Theoretical Study of the Accuracy Limits of Optical Resonance Frequency Measurements

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The principal limits for the accuracy of the resonance frequency measurements set by the asymmetry of the natural resonance line shape are studied and applied to the recent accurate frequency measurements in the two-photon $1s$-$2s$ resonance and in the one-photon $1s$-$2p$ resonance in a hydrogen atom. This limit for $1s$-$2s$ resonance is found to be $\sim 10^{-5}$ Hz compared to the accuracy achieved in experiment ± 46 Hz. In the case of a deuterium atom the limit is essentially larger: 10^{-2} Hz. For $1s$-$2p$ resonance the accuracy limit is 0.17 MHz while the uncertainty of the recent frequency measurement is about 6 MHz.

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The resonance approximation which reduces the description of the spectral line profile to the Lorentz contour is the cornerstone of all the modern transition frequency measurements in the resonance experiments. This approximation is employed for the extraction of the resonance frequency values from the experimental data. Here we address only the natural line profile, disregarding Doppler and collisional broadening. We assume that the latter effects can be minimized below the level of the natural line width. In the resonance approximation the line profile is described by two parameters, resonance frequency ω^{res} and the width Γ, and is symmetric with respect to ω^{res}. In the early paper by *F*. Low [1] it was pointed out that the resonance approximation is valid only up to a certain limit of accuracy which is defined by nonresonant (NR) corrections. Beyond this limit the line profile acquires asymmetric form and the definition of ω^{res} becomes nonunique. Moreover, the NR corrections are, in principle, process dependent, so the frequency value also will differ for different excitation processes. This means that the measurement of frequency is a well-defined procedure only up to some limit, which we define in our Letter. Going beyond this limit, it should be replaced by the direct measurement of the spectral line profile, which then becomes the only quantum-mechanical observable.

During the last few years the NR corrections were reconsidered in a series of theoretical works [2–4]. This reconsideration was triggered by the new extra accurate optical resonance experiments [5–7]. In the experiment [5] the natural line profile for the Lyman-α $1s$-$2p$ one-photon resonance in hydrogen was observed for the first time. In [6,7] the most accurate measurements in the optical region were performed for the two-photon $1s$-$2s$ resonance in hydrogen. In the latter experiments the absolute accuracy of the frequency measurements was as high as ± 46 Hz, or 10^{-14} in relative units. However, the scheme of these experiments causes serious difficulties for the theoretical description of NR corrections and hence for the definition of the accuracy limits in this case. In [6,7], the H atoms were excited from the ground $1s$ state to the $2s$ state via two-photon absorption in the space region free of external fields. After this the excited atoms moved during approximately 10^{-3} s to another space region where they meet a weak electric field. In this field $2s$ and $2p$ states are mixed and the atoms decay via the 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 $\hat{H}_{\mathrm{in}}$ and $\hat{H}_{\mathrm{out}}$.

The QED theory with different in and out Hamiltonians was developed by Fradkin, Gitman, and Shvartsman (see book [8]). This theory we will use below for the description of NR corrections in the two-photon $1s$-$2s$ resonance experiment. The earlier attempts [3,4] to describe the NR corrections in these experiments with standard QED methods cannot be considered as reliable.

The major contribution to NR corrections arises from the interference between the resonant and nonresonant terms in the expression for the scattering amplitude [2]. In [2] only the nonresonant terms with the same symmetry as the resonant one were included. As it was pointed out in [3] the most important contribution arises from the interference between the resonant term and nonresonant terms with another symmetry: the NR contribution of the $2p_{3/2}$ state to $1s$-$2p_{1/2}$ resonance was considered as an example. It was argued that this contribution can survive in differential (with respect to the angles) cross section for the resonance photon scattering on an atom. In the present work we will show that the NR contributions of this type survive both in differential and total cross section provided that the resonance line shape is fully natural. Moreover, in many cases the dominant NR contributions which define the accuracy limits for the frequency measurements arise from the neighbor hyperfine (HF) components. These components have different symmetry (values of the total angular mo-

mentum F of an atom) compared to the HF component which is used in the basic transition (see Fig. 1). The contribution of HF neighbor components was missing in the earlier investigations [2–4].

