}(k_\mu-\varkappa_u)\right.\right.$$
$$\left.\left.+\Omega-\varkappa_m-i\frac{\gamma}{2}\right]^{-1}\right\}\prod_{\nu=1}^{n}f_{i_\nu}^{*}(\mathbf{k}_\nu)a_{i_\nu}^{+}(\mathbf{k}_\nu)d\mathbf{k}_\nu\prod_{\mu=1}^{n}f_{j_\mu}(\varkappa_\mu)a_{j_\mu}^{-}(\varkappa_\mu)d\varkappa.$$

It is easy to verify that $u_{01}(-\infty)=0$.

Let us consider the coefficient in the second-order term ($n=2$) in (A.6)

$$\frac{1}{4}\sum_{P(k_\nu)P(\varkappa_\mu)}\frac{\delta(k_1+k_2-\varkappa_1-\varkappa_2)}{(k_1-\varkappa_1-i\varepsilon)(\Omega-\varkappa_1-i\gamma/2)(\Omega+k_1-\varkappa_1-\varkappa_2-i\gamma/2)}.$$

Using the equation

$$k_1+k_2=\varkappa_1+\varkappa_2, \tag{A.7}$$

this coefficient can be readily transformed into

$$\frac{1}{4}\sum_{P(k_\nu)P(\varkappa_\mu)}\frac{\delta(k_1+k_2-\varkappa_1-\varkappa_2)}{(k_1-\varkappa_1-i\varepsilon)(\Omega-\varkappa_1-i\gamma/2)(\Omega-k_2-i\gamma/2)}.$$

Further, from the identity

$$\frac{1}{k_1-\varkappa_1-i\varepsilon}\frac{1}{\Omega-\varkappa_1-i\gamma/2}$$
$$=\frac{1}{\Omega-k_1-i\gamma/2}\left(\frac{1}{k_1-\varkappa_1-i\varepsilon}-\frac{1}{\Omega-\varkappa_1-i\gamma/2}\right)$$

(which holds true in the limit as $\epsilon\to 0$) we obtain, again using (A.7), that this coefficient can be represented in the form

$$\frac{1}{4}\sum_{P(k_\nu)P(\varkappa_\mu)}\frac{1}{\Omega-k_1-i\gamma/2}\frac{1}{\Omega-\varkappa_1-i\gamma/2}$$
$$\times\left[i\pi\delta(k_1-\varkappa_1)-\frac{1}{\Omega-\varkappa_1-i\gamma/2}\right]\delta(k_1+k_2-\varkappa_1-\varkappa_2).$$

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Translated by J. G. Adashko
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