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Nonresonant corrections for the optical resonance frequency measurements in the hydrogen atom

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The deviation of the natural spectral line profile from the Lorentz shape for the optical resonant frequency measurements is considered. This deviation leads to an asymmetry, which is mainly due to nonresonant correction to the resonant Lorentz profile. The nonresonant corrections are studied for the different types of the atomic resonant experiments. The most accurate recent optical resonance experiments are analyzed, i.e., the two-photon 1*s*-2*s* resonance excitation of the hydrogen atom with the delayed decay in the external electric field. The description of the nonresonant correction in the latter case requires the employment of QED with different in and out Hamiltonians. The nonresonant corrections for this experiment are investigated and found to be about 10^{-5} Hz, while the recent experimental uncertainty is 34 Hz and in the near feature is expected to be a few hertz. The projected 1*s*-2*s* resonance excitation experiment with the three-photon ionization detection (which is now in progress) is also considered.

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I. INTRODUCTION

The recent superaccurate measurements of the resonance transition frequency in the hydrogen atom [1,2] require a careful analysis of the limits for the validity of the resonance approximation in such experiments. These limits are set by nonresonant (NR) corrections to the standard Lorentz line profile. Here we address only the natural spectral line profile, disregarding Doppler, time-of-flight, and collisional broadening. It is assumed that the accuracy of the experiment allows for the observation of natural line profile, like in [3]. In the resonance approximation, which reduces the description of the spectral line profile to the Lorentz contour, the line profile is described by two parameters, resonance frequency w^{res} and width Γ and is symmetric with respect to w^{res}. In the early paper by Low [4] it was pointed out that resonance approximation is valid only up to a certain level of accuracy, which is defined by NR corrections. Beyond this limit the line profile becomes asymmetric and the interpretation of w^{res} becomes ambiguous. Moreover, the NR corrections are, in principle, process dependent, so the measured line profile also becomes dependent on the process of measurement. The other source of the line profile asymmetry is the dependence of transition rates on frequency. The NR corrections are, however, dominant in the most cases.

The magnitude of NR correction δw^{res} as it was estimated by Low [4] for H-like ions appeared to be very small: $\delta w^{res}/\Gamma \approx \alpha(\alpha Z)^2$ for the dipole optical transition, where α is the fine structure constant and Z is the charge of the nucleus. Accordingly, for the long time after the arrival of the paper of Low [4] there was no need to consider these corrections in the optical experiments. Recently the interest to NR corrections was triggered by the new very accurate optical resonance experiments [1,2] and the various theoretical estimates were given in [5,6]. Recently the NR corrections were considered also within an astrophysical context [7]. In this work we will present the evaluation of NR corrections for the frequency measurements in experiment [1,2]. The calculations will be done for the hydrogen atom or H-like ions.

One has to distinguish between two types of the possible resonant optical experiments. All the processes consist of three stages: excitation of the certain intermediate states, its propagation, and its decay. The coherence in the sum over the intermediate state can be destroyed by the collisions (the first type of the experiments) or conserved (the second type of the experiments). The analysis shows that the NR corrections in case of the first type of resonance experiments are essentially smaller than for the experiments of second type. However, in the ultimate case when the natural line profile can be observed and there are no phase distortions, both types of resonance experiments actually coincide and the NR corrections to the frequency for both experiments are the same. In what follows we will consider only this situation. Our argumentation is the following: when the natural line profile is not observed, i.e., screened by the collisional, time-of-flight, Doppler broadening, etc. [1], the latter effects define the possible uncertainty of the experimental determination of the transitional frequency. This uncertainty is evidently larger than the effect of the NR corrections but can be decreased by the technical improvements. The distortion of the line profile caused by the NR corrections is the unavoidable systematic effect that should be always taken into account.

The absolute uncertainty remaining in the two-photon resonance frequency measurement [1] for 1*s*-2*s* transition in hydrogen is 34 Hz, which corresponds to the relative accuracy of 1.4×10^{-14}. However, in [1] a special scheme of registration was employed, which requires the special theoretical consideration. In [1] the atoms are excited by means of laser radiation via two-photon absorption from the ground 1*s* state. This excitation occurs in a spatial region free of external fields. The excited atoms are moving subsequently to another spatial region where they experience an additional weak electric field. In the presence of such a field the $2s$ and $2p$ states are mixed and the atoms decay via ordinary $2p$-$1s$ transition. This radiation is detected and provides the necessary information for the extraction of $1s$-$2s$ frequency value from the experimental data. In the rest frame of an atom it looks like the excitation occurs in the absence of the electric field and then the electric field is turned on (delayed decay). In terms of QED, the initial and final states of an atom are described then by different Hamiltonians $\hat{H}_{in}$ and $\hat{H}_{out}$. The QED theory with different in and out Hamiltonians was considered by several authors. Here, we refer to the book by Fradkin *et al.* [8] (see there the references to other works in the area).

In this paper we will discuss also another possible method for registration of the excited $2s$ state in hydrogen: the three-photon ionization [9].

The paper is organized as follows. In Sec. II the processes of the first and second types as defined above will be compared. The standard expressions for the excitation and decay probabilities will be derived. In Sec. III we describe the differential cross section for the process of the resonant photon scattering on atomic electron, having the goal to stress the difference between this process (of the second type) and process of level excitation: in the latter case the information on the excitation process is usually lost when the decay of the excited state is registered and the natural line profile is screened by the collisional and Doppler broadening (process of the first type). However we always will consider the resonant scattering process for defining the NR correction even in cases when in the real experiment the natural line profile is not yet observed. In Sec. IV the NR correction to the process of the resonant photon scattering on the atomic electron will be introduced. In Sec. V we introduce the formulation and present the generic expressions for QED with different in and out Hamiltonians, which are necessary for description of delayed decay of the $2s$ level in an external electric field. This QED theory is applied in Sec. VI to the description of the experiment [1], considered as a resonance process of two-photon excitation of $2s$ level with a subsequent delayed decay in the external electric field. These derivations will result in Sec. VII as the NR correction values for the frequency measurement with this type of experiment. In Sec. VIII all results are summarized and the final conclusions are made.

II. RESONANT EXCITATION OF ATOMIC LEVELS AND RESONANCE PHOTON SCATTERING (TOTAL CROSS SECTIONS)

We start with introducing the resonant elastic scattering of the photon on the bound electron in an atom in the absence of external fields. The corresponding Feynman graphs are depicted in Fig. 1. The amplitude of this process can be written in the form

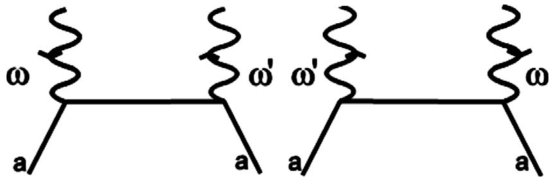

FIG. 1. The Feynman graphs describing the elastic photon scattering on atomic electron. The solid line describes the electron in the field of the nucleus; the wavy line with an arrow denotes the absorption or the emission of a photon. Symbol a corresponds to the certain electron level (njl) in an atom; ω and ω' denote the absorbed and emitted photon frequencies. The right-hand side of both graphs corresponds to the initial state and the left-hand side corresponds the final state. In case of the elastic scattering (the initial and final atomic states are the same) $\omega=\omega'$.

$$U^{JM_f,JM_i}_{n_0j_0l_0m_f,n_0j_0l_0m_i} = \sum_{njlm} \frac{\langle n_0j_0l_0m_f|A^{(E,M)^\dagger}_{JM_f}|njm\rangle\langle njm|A^{(E,M)}_{JM_i}|n_0j_0l_0m_i\rangle}{E_{njl}-E_{n_0j_0l_0}-\omega} + \sum_{njlm} \frac{\langle n_0j_0l_0m_f|A^{(E,M)}_{JM_i}|njm\rangle\langle njm|A^{(E,M)^\dagger}_{JM_f}|n_0j_0l_0m_i\rangle}{E_{njl}-E_{n_0j_0l_0}+\omega}. \tag{1}$$

Here the quantum numbers $njlm$ represent the standard set of one-electron quantum numbers for the electron in the hydrogen atom: principal quantum number n, total angular momentum j, its projection m, and orbital angular momentum l which fixes the parity of the state. The photons are characterized, apart from the frequency ω, by the total angular momentum J, its projection M, and the type of the photon (electric E or magnetic M), which fixes the parity. For simplicity we assume that the absorbed and emitted photons have the same J values as it happens, for example, for $1s$-$2p$ (E1 photon) transition. The indices i,f denote the initial and final states of the particles. The operator $A^{E,M}_{JM}$ corresponds to the photon absorption and $A^{(E,M)^\dagger}_{JM}$ denotes the photon emission.

The absorption operator looks like

$$A^{E,M}_{JM} = e\vec{\gamma}\cdot\vec{A}^{E,M}_{\omega JM}, \tag{2}$$

$$\vec{A}^{E}_{\omega JM}(\vec{r}) = \sqrt{\frac{\omega}{2\pi}}\left(\sqrt{\frac{J}{2J+1}}g_{J+1}(\omega r)\vec{Y}_{JJ+1M}(\vec{n}) - \sqrt{\frac{J+1}{2J+1}}g_{J-1}(\omega r)\vec{Y}_{JJ-1M}(\vec{n})\right), \tag{3}$$

$$\vec{A}^{M}_{\omega JM}(\vec{r}) = \sqrt{\frac{\omega}{2\pi}}\vec{Y}_{JJM}(\vec{n}). \tag{4}$$

Here the spherical vector functions $\vec{Y}_{JLM}(\vec{n})$ are introduced,

$$\vec{Y}_{JLM}(\vec{n}) = \sum_{M_L,\mu} C^{L1}_{JM}(M_L,\mu) Y_{LM_L}(\vec{n}) \vec{\chi}_\mu, \tag{5}$$

where $\vec{n}=\frac{\vec{r}}{r}$, $Y_{LM_L}(\vec{n})$ is the ordinary spherical function, $\vec{\chi}_\mu$ is the vector spin function for the photon,

$$g_l(\omega r) = \frac{(2\pi)^{3/2}}{\sqrt{\omega r}} J_{l+1/2}(\omega r),$$

and $J_{l+1/2}(\omega r)$ is the Bessel function. We use the standard notation $\vec{\gamma}$ for the Dirac matrices and e is the electron charge. The relativistic units $\hbar = c = 1$ ($\hbar$ is the Planck constant and c is the speed of light) are employed.