We start with the simplest case of the elastic photon scattering on the hydrogen atom in the absence of the electric field. The amplitude for this process can be written in a standard form:

$$A^{JM_f,JM_i}_{n_0j_0l_0m_f;n_0j_0l_0m_i} = \sum_{njlm} \frac{\langle n_0j_0l_0m_f|A^*_{JLM_f}|njlm\rangle\langle njlm|A_{JLM_i}|n_0j_0l_0m_i\rangle}{E_{njl} - E_{n_0j_0l_0} - \omega} \tag{1}$$

where the quantum numbers $njlm$ represent the standard set of one-electron quantum numbers for the electron in the hydrogen atom. The photons are characterized by the total angular momentum J, its projection M, and the orbital angular momentum L. The latter defines parity, or the type of the photon: electric or magnetic. The operator A_{JLM} corresponds to photon absorption, A^*_{JLM} denotes photon emission. In Eq. (1) the additional term where the operators A^* and A are interchanged and the sign of ω is reversed, is omitted. This term vanishes in the resonance approximation and does not contribute to the leading NR corrections. The resonance condition for the frequency is $\omega = E_{n_1j_1l_1} - E_{n_0j_0l_0}$ and in the resonance approximation in Eq. (1) only the terms with $njl = n_1j_1l_1$ are retained.

To avoid the singularity in Eq. (1) we insert in the denominator the width of the excited state $\Gamma_{n_1j_1l_1}$. The regular method for this insertion within the framework of QED is described in [1].

With this description of the resonant process the NR corrections arise as the other terms of the expansion in Eq. (1) with $njl \neq n_1j_1l_1$. Performing the summations over all the angular momentum projections, we obtain for the process probability in the resonance approximation

$$W_{n_0j_0l_0,n_0j_0l_0} = \frac{S^{1\gamma}(j_0j_1j_1J)}{(E_{n_1j_1l_1} - E_{n_0j_0l_0} - \omega)^2 + \frac{1}{4}\Gamma^2_{n_1j_1l_1}} \times \frac{1}{2\pi} W^{em}_{n_0j_0l_0,n_1j_1l_1} W^{ab}_{n_1j_1l_1,n_0j_0l_0}, \tag{2}$$

where W^{ab}, W^{em} are the standard absorption and emission probabilities per time unit and

$$S^{1\gamma}(j_0j_1j_2J) = \sum_a \begin{Bmatrix} j_0 & j_1 & J \\ J & J & a \\ j_2 & j_0 & J \end{Bmatrix}.$$

Taking into account the NR correction due to the closest to $n_1j_1l_1$ neighbor level $n_2j_2l_2$ results in

$$W_{n_0j_0l_0,n_0j_0l_0} = \frac{1}{2\pi}\Bigg[\frac{W^{em}_{n_0j_0l_0,n_1j_1l_1} W^{ab}_{n_1j_1l_1,n_0j_0l_0}}{(E_{n_1j_1l_1} - E_{n_0j_0l_0} - \omega)^2 + \frac{1}{4}\Gamma^2_{n_1j_1l_1}} S^{1\gamma}(j_0j_1j_1J) + 2\mathrm{Re}\frac{A^{em*}_{n_0j_0l_0,n_1j_1l_1} A^{em}_{n_0j_0l_0,n_2j_2l_2} A^{ab*}_{n_1j_1l_1,n_0j_0l_0} A^{ab}_{n_2j_2l_2,n_0j_0l_0}}{(E_{n_1j_1l_1} - E_{n_0j_0l_0} - \omega - \frac{i}{2}\Gamma_{n_1j_1l_1})(E_{n_2j_2l_2} - E_{n_0j_0l_0} - \omega - \frac{i}{2}\Gamma_{n_2j_2l_2})} S^{1\gamma}(j_0j_1j_2J)\Bigg]. \tag{3}$$

Here A^{em}, A^{ab} are the reduced emission and absorption amplitudes (reduced matrix elements of the photon emission and absorption operators). These matrix elements do not depend on the angular momentum projections. The factor $S^{1\gamma}(j_0j_1j_2J)$ does not vanish for $j_2 \neq j_1$, so that the dominant NR corrections due to the interference of the resonant and nonresonant terms with different symmetry ($j_1l_1 \neq jl$) can exist. They exist both in differential (with respect to the angles) and total probability; Eq. (3) describes the total probability. The differential one would depend on the angle $(\vec{n}_i, \vec{n}_f)$, where $\vec{n}_i$, $\vec{n}_f$ are the velocity directions for the absorbed and emitted photons. The vector $\vec{n}_i$ is fixed by the incident laser beam and $\vec{n}_f$ by the position of the detector.

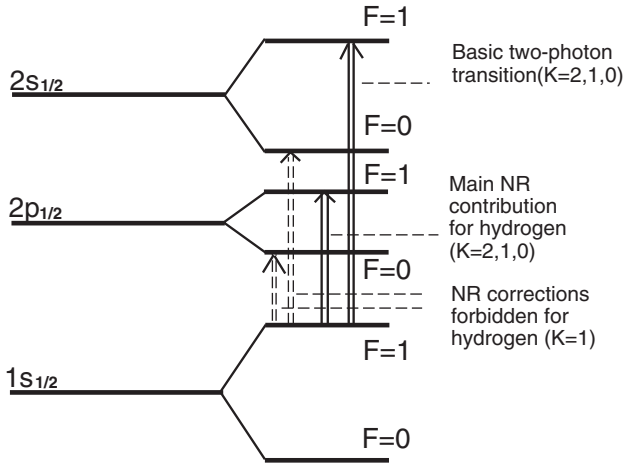

FIG. 1. Scheme of the levels for the two-photon $1s$-$2s$ transition. The vertical double lines denote the two-photon transitions. The K numbers denote the total angular momentum for a two-photon system, possible for different two-photon transitions.

Now we turn to the case of the $1s$-$2s$ two-photon absorption and delayed decay in electric field. The formalism of QED with different in and out Hamiltonians developed in [8] pursues the more complicated task: the creation of the electron-positron pairs in the strong electric field. We will apply this formalism to the case of a weak electric field. However, the situation in our case is also nonperturbative: due to the admixture of the $2p$ state into the $2s$ state the emission probability changes by 8 orders of magnitude. The change of the other atomic characteristics (Stark shift

of the energy levels, Stark splitting) we will consider as negligible. The criterion of the weak field will be $\varepsilon < \varepsilon_c = 475$ V/cm, where ε is the strength of the electric field. In the field $\varepsilon = \varepsilon_c$ the $2s$ and $2p$ levels are 100% mixed.

The Fradkin-Gitman-Shvartsman (FGS) theory [8] operates with two complete sets of eigenfunctions belonging to in and out Hamiltonians. This theory follows the standard QED approach in generalized form: S matrix, field operators in the Fock space, four-dimensional perturbation expansion for the S-matrix elements, Wick theorem, and Feynman graph techniques. Actually, the unique new element that we will have to use is the generalized FGS electron propagator. This propagator connects two vertices, which are described by in and out Hamiltonians, respectively.

In our case this means the absence or presence of the electric field. The FGS propagator looks like

$$S^{\mathrm{FGS}}(x_1x_2) = \Theta(t_1 - t_2) \sum_{\tilde{n},n(E_{\tilde{n};n}>0)} \psi_{\tilde{n}}(x_1)\omega_{\tilde{n}n}\bar{\psi}_n(x_2) - \Theta(t_2 - t_1) \sum_{\bar{n},n(E_{\tilde{n};n}<0)} \psi_n(x_1)\omega_{n\tilde{n}}\bar{\psi}_{\tilde{n}}(x_2), \quad (4)$$

where $\psi_{\tilde{n}}(x)$ are the solutions of the Dirac equation for the electron in the field of the nucleus and the external electric field, $\psi_n(x)$ are the solutions with zero external field; $E_{\tilde{n}}$, E_n are the corresponding eigenvalues. The matrix $\omega_{\tilde{n}n}$ in the weak-field limit reduces to an overlap integral

$$\omega_{\tilde{n}n} = \int \psi^+_{\tilde{n}}(\vec{x})\psi_n(\vec{x})d\vec{x} \equiv \langle\tilde{n}|n\rangle. \quad (5)$$

In the nonrelativistic limit, evidently valid for the neutral hydrogen atom, we replace the Dirac wave functions by Schrödinger ones and omit the negative-energy contribution in Eq. (4).