Imposing the resonance approximation together with the resonance condition

$$\omega = E_{n_1 j_1 l_1} - E_{n_0 j_0 l_0}, \tag{6}$$

only the terms with $njl = n_1 j_1 l_1$ in the first sum in Eq. (1) should be retained; the second sum should be fully omitted.

This yields

$$U^{JM_f,JM_i}_{n_0 j_0 l_0 m_f, n_0 j_0 l_0 m_i}(n_1 j_1 l_1) = \sum_m \frac{\langle n_0 j_0 l_0 m_f | A^{(E,M)^\dagger}_{JM_f} | n_1 j_1 l_1 m\rangle \langle n_1 j_1 l_1 m | A^{(E,M)}_{JM_i} | n_0 j_0 l_0 m_i\rangle}{E_{n_1 j_1 l_1} - E_{n_0 j_0 l_0} - \omega}. \tag{7}$$

The difference between the two types of the resonant processes, mentioned in Sec. I, concerns the summation over the projection m in Eq. (7). The process of excitation of atomic level corresponds to the case when this summation should be decoupled and restricted to one particular value of the projection. This implies that the summation over m in Eq. (7) should be omitted and $|n_1 j_1 l_1 m\rangle\langle n_1 j_1 l_1 m|$ should be replaced by $|n_1 j_1 l_1 m\rangle\langle n_1 j_1 l_1 m'|$ with the fixed m, m' values. In the expression for the transition probability, the summation over m and averaging over m' should be restored. This decoupling may occur due to some averaging process that intervenes between the absorption of the photon and emission of the photon. In particular, it could be multiple scattering, where the value of the projection m changes many times. In the process of scattering the atom may change also the values njl, but then it leaves the resonant process (of course, due to the collisions some atoms can be kicked out of the beam and thus not detected at all).

If the values njl remain equal to $n_1 j_1 l_1$ and only m changes, the atom remains within the resonance. We are interested in the latter case. The result of such changes of the m value can be interpreted as a random process and thus decoupling arise. In this way the experimental methods of the resonant excitation of atomic levels should be understood.

Now we can write down the expression for the resonant process probability (excitation cross section) as

$$dW^{(J)}_{n_0 j_0 l_0, n_0 j_0 l_0}(n_1 j_1 l_1) = \frac{1}{(2J+1)(2j_0+1)(2j_1+1)} \times \sum_{M_i M_f} \sum_{m_i m_f} \sum_{m,m'} |\langle n_0 j_0 l_0 m_f | A^{(E,M)^\dagger}_{JM_f} | n_1 j_1 l_1 m\rangle|^2 \times \frac{|\langle n_1 j_1 l_1 m' | A^{(E,M)}_{JM_i} | n_0 j_0 l_0 m_i\rangle|^2}{(E_{n_1 j_1 l_1} - E_{n_0 j_0 l_0} - \omega)^2 + \frac{1}{4}\Gamma^2_{n_1 j_1 l_1}} d\omega. \tag{8}$$

In Eq. (8) it is assumed that both the initial electrons and photons are nonpolarized.

Here the width of the excited level $\Gamma_{n_1 j_1 l_1}$ is introduced in a standard way. The operators $A^{(E,M)}_{JM}$ are the spherical tensors of the rank J with the JM component in the electron space. Then, performing the summations over all the angular momentum projections in Eq. (4) with the use of the Wigner-Eckart theorem for the matrix elements in Eq. (8),

$$\langle n' j' l' m' | A_{JLM} | njlm\rangle = (-1)^{j'-m'} \begin{pmatrix} j' & J & j \\ m' & M & \bar{m} \end{pmatrix} \langle n' j' l' \| A_{JL} \| njl\rangle, \tag{9}$$

where $\langle n' j' l' \| A_{JL} \| njl\rangle$ is the angular reduced matrix element, yields

$$dW^{(J)}_{n_0 j_0 l_0, n_0 j_0 l_0}(n_1 j_1 l_1) = \frac{W^{em(J)}_{n_0 j_0 l_0, n_1 j_1 l_1} W^{ab(J)}_{n_1 j_1 l_1, n_0 j_0 l_0}}{(E_{n_1 j_1 l_1} - E_{n_0 j_0 l_0} - \omega)^2 + \frac{1}{4}\Gamma^2_{n_1 j_1 l_1}} d\omega, \tag{10}$$

where

$$W^{ab(J)}_{n_1 j_1 l_1, n_0 j_0 l_0} = \frac{|\langle n_1 j_1 l_1 \| A^{E,M}_J \| n_0 j_0 l_0\rangle|^2}{(2j_0+1)(2J+1)} \tag{11}$$

is the absorption transition rate for the process $n_0 j_0 l_0 \to n_1 j_1 l_1$ and

$$W^{em(J)}_{n_0 j_0 l_0, n_1 j_1 l_1} = \frac{|\langle n_0 j_0 l_0 \| A^{E,M}_J \| n_1 j_1 l_1\rangle|^2}{2j_1+1} \tag{12}$$

is the emission transition rate for the process $n_1 j_1 l_1 \to n_0 j_0 l_0$.

It follows from expression (11) that the process of the atomic level excitation, as described above, demonstrates the standard resonant behavior, i.e., the frequency distribution of the absorbed photons is determined by the Lorentz profile. The atomic level emission, also described by the same Lorentz contour [Eq. (12)], is fully independent of the excitation process; the latter can be even of another nature (electron impact excitation, etc.). In principle, there are NR corrections to this process, which restore this dependence; these corrections arise from the terms of the expansion over intermediate states in Eq. (1) with $njl \neq n_1 j_1 l_1$. However we have to remember that collisional and other processes, which we supposed to intervene between the absorption of the photon and its subsequent emission, fully distort and screen the natural line profile for the emitted photons. Therefore there is no sense to discuss the role of NR corrections in this case. The systematic corrections to the resonance frequency determination from the emission spectrum are defined now by other factors which in case of $1s$-$2s$ resonance were in every detail analyzed in [1]. Still it makes sense to investigate the experiment for the measurement of $1s$-$2s$ transition frequency [1], idealizing the experimental situation and assuming that the natural line profile can be observed. Then the experiment for the measurement of $1s$-$2s$ transition frequency will become of the resonant photon scattering type with a complicated scattering scheme: absorption of two photons followed by the one-photon delayed decay in an external electric field. We will investigate the NR corrections in this idealized experiment in Secs. VI and VII.

For the investigation of the resonant photon scattering on the atomic electron, i.e., the resonant process of the second type, discussed in Sec. I, we can start with expression (7). Now no assumption is made concerning summation over m in Eq. (7); this summation is exactly performed. The expression for the transition probability in the resonance approximation now looks like

$$dW^{(J)}_{n_0j_0l_0,n_0j_0l_0}(n_1j_1l_1) = \frac{1}{(2J+1)(2j_0+1)}\sum_{M_iM_f}\sum_{m_im_f}\sum_{m,m'}\langle n_0j_0l_0m_f|A^{(E,M)^\dagger}_{JM_f}|n_1j_1l_1m\rangle \times\langle n_1j_1l_1m|A^{(E,M)}_{JM_i}|n_0j_0l_0m_i\rangle\frac{\langle n_0j_0l_0m_f|A^{(E,M)^\dagger}_{JM_f}|n_1j_1m'\rangle^*\langle n_1j_1m'|A^{(E,M)}_{JM_i}|n_0j_0l_0m_i\rangle^*}{(E_{n_1j_1l_1}-E_{n_0j_0l_0}-\omega)^2+\frac{1}{4}\Gamma^2_{n_1j_1l_1}}d\omega. \qquad (13)$$

Using again the Wigner-Eckart theorem [Eq. (9)] and performing the summation over the angular momentum projections in Eq. (13) we arrive again at Eq. (10). To see how it happens it is useful to perform first the summation over m_i, M_i and m_f, M_f in Eq. (13). Thus there is no difference between the processes of the first and the second types in the total probabilities (cross sections) and the statement made in [10] that both types of the process differ in the total cross sections is incorrect. However, this difference arises in the differential (with respect to the angles) cross sections. The NR correction estimates, given in [10], are valid for the differential cross sections as well as general derivation scheme with the use of the QED with different in and out Hamiltonians. Therefore, we now go over to the description of the differential cross sections.