Within the FGS theory, the probability of absorption of two equivalent laser photons with frequency ω' by the electron in the hydrogen atom in its ground state with the subsequent delayed decay in external electric field in the resonance approximation ($n = a$, $\tilde{n} = \tilde{a}'$) looks like

$$dW_{\tilde{a}a} = \frac{1}{2\pi} \frac{W^{(em)}_{\tilde{a}\tilde{a}'}|\langle\tilde{a}'|a'\rangle|^2 W^{(ab2\gamma)}_{a'a}}{(E_{a'} - E_a - 2\omega')^2 + \frac{1}{4}\Gamma^2_{a'}} S^{2\gamma}_{aa'}, \quad (6)$$

where $W^{(ab2\gamma)}$ is the two-photon absorption probability, $S^{2\gamma}_{aa'}$ is the angular factor similar to $S^{1\gamma}$ in Eq. (2), and the resonance condition is $E_{a'} - E_a = 2\omega'$.

Taking into account the NR correction due to the closest to a' neighbor level a'' results in an expression similar to Eq. (3)

$$dW_{\tilde{a}a} = \frac{1}{2\pi}\left\{\frac{W^{em}_{\tilde{a}\tilde{a}'}|<\tilde{a}'|a'>|^2 W^{(ab2\gamma)}_{a'a}}{(E_{a'} - E_a - 2\omega')^2 + \frac{1}{4}\Gamma^2_{a'}} S^{2\gamma}_{aa'} + 2\mathrm{Re}\left[\frac{A^{em*}_{\tilde{a}\tilde{a}'}\langle\tilde{a}'|a'\rangle^* A^{em}_{\tilde{a}\tilde{a}''}\langle\tilde{a}''|a''\rangle A^{(ab2\gamma)*}_{a'a} A^{(ab2\gamma)}_{a''a}}{(E_{a'} - E_a - 2\omega' - \frac{i}{2}\Gamma_{a'})(E_{a''} - E_a - 2\omega' - \frac{i}{2}\Gamma_{a''})}\right] S^{2\gamma}_{aa''}\right\}, \quad (7)$$

where $A^{(ab2\gamma)}_{a'a}$, $A^{(ab2\gamma)}_{a''a}$ are the reduced two-photon absorption amplitudes.

For moving further we have to choose the procedure for determination of the resonance photon frequency. In [2–4] the evaluation of the maximum value of the frequency distribution was used for this purpose. As it was shown in [3] any other procedure (e.g., finding a "center of gravity" for the line profile) would give the result quite close to the choice formulated above. In case of the Lorentz profile all the methods of defining ω^{res} give the same result $\omega^{\mathrm{res}}_0 = E_{a'} - E_a$. With our choice, the NR correction will look like

$$\delta\omega^{\mathrm{NR}} = \omega^{\max} - \omega^{\mathrm{res}}_0, \quad (8)$$

where $\omega^{\max}$ is the frequency value, corresponding to the maximum of the frequency distribution.