III. RESONANCE PHOTON SCATTERING ON THE ATOMIC ELECTRON: DIFFERENTIAL CROSS SECTIONS

To write down the differential cross sections we have to introduce first the wave functions for the incoming and outcoming photons. We characterize these photons by their momentum $\vec{k}=\omega\vec{\nu}$ ($\omega=|\vec{k}|$ is the frequency) and the polarization vector $\vec{e}$,

$$\vec{A}_{\vec{k}\cdot\vec{e}}(\vec{r}) = \sqrt{\frac{2\pi}{\omega}}\vec{e}e^{i\vec{k}\cdot\vec{r}}. \qquad (14)$$

Function (14) represents the absorbed photon, the emitted photon is described by the wave function $\vec{A}^*_{\vec{k}\cdot\vec{e}}(\vec{r})$. Now we expand plane wave (14) in the spherical waves or what is more convenient in the electric and magnetic photon wave functions,

$$\vec{A}^*_{\vec{k}\cdot\vec{e}}(\vec{r}) = \frac{2\pi}{\omega}\sum_{JMs}[\vec{e}^*\cdot\vec{A}^{(s)}_{JM}(\vec{\nu})]\vec{A}^{(s)}_{JM}(\vec{r}), \qquad (15)$$

where $s=E,M$ and

$$\vec{A}^E_{JM}(\vec{\nu}) = \sqrt{\frac{\omega}{2\pi}}\left[\sqrt{\frac{J}{2J+1}}\vec{Y}_{JJ+1M}(\vec{\nu}) - \sqrt{\frac{J+1}{2J+1}}\vec{Y}_{JJ-1M}(\vec{\nu})\right], \qquad (16)$$

$$\vec{A}^M_{JM}(\vec{\nu}) = \sqrt{\frac{\omega}{2\pi}}\vec{Y}_{JJM}(\vec{\nu}) \qquad (17)$$

are the electric and magnetic photon wave functions in the momentum space. Instead of Eq. (7) we have the amplitude

$$U^{\vec{\nu}_f\vec{e}_f,\vec{\nu}_i\vec{e}_i}_{n_0j_0l_0m_f,n_0j_0l_0m_i} = \sum_m\frac{\langle n_0j_0l_0m_f|A^{\vec{\nu}_f\vec{e}_f^\dagger}|n_1j_1l_1m\rangle\langle n_1j_1l_1m|A^{\vec{\nu}_i\vec{e}_i}|n_0j_0l_0m_i\rangle}{E_{n_1j_1l_1}-E_{n_0j_0l_0}-\omega}, \qquad (18)$$

where

$$A^{\vec{\nu}_i,\vec{e}_i}(\vec{r}) = e\vec{\gamma}\cdot\vec{A}_{\vec{k}_i,\vec{e}_i}(\vec{r}). \qquad (19)$$

An expression, which replaces now Eq. (8), is

$$dW^{\vec{\nu}_f\vec{\nu}_i}_{n_0j_0l_0,n_0j_0l_0} = \frac{1}{(2j_0+1)}(2\pi^3)\sum_{J'_fM'_fs'_f}\sum_{J_fM_fs_f}\sum_{J'_iM'_is'_i}\sum_{J_iM_is_i}\sum_{\vec{e}_i\vec{e}_f}[\vec{e}^{\,*}_f\cdot\vec{A}^{(s_f)}_{J_fM_f}(\vec{\nu}_f)][\vec{e}_f\cdot\vec{A}^{(s'_f)}_{J'_fM'_f}(\vec{\nu}_f)]$$
$$\times[\vec{e}_i\cdot\vec{A}^{(s_i)}_{J_iM_i}(\vec{\nu}_i)][\vec{e}^{\,*}_i\cdot\vec{A}^{(s'_i)}_{J'_iM''_i}(\vec{\nu}_i)]\sum_{m_im_f}\sum_{m,m'}\langle n_0j_0l_0m_f|e\vec{\gamma}\cdot\vec{A}^{(s_f)^*}_{J_fM_f}(\vec{r})|n_1j_1l_1m\rangle\langle n_0j_0l_0m_f|e\vec{\gamma}\cdot\vec{A}^{(s'_f)^*}_{J'_fM'_f}(\vec{r})|n_1j_1l_1m'\rangle^*$$
$$\times\langle n_1j_1l_1m|e\vec{\gamma}\cdot\vec{A}^{(s_i)}_{J_iM_i}(\vec{r})|n_0j_0l_0m_i\rangle\langle n_1j_1l_1m'|e\vec{\gamma}\cdot\vec{A}^{(s'_i)}_{J'_iM'_i}(\vec{r})|n_0j_0l_0m_i\rangle^*\frac{1}{(E_{n_1j_1l_1}-E_{n_0j_0l_0}-\omega)^2+\frac{1}{4}\Gamma^2_{n_1j_1l_1}}d\vec{\nu}_id\vec{\nu}_fd\omega. \tag{20}$$

Essential simplification arises if we assume that only one type of the photon with the fixed J value can be absorbed and emitted in the resonance. Summation over the polarizations we perform with the use of the formula

$$\sum_{\vec{e}}(\vec{e}^{\,*}\cdot\vec{a})(\vec{e}\cdot\vec{b}) = [\vec{a}\times\vec{\nu}][\vec{b}\times\vec{\nu}]. \tag{21}$$

Then

$$\sum_{\vec{e}_f}[\vec{e}^{\,*}_f\cdot\vec{A}^{(s_f)}_{J_fM_f}(\vec{\nu}_f)][\vec{e}_f\cdot\vec{A}^{(s'_f)}_{J'_fM'_f}(\vec{\nu}_f)] = [\vec{\nu}_f\times\vec{A}^{(s_f)}][\vec{\nu}_f\times\vec{A}^{(s'_f)}], \tag{22}$$

$$\sum_{\vec{e}_i}[\vec{e}^{\,*}_i\cdot\vec{A}^{(s_i)}_{J_iM_i}(\vec{\nu}_i)][\vec{e}_i\cdot\vec{A}^{(s'_i)}_{J'_iM'_i}(\vec{\nu}_i)] = [\vec{\nu}_i\times\vec{A}^{(s_i)}][\vec{\nu}_i\times\vec{A}^{(s'_i)}]. \tag{23}$$

The vector products $[\vec{\nu}\times\vec{A}^{(E,M)}_{JM}(\nu)]$ can be presented like

$$[\vec{\nu}\times\vec{A}^{(E,M)}_{JM}(\nu)] = \vec{A}^{(M,E)}_{JM}(\nu). \tag{24}$$

Then Eq. (20) results in the following:

$$dW^{\vec{\nu}_f\vec{\nu}_i}_{n_0j_0l_0,n_0j_0l_0} = \frac{1}{(2j_0+1)}(2\pi^3)\sum_{M_fM'_f}\sum_{M_iM'_i}\sum_{s}[\vec{A}^{(s)}_{JM_f}(\vec{\nu}_f)\cdot\vec{A}^{(s)^*}_{JM'_f}(\vec{\nu}_f)][\vec{A}^{(s)}_{JM_i}(\vec{\nu}_i)\cdot\vec{A}^{(s)^*}_{JM'_i}(\vec{\nu}_i)]$$
$$\times\sum_{m_im_f}\sum_{m,m'}\langle n_0j_0l_0m_f|A^{(s)}_{JM_f}(\vec{r})|n_1j_1l_1m\rangle\langle n_0j_0l_0m_f|A^{(s)}_{JM'_f}(\vec{r})|n_1j_1l_1m'\rangle^*$$
$$\times\langle n_1j_1l_1m|A^{(s)}_{JM_i}(\vec{r})|n_0j_0l_0m_i\rangle\langle n_1j_1l_1m'|A^{(s)}_{JM'_i}(\vec{r})|n_0j_0l_0m_i\rangle^*\frac{1}{(E_{n_1j_1l_1}-E_{n_0j_0l_0}-\omega)^2+\frac{1}{4}\Gamma^2_{n_1j_1l_1}}d\vec{\nu}_id\vec{\nu}_fd\omega. \tag{25}$$

where we have used again notation (2).

As the next step we employ once more Wigner-Eckart theorem (9) and perform the summations over m_i,m_f,m,m' in Eq. (25). This results in the following:

$$dW^{\vec{\nu}_f\vec{\nu}_i}_{n_0j_0l_0,n_0j_0l_0} = \frac{2j_1+1}{(2J+1)}(2\pi^3)\sum_{M_fM_i}|(\vec{A}^{(E,M)}_{JM_f}(\vec{\nu}_f)\;|^2|(\vec{A}^{(E,M)}_{JM_i}(\vec{\nu}_i)\;|^2$$
$$\times W^{em(J)}_{n_0j_0l_0,n_1j_1l_1}W^{ab(J)}_{n_1j_1l_1,n_0j_0l_0}\frac{1}{(E_{n_1j_1l_1}-E_{n_0j_0l_0}-\omega)^2+\frac{1}{4}\Gamma^2_{n_1j_1l_1}}d\vec{\nu}_id\vec{\nu}_fd\omega. \tag{26}$$

The sum over $s=E,M$ is absent in Eq. (26) since the value of s is fixed by the choice of l_0,l_1. Since the angular dependence on the angles $\vec{\nu}_i$ and $\vec{\nu}_f$ is separated in Eq. (26), both absorption and emission differential probabilities do not depend on the angles and we return to formula (10) (see the details of calculations in Appendix A).

Now we go over to the description of the second type of the experiment when the differential (with respect to the angles) cross section is evaluated. Equation (25) should be replaced by

$$dW^{\vec{\nu}_f\vec{\nu}_i}_{n_0j_0l_0,n_0j_0l_0} = \frac{1}{(2j_0+1)}(2\pi^3)\sum_{M_fM'_f}\sum_{M_iM'_i}[\vec{A}^{(E,M)}_{JM_f}(\vec{\nu}_f)\cdot\vec{A}^{(E,M)^*}_{JM'_f}(\vec{\nu}_f)][\vec{A}^{(E,M)}_{JM_i}(\vec{\nu}_i)\cdot\vec{A}^{(E,M)^*}_{JM'_i}(\vec{\nu}_i)]$$
$$\times\sum_{m_im_f}\sum_{m,m'}\langle n_0j_0l_0m_f|A^{(E,M)}_{JM_f}(\vec{r})|n_1j_1l_1m\rangle\langle n_0j_0l_0m_f|A^{(E,M)}_{JM'_f}(\vec{r})|n_1j_1l_1m'\rangle^*\times\langle n_1j_1l_1m|A^{(E,M)}_{JM_i}(\vec{r})|n_0j_0l_0m_i\rangle$$
$$\times\langle n_1j_1l_1m'|A^{(E,M)}_{JM'_i}(\vec{r})|n_0j_0l_0m_i\rangle^*\frac{1}{(E_{n_1j_1l_1}-E_{n_0j_0l_0}-\omega)^2+\frac{1}{4}\Gamma^2_{n_1j_1l_1}}d\vec{\nu}_id\vec{\nu}_fd\omega. \tag{27}$$