The first example that we will consider is the two-photon $1s_{1/2}(F = 1) + 2\gamma \to 2s_{1/2}(F = 1)$ transition in hydrogen [6,7]. The value $K = 1$ for the total angular momentum K of the two-photon system with equal photon frequencies is strictly forbidden. This concerns exactly the case of the two-photon absorption of laser photons in experiment [6,7] (see Fig. 1). According to this picture, the main NR contribution arises from the transition $1s_{1/2}(F = 1) \to 2p_{1/2}(F = 1)$. For deriving the NR correction we have to use an expression (7) where we have to set $a = 1s_{1/2}(F = 1)$, $a' = 2s_{1/2}(F - 1)$ and $a'' = 2p_{1/2}(F = 1)$. In a weak electric field $\psi_{\tilde{a}} \simeq \psi_a$, $\psi_{\tilde{a}'} \simeq \psi_{a'} + \eta\psi_{a''}$, $\psi_{a''} \simeq \psi_{a''} - \eta\psi_{a'}$ and the overlap integrals are $\langle\tilde{a}|a\rangle \simeq \langle\tilde{a}''|a''\rangle \simeq 1$. Here $\eta = |\Delta E_S|/\Delta E_L$ is the Stark shift to the Lamb shift ratio. This ratio is $\eta = 1$ for a field $\varepsilon = \varepsilon_c$. For deriving the NR correction we set in Eq. (7) $E_{a'} - E_a = \omega^{\mathrm{res}}_0$, $E_{a''} - E_a = \omega^{\mathrm{res}}_0 - \Delta E_L$, where $\Delta E_L = E_{a''} - E_{a'} \approx 10^3$ MHz is the Lamb shift. The width $\Gamma_{a'}$ in case of the experiment [6,7] is determined by the experimental setup (the time delay before the excited atoms enter the electric field region) and is equal to $\Gamma_{a'} = \Gamma_{\mathrm{exp}} \approx 1$ kHz. Insertion of the wave functions $\psi_{\tilde{a}}$, $\psi_{\tilde{a}'}$, and $\psi_{\tilde{a}''}$ in the emission amplitudes yields: $A^{em}_{\tilde{a}\tilde{a}'} \approx \eta(\Gamma_{2p})^{1/2}$, $A^{em}_{\tilde{a}\tilde{a}''} \approx (\Gamma_{2p})^{1/2}$, where Γ_{2p} is the width of the $2p$ state. Then, evaluating the maximum value of Eq. (7) with respect to ω' and defining the NR correction according to Eq. (8) we find

$$|\delta\omega^{\mathrm{NR}}| \approx \frac{1}{4}\frac{\Gamma^2_{\mathrm{exp}}}{\Delta E_L}\left(\frac{W^{2\gamma}_{1s,2p_{1/2}}}{W^{2\gamma}_{1s,2s}}\right)^{1/2}\frac{1}{\eta}\frac{S^{2\gamma}_{aa''}(F, F'')}{S^{2\gamma}_{aa'}(F, F')}, \quad (9)$$

where $S^{2\gamma}_{aa'}(F, F')$ is the angular factor defined for the transition between the two hyperfine sublevels. In the present case $F = F' = F'' = 1$, and the corresponding angular factors are equal: $S^{2\gamma}_{aa'}(F, F') = S^{2\gamma}_{aa''}(F, F'') = 11/18$. The probability for the two-photon transition $W^{2\gamma(E1E1)}_{1s,2s}$ is very well known. An accurate nonrelativistic

value for this transition was obtained in [9]: $W^{2\gamma(E1E1)}_{1s,2s} = 1.32 \times 10^{-3}(\alpha Z)^6$ a.u. Here α is the fine structure constant, Z is the nuclear charge and it is assumed that $\alpha Z \ll 1$. The probability $W^{2\gamma}_{1s,2p_{1/2}}$ was evaluated recently numerically for all Z values ($1 \ll Z \ll 100$) [10]. The result is $W^{2\gamma}_{1s,2p_{1/2}} = W^{2\gamma(E1M1)}_{1s,2p_{1/2}} + W^{2\gamma(E1E2)}_{1s,2p_{1/2}} = 2.907 \times 10^{-5}(\alpha Z)^8 + 3.69 \times 10^{-6}(\alpha Z)^8$ (a.u.).

The dependence η^{-1} in Eq. (9) cannot be continued to zero field; the meaningful limit is set by the field where the decay rate of the $2s$ level due to the admixture of the $2p$ state becomes equal to the natural decay width of the $2s$ state. This limiting field strength would be so small, that it cannot be used in a real experiment. Inserting all the numbers in Eq. (9) and taking $\eta = 0.1$, which corresponds to the weak electric field $\varepsilon = 47,5$ V/cm, we obtain the final result $|\delta\omega^{\mathrm{NR}}| \approx 10^{-5}$ Hz. This accuracy limit is still far from the recent inaccuracy estimate in the experiments [6,7]: ± 46 Hz. However, it is important to notice that the situation in hydrogen seems to be rather fortunate for the accurate resonance frequency measurement due to the absence of the transition to another ($F = 0$) hyperfine sublevel of the $2s$ state (see Fig. 1). For comparison, in the deuterium where the total atom angular momentum values for the $1s$ and $2s$ levels are $F = 1/2, 3/2$, respectively, this transition is allowed and NR correction by order of magnitude is