Application of the Wigner-Eckart theorem to the matrix element in Eq. (2) and the subsequent summation over indices m_i, m_f, m, m' yields

$$dW^{\vec{\nu}_f\vec{\nu}_i}_{n_0j_0l_0,n_0j_0l_0} = \frac{1}{(2j_0+1)}(2\pi^3)\sum_{M_fM'_f}\sum_{M_iM'_i}[\vec{A}^{(E,M)}_{JM_f}(\vec{\nu}_f)\cdot\vec{A}^{(E,M)^*}_{JM'_f}(\vec{\nu}_f)][\vec{A}^{(E,M)}_{JM_i}(\vec{\nu}_i)\cdot\vec{A}^{(E,M)^*}_{JM'_i}(\vec{\nu}_i)]\times\sum_{x\xi}(-1)^{\xi}(2x+1)\begin{pmatrix} J & J & x \\ \bar{M}_f & M'_f & \xi \end{pmatrix}$$
$$\times\begin{pmatrix} J & J & x \\ \bar{M}_i & M'_i & \xi \end{pmatrix}\begin{Bmatrix} J & x & J \\ j_1 & j_0 & j_1 \end{Bmatrix}^2\times W^{em(J)}_{n_0j_0l_0,n_1j_1l_1}W^{ab(J)}_{n_1j_1l_1,n_0j_0l_0}\frac{1}{(E_{n_1j_1l_1}-E_{n_0j_0l_0}-\omega)^2+\frac{1}{4}\Gamma^2_{n_1j_1l_1}}d\vec{\nu}_id\vec{\nu}_fd\omega. \tag{28}$$

Equation (28) corresponds to the photon scattering cross section (probability) for the process of the second type: the resonance photon scattering on the atomic electron. Namely, Eq. (28) describes the contribution of the resonant term to the differential cross section. Due to the summation over $x\xi$ in Eq. (28), unlike Eq. (26) the connection holds between the angles $\vec{\nu}_i$ and $\vec{\nu}_f$ for the incoming and outcoming photons. Finally, we can write down expression (29) for the transition probability in the form

$$dW^{\vec{\nu}_f\vec{\nu}_i}_{n_0j_0l_0,n_0j_0l_0} = S_{Jj_0j_1}(\vec{\nu}_f\vec{\nu}_i)\frac{W^{em(J)}_{n_0j_0l_0,n_1j_1l_1}W^{ab(J)}_{n_1j_1l_1,n_0j_0l_0}}{(E_{n_1j_1l_1}-E_{n_0j_0l_0}-\omega)^2+\frac{1}{4}\Gamma^2_{n_1j_1l_1}}d\vec{\nu}_id\vec{\nu}_fd\omega, \tag{29}$$

where

$$S_{Jj_0j_1}(\vec{\nu}_f\vec{\nu}_i) = \frac{1}{(2j_0+1)}\sum_{M_fM'_f}\sum_{M_iM'_i}[\vec{A}^{(E,M)}_{JM_f}(\vec{\nu}_f)\cdot\vec{A}^{(E,M)^*}_{JM'_f}(\vec{\nu}_f)][\vec{A}^{(E,M)}_{JM_i}(\vec{\nu}_i)\cdot\vec{A}^{(E,M)^*}_{JM'_i}(\vec{\nu}_i)]$$
$$\times\sum_{x\xi}(-1)^{\xi}(2x+1)\begin{pmatrix} J & J & x \\ \bar{M}_f & M'_f & \xi \end{pmatrix}\begin{pmatrix} J & J & x \\ \bar{M}_i & M'_i & \xi \end{pmatrix}\begin{Bmatrix} J & x & J \\ j_1 & j_0 & j_1 \end{Bmatrix}^2. \tag{30}$$

The only angular dependence that can arise in the resonant photon experiment in the absence of external electric fields is the dependence on the $\angle\vec{\nu}_i\vec{\nu}_f$ between the direction $\vec{\nu}_i$ of the velocity of the absorbed photon and the direction $\vec{\nu}_f$ of the velocity of the emitted photon. While the vector $\vec{\nu}_i$ is fixed by the incident laser beam, the vector $\vec{\nu}_f$ is fixed by the position of the detector. In the experiment of the first type no angular dependence can arise since the integration over angles is performed independently for the absorption and the emission rate, respectively. In the processes of the second type the angular dependence can occur.

To obtain the angular dependence in the explicit form we can choose the direction of z axis in the photon momentum space along the vector $\vec{\nu_i}$. Then the vector $\vec{\nu_f}$ will represent the dependence on the $\angle\vec{\nu_i}\vec{\nu_f}$. Expression (30) simplifies and is given in Appendix B. In case of the magnetic dipole photons ($J=1$) the corresponding expression reads

$$S^M_{1j_0j_1}(\Theta) = \frac{\omega^2}{32\pi(2j_0+1)}\begin{Bmatrix} 1 & 0 & 1 \\ j_1 & j_0 & j_1 \end{Bmatrix}^2(a_M+b_M\cos^2\Theta), \tag{31}$$

where

$$a_M = \frac{1}{3}-\frac{15}{2\sqrt{6}}, \tag{32}$$

$$b_M = \frac{45}{2\sqrt{6}}. \tag{33}$$

In case of the electric dipole photons ($J=1$) the corresponding expression reads

$$S^E_{1j_0j_1}(\Theta) = \frac{\omega^2}{32\pi(2j_0+1)}\times\begin{Bmatrix} 1 & 0 & 1 \\ j_1 & j_0 & j_1 \end{Bmatrix}^2(a_E+b_E\cos^2\Theta), \tag{34}$$

where

$$a_E = \frac{1+\sqrt{30}-4\sqrt{10}}{9} + \frac{5}{36\sqrt{30}} - \frac{5(31\sqrt{3}-12)}{12\sqrt{3}}, \qquad (35)$$

$$b_E = \frac{5}{2\sqrt{30}} + \frac{5}{12\sqrt{3}}. \qquad (36)$$

IV. NONRESONANT CORRECTIONS TO THE RESONANCE TRANSITION PROBABILITY

In this section we will derive the NR corrections to the resonance transition probability for the process of resonant photon scattering on the atomic electron. The nonresonant corrections to the resonant process arise as the terms of expansion in Eq. (1) with $njl = n_2 j_2 l_2 \neq n_1 j_1 l_1$. In the expression for the transition probability the major NR contribution will arise from the interference between the amplitudes corresponding to $n_2 j_2 l_2$ and $n_1 j_1 l_1$. The contribution from the level $n_2 j_2 l_2$ most close to $n_1 j_1 l_1$ level will dominate. Repeating the derivations which led to the expression for the transition probability [Eq. (29)] and retaining now two terms $n_1 j_1 l_1$ and $n_2 j_2 l_2$ ($n_2 j_2 l_2$ being considered as the closest neighbor to the $n_1 j_1 l_1$) in the sum over njl in Eq. (1) result in

$$dW^{\vec{\nu}_f \vec{\nu}_i}_{n_0 j_0 l_0, n_0 j_0 l_0} = S_{J j_0 j_1}(\vec{\nu}_f \vec{\nu}_i) \frac{W^{em(J)}_{n_0 j_0 l_0, n_1 j_1 l_1} W^{ab(J)}_{n_1 j_1 l_1, n_0 j_0 l_0}}{(E_{n_1 j_1 l_1} - E_{n_0 j_0 l_0} - \omega)^2 + \frac{1}{4}\Gamma^2_{n_1 j_1 l_1}} d\vec{\nu}_i d\vec{\nu}_f d\omega + S_{J j_0 j_1 j_2}(\vec{\nu}_f \vec{\nu}_i)$$
$$\times 2\,\mathrm{Re}\left(\frac{\langle n_0 j_0 l_0 \| A^{E,M}_J \| n_1 j_1 l_1 \rangle \langle n_0 j_0 l_0 \| A^{E,M}_J \| n_2 j_2 l_2 \rangle \langle n_1 j_1 l_1 \| A^{E,M}_J \| n_0 j_0 l_0 \rangle \langle n_2 j_2 l_2 \| A^{E,M}_J \| n_0 j_0 l_0 \rangle}{\left(E_{n_1 j_1 l_1} - E_{n_0 j_0 l_0} - \omega - \frac{i}{2}\Gamma_{n_1 j_1 l_1}\right)\left(E_{n_2 j_2 l_2} - E_{n_0 j_0 l_0} - \omega - \frac{i}{2}\Gamma_{n_2 j_2 l_2}\right)} \right) d\vec{\nu}_i d\vec{\nu}_f d\omega, \qquad (37)$$

where

$$S_{J j_0 j_1 j_2}(\vec{\nu}_f \vec{\nu}_i) = \frac{1}{(2j_0+1)}(2\pi^3) \sum_{M_f M_f'} \sum_{M_i M_i'} [\vec{A}^{(E,M)}_{JM_f}$$
$$\times (\vec{\nu}_f) \cdot \vec{A}^{(E,M)^*}_{JM_f'}(\vec{\nu}_f)][\vec{A}^{(E,M)}_{JM_i}(\vec{\nu}_i) \cdot \vec{A}^{(E,M)^*}_{JM_i'}(\vec{\nu}_i)]$$
$$\times \sum_{x\xi} (-1)^{\xi}(2x+1) \begin{pmatrix} J & J & x \\ \bar{M}_f & M_f' & \xi \end{pmatrix}$$
$$\times \begin{pmatrix} J & J & x \\ \bar{M}_i & M_i' & \xi \end{pmatrix} \begin{Bmatrix} J & x & J \\ j_1 & j_0 & j_2 \end{Bmatrix}^2 . \qquad (38)$$

It is important that the factor $S_{J j_0 j_1 j_2}(\vec{\nu}_f \vec{\nu}_i)$ does not vanish for $j_2 l_2 \neq j_1 l_1$, i.e., for the levels with different symmetry properties (different total angular momenta or different parities). Such terms, as a rule, give the major contribution to the NR corrections. In [5] only NR corrections from the terms $n_2 j_2 l_2$ with $j_2 l_2 = j_1 l_1$ were considered.

Expression (38) again can be simplified if we choose the direction of z axis in photon momentum space along the vector $\vec{\nu}_i$. The corresponding expression is given in Appendix B. In case of the magnetic dipole photon ($J=1$) this expression reads

$$S^M_{1 j_0 j_1 j_2}(\Theta) = \frac{\omega^2}{32\pi(2j_0+1)} \begin{Bmatrix} 1 & 0 & 1 \\ j_1 & j_0 & j_2 \end{Bmatrix}^2 (a_M + b_M \cos^2\Theta). \qquad (39)$$

In case of the electric dipole photons ($J=1$) the corresponding expression reads

$$S^E_{1 j_0 j_1 j_2}(\Theta) = \frac{\omega^2}{32\pi(2j_0+1)} \begin{Bmatrix} 1 & 0 & 1 \\ j_1 & j_0 & j_2 \end{Bmatrix}^2 (a_E + b_E \cos^2\Theta). \qquad (40)$$

Note that factors $a_{E,M}, b_{E,M}$ in Eqs. (31) and (34) coincide with corresponding factors $a_{E,M}, b_{E,M}$ in Eqs. (39) and (40).

V. QED THEORY WITH DIFFERENT IN AND OUT HAMILTONIANS

Aiming at the accuracy estimates for the 1s-2s resonance experiment in hydrogen [1] we will first introduce necessary generalization of QED, mentioned in Sec. I, i.e., we will present the formulation of QED with different in and out Hamiltonians. In this formulation we will follow Fradkin-Gitman-Shvartsman book [8] (FGS formalism). In principle, Fradkin *et al.* [8] pursued the more complicated task; then one considered here, namely, the case when the external electric field creates the electron-positron pairs. Accordingly, we shall describe this approach in a simplified version, sufficient for our purposes here: the presence of weak electric field that enables the more efficient one-photon decay of 2s level in hydrogen. However due to the admixture of 2p state to 2s state the emission probability changes by 7 orders of magnitude. We will consider the change in the other atomic characteristics (Stark effect of atomic levels and Stark splitting) negligible. The criterion of the weak field will be

$\varepsilon \ll \varepsilon_c = 475$ V/cm, where ε is the strength of the electric field. In the field $\varepsilon = \varepsilon_c$ the Stark matrix element is equal to the Lamb shift and the $2s$ and $2p$ levels are strongly mixed [11].

The applicability of the FGS picture is defined by the two inequalities: $\tau_{at} \ll \tau_{field} \ll \tau_d$. Here τ_{field} is the time showing how fast the field is changing in the rest frame of atom, τ_{at} is the characteristic atomic time necessary for the formation of the stationary atomic states, and τ_d is the atomic decay time. The inequality $\tau_{at} \ll \tau_{field}$ means that the field is changing slowly enough not to destroy the stationary atomic states. The other inequality $\tau_{field} \ll \tau_d$ implies that the field is changing quite abruptly in space (and, corresponding, in time in the rest frame of the atom) so that the detection signal has a sharp peak structure which allows for the accurate determinations of the frequency. Evidently, the second inequality $\tau_{field} \ll \tau_d$ is satisfied in the $1s$-$2s$ experiments [1,2]. To estimate τ_{at} we have to recall that for the formation of the atomic states the electron has to perform at least several revolutions around the nucleus, the time of one revolution being of the order 10^{-16} s. For the formation of the distinct $2s$, $2p$ levels split by the energy interval (Lamb shift) 1000 MHz, the atomic time $\tau_{at} \approx 10^{-9}$ s will be necessary. The decay time for the $2p$ level is about 10^{-9} s; the same decay time acquires the $2s$ level in the critical field ε_c. Assuming that the "weak field" in the experiments [1,2] is by 1 order of the magnitude smaller than ε_c and remembering that admixture of the $2s$ state in the external field ε is proportional to ε^2 we obtain $\tau_d \approx 10^{-7}$ s. Thus the both inequalities mentioned above are compatible and the applicability of the FGS picture is a plausible assumption.

Another assumption concerns the duration of the excitation time, i.e., the time interval when laser, exciting $2s$ level, is switched on. This time should not be less than the lifetime of the $2s$ level in the absence of the field, i.e., 1/7 s. Then the line profile is not influenced by the excitation time.

The FGS theory operates with two complete sets of eigenfunctions, belonging to in and out Hamiltonians. For the experimental situation in [1] the in states correspond to the solution of the Dirac (Schrödinger) equation in the field of the nucleus. The nonrelativistic approximation utilizing Schrödinger wave function will be sufficient for our purposes. The out set will correspond to the perturbed wave function in the presence of an electric field; according to the experimental conditions in [1] this field is assumed to be weak.

The advantage of the FGS approach is that it employs the standard QED methods in a generalized form: S matrix, field operators in the Fock space, four-dimensional perturbation theory, and Feynman graph techniques in the Furry picture. Although inspired by the generic lines of FGS theory, a simplified version of Feynman graph techniques, more close to the conventional bound-state QED, will be employed here.

Actually the only new element which needs to be used is the generalized electron propagator, connecting vertices which refer to the different in and out spaces. The eigenmode decomposition of this propagator reads

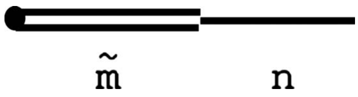

FIG. 2. The diagrammatic representation of the FGS electron propagator [Eq. (33)] in the coordinate space. See explanations in the text.

$$S^{\mathrm{FGS}}(x_1,x_2) = \theta(t_1-t_2)\sum_{\tilde m,n}^{E_{\tilde m},E_n>0}\Psi_{\tilde m}(x_1)\omega_{\tilde m n}\bar\Psi_n(x_2) - \theta(t_2-t_1)\sum_{\tilde m,n}^{E_{\tilde m},E_n<0}\Psi_n(x_1)\omega_{\tilde m n}\bar\Psi_{\tilde m}(x_2), \qquad (41)$$

where $\Psi_{\tilde m}(x)$ are the solutions of the Dirac equation for the electron in the field of the nucleus and external electric field, $\Psi_n(x)$ are the solutions with zero external field, and $E_{\tilde m}, E_n$ are corresponding eigenvalues. Within the nonrelativistic theory, sufficient for the description of the process in the neutral hydrogen atom, in Eq. (41) the contribution due to the negative energy states can be neglected.

The matrix $\omega_{\tilde m n}$ is defined as [8]

$$\omega_{\tilde m n} = \langle \cdots \tilde m \cdots | \hat{\tilde a}^{\dagger} \hat a | \cdots n \cdots \rangle, \qquad (42)$$

where $\langle \cdots \tilde m \cdots |$ denotes the state vector of the out-Fock-space with an electron in the state $\tilde m$, $| \cdots n \cdots \rangle$ denotes the state vector of the in-Fock-space with an electron in the state n, $\hat{\tilde a}^{\dagger}$ is the creation operator in the out-Fock-space, and $\hat a$ is the annihilation operator in the in-Fock-space. In the simple case, when no particles are created by the electric field, the matrix $\omega_{\tilde m n}$ is reduced to the overlap integral,

$$\omega_{\tilde m n} = \int \Psi^{\dagger}_{\tilde m}(\vec x)\Psi_n(\vec x)d\vec x \equiv \langle \tilde m | n \rangle. \qquad (43)$$

In what follows the conventional Feynman diagrams involving the propagator S^{FGS}, defined by Eq. (41), will be used. The ordinary solid line will represent an electron in the field of nucleus only, while a double solid line will describe an electron propagator in the field of the nucleus together with an additional external electric field. We will denote the vertex, corresponding to the out Hamiltonian, as a fat dot, and the vertex, corresponding to the in Hamiltonian, as an ordinary dot. The new graphic element, corresponding to the propagator S^{FGS}, is depicted in Fig. 2.

An important question is the evaluation of the transition probabilities within FGS theory. The problem arise how to connect the matrix element of the S matrix with the transition rate (or cross section) of the process. In the traditional QED this problem is solved in the following way: due to the energy conservation law the S-matrix element between the initial (i) and final (f) states can be always presented as

$$S_{if} = 2\pi i \delta(E_i - E_f) U_{if}, \qquad (44)$$

where E_i, E_f are the energies of the initial (final) states and U_{if} is the amplitude of the process. The squared δ function $\delta(E_i - E_f)$ is presented like

$$\delta(E_i - E_f)\frac{1}{2\pi}\int_{-T/2}^{T/2} e^{i(E_i-E_f)t}dt = \delta(E_i - E_f)\frac{T}{2\pi}, \tag{45}$$

where T can be interpreted as the observation time. Then the transition rate (probability per time unit) can be defined as

$$dW_{if} = \frac{|S_{if}|^2}{T} = 2\pi\delta(E_i - E_f)|U_{if}|^2 d\omega. \tag{46}$$

Within FGS theory the energy is not conserved and definition (44) for the transition probability fails, in principle.

However, in the case of the weak field, assuming that the Stark shifts are neglected, the energy conservation actually remains. Then we can keep standard formula (44) for the evaluation of transition probabilities using however the wave functions, perturbed by the electric field.

VI. TWO-PHOTON 1*s*-2*s* RESONANCE WITH DELAYED DECAY REGISTRATION IN AN EXTERNAL ELECTRIC FIELD

In this section we will apply the formalism described in Sec. V to the derivation of NR correction to the resonance 1*s*-2*s* transition in the idealized experiment [1], i.e., in the process of resonant two-photon excitation of 2*s* level in hydrogen with the subsequent delayed decay in an external electric field. In the experiment [1] excited by the two-photon absorption hydrogen atom in 2*s* state left the excitation region (in space) and entered the decay region (out space), where the weak electric field was present. The decay occurred predominantly via the fast 2*p*-1*s* transition (transition rate $\sim 10^9\ \mathrm{s}^{-1}$). Thus the lifetime of the excited 2*s* state as defined by the experimental setup was in the millisecond range. This lifetime is much smaller than the natural lifetime for a 2*s* level associated with the two-photon decay ($\sim\frac{1}{7}$ s). Within the reference frame of hydrogen atom this situation corresponds to a time-dependent problem, where the in and out QED formalisms involving different asymptotic Hamiltonians are applicable. Employing the diagrammatic techniques for the FGS formalism introduced in Sec. V, the process of 1*s*-2*s* excitation, described above, is represented by the Feynman graph depicted in Fig. 3.