$$|\delta\omega^{\mathrm{NR}}| \sim \frac{1}{4}\frac{\Gamma^2_{\mathrm{exp}}}{\Delta E_{\mathrm{HFS}}}, \tag{10}$$

where $\Delta E_{\mathrm{HFS}} = E_{2s_{1/2}(F=3/2)} - E_{2s_{1/2}(F=1/2)} \approx 100$ MHz is the hyperfine-structure interval for $2s$ level. Taking the same Γ_{exp} value as in [6,7] we would have $|\delta\omega^{\mathrm{NR}}| \sim 10^{-2}$ Hz, which is 3 orders of magnitude larger than for hydrogen. This is not so far from the accuracy limit of about 0.1 Hz that was considered in [7] as achievable in the future with the use of colder hydrogen atoms.

A quite different situation arises for the $1s$-$2p$ resonant experiment in hydrogen [5]. In [5] the resonance $1s(F = 1) \rightarrow 2p_{3/2}(F = 2, 1)$ was measured. The hyperfine structure for the $2p_{3/2}$ level was not resolved since $\Gamma_{2p} > \Delta E_{\mathrm{HFS}}(2p_3/2)$: $\Gamma_{2p} \approx 100$ MHz, $\Delta E_{\mathrm{HFS}}(2p_{3/2}) = E(2p_{3/2}, F = 2) - E(2p_{3/2}, F = 1) = 23.7$ MHz. This is a typical case of overlapping resonances for two hyperfine sublevels. However, due to the presence of interference terms [see Eq. (3)] the line shape deviates from the overlap of two Lorentz profiles and can be presented by an expression

$$F(\omega) = \frac{f(F,F')}{(\omega_1^{\mathrm{res}} - \omega)^2 + \frac{1}{4}\Gamma^2_{2p}} + \frac{f(F,F'')}{(\omega_2^{\mathrm{res}} - \omega)^2 + \frac{1}{4}\Gamma^2_{2p}} + 2\mathrm{Re}\frac{g(F,F',F'')}{(\omega_1^{\mathrm{res}} - \omega - \frac{i}{2}\Gamma_{2p})(\omega_2^{\mathrm{res}} - \omega - \frac{i}{2}\Gamma_{2p})}, \tag{11}$$

where $\omega_1^{\mathrm{res}} = E(2p_{3/2}, F = 2) - E(1s_{1/2}, F = 1)$, $\omega_2^{\mathrm{res}} = E(2p_{3/2}, F = 1) - E(1s_{1/2,F=1})$, f, g are the angular factors, similar to $S^{1\gamma}$ in Eq. (2). These angular factors play an important role in defining the line shape Eq. (11): $f(1,2):f(1,1):g(1,2,1) = 181:1:0.307$. Thus the line shape exhibits the one-peak structure (the larger peak fully screens the smaller one). The deviation from the overlap of two Lorentz profiles is of order $0.307/181 \approx 0.17\%$. Then the absolute accuracy limit for determination of the $1s(F = 1) \rightarrow 2p_{3/2}$ transition frequency can be estimated as $\Gamma_{2p} \times 0.0017 \approx 0.17$ MHz. The existence of the interference terms that distort the closely lying resonances is well known (see, for example, [11]). However, in [11] this distortion was included in the error bars for the transition frequencies not distinguishing between other contributions of purely technical origin, such as the laser intensity distribution and the spread of atomic velocities. Nowadays, when the two latter effects in practice can be diminished completely (e.g., by the use of colder atoms) it becomes more useful to introduce such quantity as an ''absolute limit'' for the frequency determination in the resonance experiments. Actually any resonance experiment pretending on the utmost accuracy should contain fittings of the experimental data with a suitable theoretical line shape including the effects of interference, originating from NR corrections. In the case of $1s - 2p_{3/2}$ transitions Eq. (11) provides such a theoretical line shape, derived from first principles of QED.

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