Writing down the *S*-matrix element, corresponding to Fig. 3, we find

$$S_{\tilde{a}a} = (-i)^3\int \bar{\Psi}_{\tilde{a}}(x_1)\gamma^{\mu 1}A_{\mu 1}^{(JLM)*}(x_1)S^{\mathrm{FGS}}(x_1x_2)\gamma^{\mu 2}A_{\mu 2}^{(J'L'M')}(x_2)S(x_2x_3)\gamma^{\mu 3}A_{\mu 3}^{(J'L'M'')}(x_3)\Psi_a(x_3)\, d^4x_1d^4x_2d^4x_3. \tag{47}$$

Here $\Psi_a(x)$ is the initial electron wave function (in the in space), $\bar{\Psi}_{\tilde{a}}(x)$ is the Dirac-conjugated final state wave function (in the out space), γ^μ are the standard Dirac matrices, $A_\mu^{(JLM)}$ is the photon wave function with quantum numbers JLM [12], $S(x_1x_2)$ is the ordinary electron propagator in the Furry picture, and S^{FGS} is the propagator defined by Eq. (41).

The integration over the time variables in Eq. (47) in the nonrelativistic limit and with neglect of Stark shifts of the levels in an external electric field yields

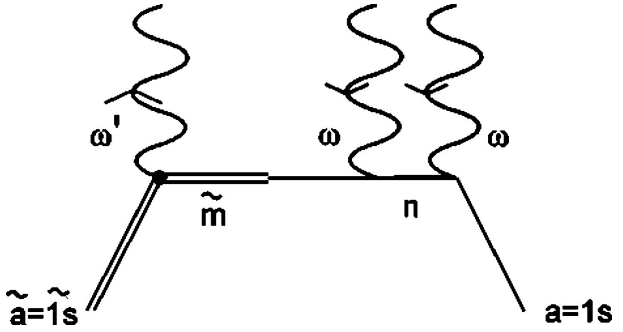

FIG. 3. The process of the two-photon excitation 1*s*-2*s* followed by the delayed decay in an external electric field. The picture is understood with respect to the frame of reference of the hydrogen atom. The ordinary external and internal solid lines describe the electron wave function and electron propagator for the bound electron in the absence of external electric field. The double internal electron line represents the FGS electron propagator as described in Fig. 2. The external double solid line corresponds to the electron wave function in the external electric field. The two absorbed photons are the laser photons with frequencies $\omega=\frac{1}{2}(E_{2s}-E_{1s})$, where E_i are the energies of the atomic electron states in the absence of the electric field. The notations $n,\tilde{n}$ for the wave functions were introduced in Sec. V.

$$S_{\tilde{a}a} = 2\pi i\delta(\omega - 2\omega')\sum_{nn_1}\langle\tilde{a}|A_{JLM}^{\dagger}|\tilde{n}\rangle\langle\tilde{n}|n\rangle \times \frac{\langle n|A_{J'L'M'}|n_1\rangle\langle n_1|A_{J'L'M''}|a\rangle}{(E_n - E_a - 2\omega')(E_{n_1} - E_a - \omega')}. \tag{48}$$

In Eq. (48) we omitted the sum over $\tilde{m}$ in the FGS propagator [Eq. (41)] and kept only the term $m=\tilde{n}$ having in mind that $\tilde{n}\rightarrow n$ when the electric field is switched off. Keeping the other terms $\tilde{m}\neq\tilde{n}$ would mean the energy nonconservation and would contradict to our basic assumption about the energy conservation in the weak field limit. More accurately, $m=\tilde{n}$ means that we keep summation over the degenerate substrates; in case of hydrogen for $n=2s,2p$ we keep $m=\tilde{n}=\widetilde{2s},\widetilde{2p}$.

In Eq. (48) the last two matrix elements in the numerator together with the last energy denominator after the summation over intermediate states n_1 represent the ordinary two-photon absorption amplitude, corresponding to the transition from the state $|a\rangle$ to the state $|n\rangle$ in the absence of the electric field. The resonance condition, as it was realized in the experiment [1], is

$$2\omega' = E_{a'} - E_a, \tag{49}$$

where $a=1s, a'=2s$. Then, setting in Eq. (48) $n=a', \tilde{n}=\tilde{a}$, using Eq. (46), integrating over ω, and summing over angular momentum projections, except the initial M_i, M_i' and final M_f photon projections in the resonance approximation we receive the following expression for the differential probability of the process:

$$dW_{\tilde{a}a}(\omega') = \frac{1}{2\pi} S^{2\gamma}_{aa'J}(\vec{\nu}_f \vec{\nu}_i' \vec{\nu}_i) \times \frac{W^{em(JL)}_{\tilde{a}\tilde{a}'} |\langle \tilde{a}' | a' \rangle|^2 W^{ab(JL,J'L')}_{a'a}}{(E_{a'} - E_a - 2\omega')^2 + \frac{1}{4}\Gamma^2_{a'}} d\vec{\nu}_f d\vec{\nu}_i' d\vec{\nu}_i d\omega', \qquad (50)$$

where $W^{ab(JL,J'L')}_{a'a}$ is the two-photon absorption probability and $S^{2\gamma}_{aa'J}(\vec{\nu}_f \vec{\nu}_i' \vec{\nu}_i)$ is the angular factor, similar to the factor in Eq. (30) in case of one-photon scattering.

In case of the Doppler-free spectroscopy employed in [1], $\vec{\nu}_i' = -\vec{\nu}_i$. Taking into account the NR correction due to the closest to a' neighbor level a'' results in the expression similar to Eq. (37),

$$dW_{\tilde{a}a}(\omega') = \frac{1}{2\pi} S^{2\gamma}_{aa'J}(\vec{\nu}_f \vec{\nu}_i' \vec{\nu}_i) \frac{W^{em(JL)}_{\tilde{a}\tilde{a}'} |\langle \tilde{a}' | a' \rangle|^2 W^{ab(JL,J'L')}_{a'a}}{(E_{a'} - E_a - 2\omega')^2 + \frac{1}{4}\Gamma^2_{a'}} d\vec{\nu}_f d\vec{\nu}_i' d\vec{\nu}_i d\omega' + \frac{1}{2\pi} S^{2\gamma}_{aa'a''J}(\vec{\nu}_f \vec{\nu}_i' \vec{\nu}_i) \times 2\,\mathrm{Re}\left(\frac{\langle \tilde{a} || A^{E,M}_J || \tilde{a}' \rangle \langle \tilde{a} || A^{E,M}_J || \tilde{a}'' \rangle \langle \tilde{a}' | a' \rangle^* \langle \tilde{a}'' | a'' \rangle \langle \tilde{a}' || A^{ab}_{J'L',J'L'} || a \rangle \langle a'' || A^{ab}_{J'L',J'L'} || a \rangle}{\left(E_{a'} - E_a - 2\omega' - \frac{i}{2}\Gamma_{a'}\right)\left(E_{a''} - E_a - 2\omega' - \frac{i}{2}\Gamma_{a''}\right)} \right) d\vec{\nu}_f d\vec{\nu}_i' d\vec{\nu}_i d\omega'. \qquad (51)$$

Here $\langle \tilde{a}' || A^{ab}_{J'L',J'L'} || a \rangle, \langle a'' || A^{ab}_{J'L',J'L'} || a \rangle$ are the angular reduced two-photon absorption amplitudes and $S^{2\gamma}_{aa'a''J}(\vec{\nu}_f \vec{\nu}_i' \vec{\nu}_i)$ is the angular factor, similar to the factor in Eq. (38) in case of one-photon scattering.

VII. NR CORRECTIONS FOR THE FREQUENCY MEASUREMENTS IN 1*s*-2*s* RESONANT HYDROGEN EXPERIMENT

For moving further we have to choose the procedure for the determination of the resonance photon frequency. In [5,6] the evaluation of the maximum value of the frequency distribution was used for this purpose. As it was shown in [6], any other procedure (e.g., finding a "center of gravity" for a line profile) would give the result close to the choice formulated above. In case of the Lorentz profile all the methods of defining ω^{res} give the same result,

$$\omega_0^{res} = E_{a'} - Ea.$$

With our choice of the resonance frequency definition the NR correction will look like

$$\delta\omega^{\mathrm{NR}} = \omega^{\max} - \omega_0^{res}, \qquad (52)$$

where $\omega^{\max}$ corresponds to the position of the maximum of the asymmetrical frequency distribution. In this section we will investigate the two-photon transition in hydrogen [1],

$$1s_{1/2}(F=1) + 2\gamma \to 2s_{1/2}(F=1).$$

Here F is the total angular momentum for an atom, which defines the hyperfine structure sublevel. The value $K=1$ for the total angular momentum K of the two-photon system with equal photon frequencies is strictly forbidden by the Landau theorem [13]. Then the NR correction due to the transition

$$1s_{1/2}(F=1) + 2\gamma \to 2s_{1/2}(F=0)$$

is forbidden because of the Landau theorem and main NR contribution arises from the transition

$$1s_{1/2}(F=1) + 2\gamma \to 2p_{1/2}(F=1).$$

Note that according to Eq. (38) this NR contribution vanishes for the nonpolarized initial photons. In experiments, however, the incident laser light should have nonzero polarization.

For deriving the NR correction we have to use expression (51) where we have to set

$$a = 1s_{1/2}(F=1),$$

$$a' = 2s_{1/2}(F=1),$$

$$a'' = 2p_{1/2}(F=1).$$

In the weak electric field,

$$\Psi_{\tilde{a}} = \Psi_a,$$

$$\Psi_{\tilde{a}'} = \Psi_{a'} + \eta\Psi_{a''},$$

$$\Psi_{\tilde{a}''} = \Psi_{a''} - \eta\Psi_{a'},$$

and the overlap integrals are

$$\langle \tilde{a}' | a' \rangle = \langle \tilde{a}'' | a'' \rangle = 1.$$

Here

$$\eta = \frac{|\Delta E_S|}{\Delta E_L}$$

is the Stark shift to the Lamb shift ratio. This ratio is 1 for the field $\varepsilon = \varepsilon_c$. For deriving the NR correction we set in Eq. (51)

$$E_{a'} - E_a = \omega_0^{res},$$

$$E_{a''}-E_a=E_{a''}-E_{a'}+E_{a'}-E_a=\omega_0^{res}-\Delta E_L,$$

where

$$\Delta E_L=E_{2s_{1/2}}-E_{2p_{1/2}}\approx 10^3\ \text{MHz}$$

is the Lamb shift. Here we assume that the hyperfine splitting for the $2s$, $2p$ levels is much smaller than Lamb shift.

The width $\Gamma_{a'}$ follows from the experimental conditions

$$\Gamma_{a'}=\Gamma_{\text{expt}}\approx 1\ \text{kHz}.$$

The width

$$\Gamma_{a''}=\Gamma_{2p}\approx 100\ \text{MHz}$$

is very large but still can be neglected in the corresponding denominator of Eq. (26) compared to ΔE_L. Insertion of the wave functions $\Psi_{\tilde{a}}$, $\Psi_{\tilde{a}'}$ and $\Psi_{\tilde{a}''}$ in the emission amplitudes in Eqs. (50) and (51) yields

$$\langle\tilde{a}\|A_J^{E,M}\|\tilde{a}'\rangle=\eta(\Gamma_{2p})^{1/2}, \tag{53}$$

$$\langle\tilde{a}\|A_J^{E,M}\|\tilde{a}''\rangle=(\Gamma_{2p})^{1/2}. \tag{54}$$

For the estimates of the NR corrections we omit the angular factors in Eqs. (51) and (52), assuming them to be of the same order of magnitude. Then, performing the evaluation of the maximum value in Eq. (51) with respect to ω' we write

$$\frac{d}{d\omega'}\left\{\frac{\eta^2W^{2\gamma}_{1s,2s_{1/2}}}{(\omega_0^{res}-2\omega')^2+\frac{1}{4}\Gamma^2_{\text{expt}}}+2\,\text{Re}\left(\frac{\eta W^{2\gamma}_{1s,2p_{1/2}}}{\left(\omega_0^{res}-2\omega'-\frac{i}{2}\Gamma_{\text{expt}}\right)(\omega_0^{res}-\triangle E_L-2\omega')}\right)\right\}=0, \tag{55}$$

where $W^{2\gamma}_{1s,2s_{1/2}}$ and $W^{2\gamma}_{1s,2p_{1/2}}$ are the two-photon transition rates for the decay of $2s_{1/2}$ and $2p_{1/2}$ levels, respectively. We should remind that the interference between two channels

$$1s_{1/2}+2\gamma\to 2s_{1/2}$$

and

$$1s\tfrac{1}{2}+2\gamma\to 2p\tfrac{1}{2}$$

can arise in the differential (with respect to the angles) cross section according to Eq. (41).

In case of experiment [1],

$$F_0=F'=F''=1$$

(see Fig. 4). The probability for the two-photon emission $W^{em(2\gamma)}_{1s,2s_{1/2}}$ is very well known. The accurate nonrelativistic value for this transition was obtained in [14],

$$W^{2\gamma(E1E1)}_{1s,2s_{1/2}}=1.32\times 10^{-3}(\alpha Z)^6\ \text{a.u.}$$

It is assumed that $\alpha Z\ll 1$. The probability $W^{em(2\gamma)}_{1s,2s_{1/2}}$ was evaluated recently numerically for all Z values [15], $1\ll Z\ll 100$, and analytically for [16] $\alpha Z\ll 1$. The result is

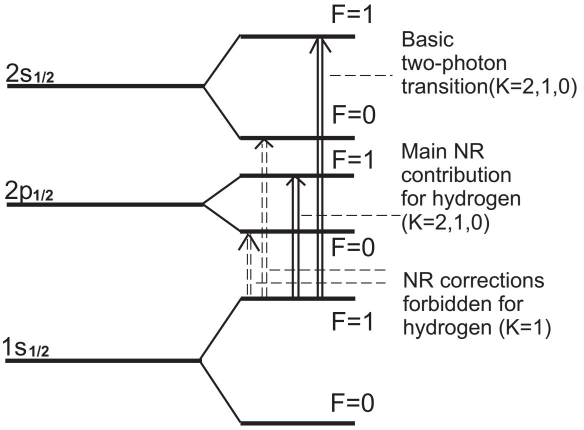

FIG. 4. Scheme of the levels for the two-photon $1s$-$2s$ transition in hydrogen. The vertical double lines denote the two-photon transitions. The K numbers denote the total angular momentum for a two-photon system, possible for the different two-photon transitions.

$$W^{2\gamma}_{1s,2p_{1/2}}=W^{2\gamma(E1M1)}_{1s,2p_{1/2}}+W^{2\gamma(E1E2)}_{1s,2p_{1/2}}$$
$$=2.907\times 10^{-5}(\alpha Z)^8+3.69\times 10^{-6}(\alpha Z)^8\ \text{a.u.}$$

However, for our purposes we need the two-photon absorption probability calculated for the case of two photons of equal frequencies,

$$\omega=\omega'=\tfrac{1}{2}(E_{2s}-E_{1s}).$$

These probabilities can be easily obtained from the results in [15,16] and are equal to

$$W^{ab(2\gamma)}_{1s,2s_{1/2}}=0.91\times 10^{-2}(\alpha Z)^6,$$

$$W^{ab(2\gamma)}_{1s,2p_{1/2}}=W^{ab[2\gamma,(E1M1)]}_{1s,2p_{1/2}}+W^{ab[2\gamma(E1E2)]}_{1s,2p_{1/2}}$$
$$=1.94\times 10^{-4}(\alpha Z)^8+0.887\times 10^{-4}(\alpha Z)^8.$$

Solving Eq. (55) and employing definition (52) yield

$$|\delta\omega^{\text{NR}}|=\frac{1}{4}\frac{\Gamma^2_{\text{expt}}}{\Delta E_L}\left(\frac{W^{ab(2\gamma)}_{1s,2p_{1/2}}}{W^{ab(2\gamma)}_{1s,2s_{1/2}}}\right)^{1/2}\frac{1}{\eta}. \tag{56}$$

Inserting all numbers in Eq. (56) and taking η=0.1 which corresponds to the weak electric field ε=47.5 V/cm, we obtain the final result

$$|\delta\omega^{\text{NR}}|\approx 10^{-5}\ \text{Hz}. \tag{57}$$

This accuracy is still far from the recent experimental inaccuracy estimate of 34 Hz [2]. However as it was stated in [1] the refitment of the experimental techniques will allow one to reach the better accuracy in the future.

In [10] it was incorrectly claimed that the NR correction in case of deuterium can be several orders of magnitude higher than in case of hydrogen due to the different selection rules between the levels with the half-integer F values. In reality these selection rules are the same as for the integer F values for the transitions between the s states in any atoms (i.e., for any isotopes of hydrogen atom, etc.).

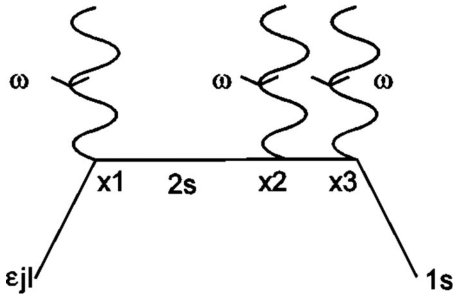

FIG. 5. The Feynman graph corresponding to the process of three-photon ionization of hydrogen atom. Here εjl are the quantum numbers of the electron in continuous spectrum with the energy $\varepsilon = 3\omega - |E_{1s}| = \frac{1}{16}$ a.u.

There exists another idea how to register the $2s$ level excitation in the $1s$-$2s$ experiment [9]. This idea consists of the registration of the $2s$ state ionization with the third laser photon with the same frequency

$$\omega' = \tfrac{1}{2}(E_{2s} - E_{1s}).$$

The energy of the three photons

$$3\omega' = \tfrac{3}{2}(E_{2s} - E_{1s}) = \tfrac{9}{16} \text{ a.u.}$$

is higher than the ionization potential $|E_{1s}| = \frac{1}{2}$ a.u. The registration can be performed via the registration of outcoming electron. The Feynman graph corresponding to this process is depicted in Fig. 5.

This kind of registration means actually the integration over the direction $\vec{\nu}_f$. This eliminates fully the NR corrections connected with the interference terms and makes these corrections fully negligible.

VIII. CONCLUSIONS

First, the results of the measurement begin to depend on the method of extraction of the frequency value from the experimental line profile: the frequency value corresponding to the maximum of the line profile will differ from the value obtained for the center of gravity, etc.

Second, the results will depend also on the method of excitation of atomic level, i.e., the excitation by photons will provide the frequency value different from the value obtained with excitation by electrons.

Third, the method of detection will also influence the measured frequency value. In particular, as it follows from Eq. (44) in case of $1s$-$2s$ excitation in hydrogen, the dependence on both excitation and detection method is present.

Extracting from the experiment the frequency value "invariant" with respect to the different kinds of measurements and characterizing pure atomic property one has to take into account the NR corrections which should be considered as specific systematic corrections in every particular type of experiment.

Moreover, in case of $1s$-$2s$ experiment using another detection scheme (e.g., three-photon ionization) one can fully avoid the necessity to take into account the NR corrections.

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APPENDIX A: THE FORMAL PROOF OF THE ANGULAR INDEPENDENCE IN EQ. (26)

Here we evaluate the scalar products

$$S^{(s)}_{JM}(\vec{\nu}) \equiv |(\vec{A}^{(s)}_{JM} \cdot (\vec{\nu}))|^2 = \sum_{\tau} (-1)^{\tau} [\vec{A}^{(s)}_{JM} \cdot (\vec{\nu})]_{\tau} [\vec{A}^{(s)}_{JM} \cdot (\vec{\nu})]^{*}_{-\tau}. \tag{A1}$$

First we introduce the expression for the spherical components of the spherical photon wave function $\vec{A}^{(s)}_{JM}(\vec{\nu})$,

$$[\vec{A}^{(M)}_{JM}(\vec{\nu})]_{\alpha} = \sum_{m_l \mu} C^{J1}_{JM}(m_l \mu) Y_{Jm_l}(\vec{\nu}) (\vec{\chi}_{\mu})_{\alpha}. \tag{A2}$$

$$[\vec{A}^{(E)}_{JM}(\vec{\nu})]_{\alpha} = -\sqrt{\frac{J}{2J+1}} \sum_{m_l \mu} C^{J+1,1}_{JM}(m_l \mu) Y_{J+1 m_l}(\vec{\nu}) (\vec{\chi}_{\mu})_{\alpha} + \sqrt{\frac{J+1}{2J+1}} \sum_{m_l \mu} C^{J-1,1}_{JM}(m_l \mu) Y_{J-1 m_l}(\vec{\nu}) (\vec{\chi}_{\mu})_{\alpha}, \tag{A3}$$

where the spherical components of the photon spin vector are

$$(\vec{\chi}_{\mu})_{\alpha} = \delta_{\mu\alpha}. \tag{A4}$$

We consider first the magnetic photons. The insertion of Eqs. (A2) and (A4) in Eq. (A1) results

$$S^{(M)}_{JM}(\vec{\nu}) = \sum_{\tau} (-1)^{\tau} \sum_{m_l, m_l'} C^{J1}_{JM}(m_l \tau) Y_{Jm_l}(\vec{\nu}) C^{J1}_{JM}(m_l' \bar{\tau}) Y^{*}_{Jm_l'}(\vec{\nu}). \tag{A5}$$

Then

$$\sum_{M_f} S^{(M)}_{JM_f}(\vec{\nu}_f) = \sum_{M_f \tau} \sum_{m_l, m_l'} C^{J1}_{JM_f}(m_l \tau) C^{J1}_{JM_f}(m_l' \bar{\tau}) Y_{Jm_l}(\vec{\nu}_f) Y^{*}_{Jm_l'}(\vec{\nu}_f). \tag{A6}$$

The orthogonality condition for the Clebsh-Gordan coefficients reads

$$\sum_{M_f \tau} (-1)^{\tau} C^{l1}_{JM_f}(m_l \tau) C^{l'1}_{JM_f}(m_l' \bar{\tau}) = \delta_{ll'} \delta_{m_l m_l'}. \tag{A7}$$

Using Eq. (A6) for the $l = l' = J$ we arrive at

$$\sum_{M_f} S^{(M)}_{JM_f}(\vec{\nu}_f) = \sum_{m_l} |Y_{Jm}(\vec{\nu})_f|^2 = \frac{2J+1}{4\pi}, \tag{A8}$$

i.e., the probability given by Eq. (26) does not depend on $\vec{\nu}_f$. The same proof concerns the dependence on $\vec{\nu}_i$. The proof for the electric photon follows again from Eq. (A7), which gives zero result for $l \neq l'$.

APPENDIX B: EXPRESSION FOR THE COEFFICIENTS [EQS. (31), (34), (39), and (42)] VIA THE ANGLE

Choosing the direction of z axis in the photon momentum space along the vector ν_i we obtain the following expression for the coefficients [Eq. (30)] (for magnetic photons):

$$
\begin{aligned}
S^M_{Jj_0j_1}(\overrightarrow{\nu_f}) = {} & \frac{\omega^2(2J+1)^4}{8(2j_0+1)} \sum_{M_fM_i} (-1)^{M_f+M_i} \sum_{\hat{K}} (2\hat{K}+1) \\
& \times \begin{pmatrix} J & J & \hat{K} \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} J & \hat{K} & J \\ M_i & 0 & \bar{M}_i \end{pmatrix} \begin{Bmatrix} J & J & J \\ J & \hat{K} & J \end{Bmatrix} \\
& \times \sum_K \sqrt{\frac{2K+1}{4\pi}} \begin{pmatrix} J & J & K \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} J & K & J \\ M_f & 0 & \bar{M}_f \end{pmatrix} \\
& \times \begin{Bmatrix} J & J & J \\ J & K & J \end{Bmatrix} Y_{K0}(\overrightarrow{\nu_f}) \times \sum_x (2x+1) \\
& \times \begin{pmatrix} J & J & x \\ \bar{M}_f & M_f & 0 \end{pmatrix} \begin{pmatrix} J & J & x \\ \bar{M}_i & M_i & 0 \end{pmatrix} \begin{Bmatrix} J & x & J \\ j_1 & j_0 & j_1 \end{Bmatrix}^2 .
\end{aligned}
\tag{B1}
$$

Here we have taken into account that according to formulas (3)–(5) we represent operators $A_{JM_f}(\vec{\nu}_f)$, $A_{JM'_f}(\vec{\nu}_f)$, $A_{JM_i}(\vec{\nu}_i)$, and $A_{JM'_i}(\vec{\nu}_i)$ via spherical functions $Y_{JM_L}(\vec{\nu}_f)$, $Y_{JM_{L'}}(\vec{\nu}_f)$, $Y_{J\hat{M}_L}(\vec{\nu}_i)$, and $Y_{J\hat{M}_{L'}}(\vec{\nu}_i)$. Then having combined the corresponding functions into $Y_{KM}(\vec{\nu}_f)$, $Y_{\hat{K}\hat{M}}(\vec{\nu}_i)$ and using equation $Y_{\hat{K}\hat{M}}(\vec{\nu}_i)_{\vec{\nu}_i\uparrow\uparrow\vec{z}} = \sqrt{\frac{2\hat{K}+1}{4\pi}}\,\delta_{\hat{M}0}$ we can get rid from the direction $\vec{\nu}_i$. That is why there is no dependence of the $\vec{\nu}_i$ in Eq. (B1). In case of $J=1$, representing spherical functions in a standard way,

$$
Y_{K0}(\Theta) = \sqrt{\frac{2K+1}{4\pi}} P_K(\cos\Theta),
\tag{B2}
$$

where $\Theta = \angle\, \vec{\nu}_i \vec{\nu}_f$, $P_K(\cos\Theta)$ are Legendre polynomials, and summing over momentum projections and $K=0,2$, $\hat{K}=0,2$, $x=0,1,2$, we arrive finally at Eq. (31). Similarly we get expression (39).

For the electric photon instead of Eq. (B1) we get

$$
\begin{aligned}
S^E_{Jj_0j_1}(\overrightarrow{\nu_f}) = {} & \frac{\omega^2}{8(2j_0+1)} \sum_{M_fM_i} (-1)^{M_f+M_i} \sum_x (2x+1) \begin{pmatrix} J & J & x \\ \bar{M}_f & M_f & 0 \end{pmatrix} \begin{pmatrix} J & J & x \\ \bar{M}_i & M_i & 0 \end{pmatrix} \begin{Bmatrix} J & x & J \\ j_1 & j_0 & j_1 \end{Bmatrix}^2 \\
& \times \sum_{K\hat{K}} \sqrt{\frac{2K+1}{4\pi}} (2\hat{K}+1) Y_{K0}(\overrightarrow{\nu_f}) \times (J^2(2J+3)^2 \begin{pmatrix} J+1 & J+1 & K \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} J+1 & J+1 & \hat{K} \\ 0 & 0 & 0 \end{pmatrix} \begin{Bmatrix} J+1 & 1 & J \\ J & K & J+1 \end{Bmatrix} \\
& \times \begin{Bmatrix} J+1 & 1 & J \\ J & \hat{K} & J+1 \end{Bmatrix} + J(J+1)(2J-1)(2J+3) \begin{pmatrix} J-1 & J-1 & K \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} J+1 & J+1 & \hat{K} \\ 0 & 0 & 0 \end{pmatrix} \times \begin{Bmatrix} J-1 & 1 & J \\ J & K & J-1 \end{Bmatrix} \\
& \times \begin{Bmatrix} J+1 & 1 & J \\ J & \hat{K} & J+1 \end{Bmatrix} - (4J^2+6J)\sqrt{(J^2+J)(2J-1)(2J+3)} \begin{pmatrix} J+1 & J+1 & K \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} J-1 & J+1 & \hat{K} \\ 0 & 0 & 0 \end{pmatrix} \\
& \times \begin{Bmatrix} J+1 & 1 & J \\ J & K & J+1 \end{Bmatrix} \begin{Bmatrix} J+1 & 1 & J \\ J & \hat{K} & J-1 \end{Bmatrix} + 2J(J+1)(2J-1)(2J+3) \begin{pmatrix} J-1 & J+1 & K \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} J-1 & J+1 & \hat{K} \\ 0 & 0 & 0 \end{pmatrix} \\
& \times \begin{Bmatrix} J+1 & 1 & J \\ J & K & J-1 \end{Bmatrix} \begin{Bmatrix} J+1 & 1 & J \\ J & \hat{K} & J-1 \end{Bmatrix} - (2J^2+J-1)\sqrt{(J^2+J)(2J+1)(2J+3)} \begin{pmatrix} J-1 & J-1 & K \\ 0 & 0 & 0 \end{pmatrix} \\
& \times \begin{pmatrix} J-1 & J+1 & \hat{K} \\ 0 & 0 & 0 \end{pmatrix} \times \begin{Bmatrix} J-1 & 1 & J \\ J & K & J-1 \end{Bmatrix} \begin{Bmatrix} J+1 & 1 & J \\ J & \hat{K} & J-1 \end{Bmatrix} + 2J(J+1)(2J-1)(2J+3) \\
& \times \begin{pmatrix} J-1 & J+1 & K \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} J+1 & J-1 & \hat{K} \\ 0 & 0 & 0 \end{pmatrix} \times \begin{Bmatrix} J+1 & 1 & J \\ J & K & J-1 \end{Bmatrix} \begin{Bmatrix} J-1 & 1 & J \\ J & \hat{K} & J+1 \end{Bmatrix} \\
& - (2J^2+3J+1)\sqrt{(J^2+J)(2J+1)(2J+3)} \begin{pmatrix} J-1 & J-1 & K \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} J+1 & J-1 & \hat{K} \\ 0 & 0 & 0 \end{pmatrix} \\
& \times \begin{Bmatrix} J-1 & 1 & J \\ J & K & J-1 \end{Bmatrix} \begin{Bmatrix} J-1 & 1 & J \\ J & \hat{K} & J+1 \end{Bmatrix} .
\end{aligned}
\tag{B3}
$$

In case of $J=1$, performing the same operations as for the magnetic photon, we arrive at Eqs. (39) and (40).

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