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Dipole scattering of a short radiation pulse on hydrogen-like atoms

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Abstract: Our theoretical model of forced dipole oscillation demonstrates that when the amplitude of the forcing field is changing fast, the oscillations of the bound electron in the atom or molecule initially proceed at two frequencies: the frequency of the natural electron oscillations and the frequency of the forcing field. Particularly, applied to the science of scattering, this model of transient forced atomic and molecular oscillations suggests that accurate interpretation of the laser scattering experiments using short laser pulses must include both the conventionally known scattering at the laser frequency (Rayleigh) and the predicted by our theoretical spectral emission that corresponds to the natural frequency of the electronic oscillations. This article presents the results of numerical simulations using our model performed for the hydrogen atom. The characteristics of the components of scattered radiation, their polarization, and Doppler thermal broadening are discussed.

1. Introduction

Study of Rayleigh scattering of light by atoms and molecules has a long history. Currently, Rayleigh scattering of laser radiation is widely used for diagnostics of gases and weakly ionized plasma [1]. Considering the long history of research one would assume that the fundamentals of the classical theory of Rayleigh scattering are well established and the science is settled. However, it appears that one important issue was omitted from the fundamentals of Rayleigh scattering theory - the transient (time-dependent) model of light scattering. The current scattering theory is solely based on the Lorentz's model that considers temporally established forced oscillator [2]. It is commonly understood that, for the typical atomic and molecular oscillators the time to reach established (repeatable) forced oscillations is determined by the characteristic damping time of the natural oscillation of the optical electrons. Then simple estimates based on the available data for light attenuation in gases due to scattering show that the time required to reach established electron oscillations is ranging from tens of picoseconds to nanoseconds. Such time scale was in the realm of pure fiction during the period from the 17th to the 19th century when the foundations of the modern optics were developed. Hence, the established atomic oscillation model seemed to be well suited. In the middle of the 20th century, after the invention of lasers, sub-nanosecond time scale became a common reality. However, the basic assumptions of the foundational paradigm of the optics remained unchanged. Struggling with the temptation to significantly deviate for the topic, we still would like to state that, both the electrodynamics and quantum mechanics theories while applied for the needs of optics, describe the light-matter interaction utilizing the same antique paradigm of slowly changing and established forced oscillations of optical electrons. Returning to the topic of this publication: we suggest that under the conditions typical for a pulsed laser interaction, the forced atomic oscillations of optical electrons could occur in a transient mode during which the optical electron oscillations contain component that has a frequency close to the natural oscillation frequency.

We described the transient atomic oscillation in a recent paper [3] by considering scattering of a relatively short pulse of electromagnetic radiation. This theoretical work predicted that, when the

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dipole approximation is valid, the re-radiated optical field contains at least two components: one at the frequency of the incident electromagnetic wave, ω_L, the other corresponds to the frequency $\omega' \approx \omega_0$, where ω_0 is the natural frequency electron oscillations. The former component is known as scattering and the latter component, that we called "pinging" emission, is previously unknown and yet not observed. Our work showed that the "pinging" emission is predicted to occur regardless of which oscillator model is used to describe the interaction of an optical electron with an electromagnetic wave: a classical Lorentzian oscillator [2] or an oscillator with realistic nonlinear potential [35]. Considering the widespread prevalence of laser diagnostic methods based on Rayleigh scattering, it is necessary to keep in mind when interpreting these experiments that for relatively short pulses the total cross-section of Rayleigh scattering can be very different from that obtained for a long pulse or stationary radiation.

2. Theoretical model

As an example, in this paper we consider the classical approximation of interaction of a hydrogen atom with an electromagnetic wave, when the dipole approximation is valid. As in our previous works [3–5], the equation of motion for electron displacement from the stationary Bohr's orbit of the s-state of a hydrogen atom is provided by the solution of the following equation of motion:

$$\ddot{\boldsymbol{r}} + \frac{2\boldsymbol{U}_0\boldsymbol{r}_0^2}{\boldsymbol{m}}\left(\frac{1}{\boldsymbol{r}_0\boldsymbol{r}^2} - \frac{1}{\boldsymbol{r}^3}\right) - \frac{\boldsymbol{\xi}}{\boldsymbol{m}}\dddot{\boldsymbol{r}} = -\frac{\boldsymbol{e}}{\boldsymbol{m}}\boldsymbol{E}(\mathrm{t}), \tag{1}$$

where $\boldsymbol{\xi} = \boldsymbol{e}^2/6\pi\boldsymbol{\varepsilon}_0\boldsymbol{c}^3$, $\boldsymbol{\varepsilon}_0$ is the vacuum permittivity, $\boldsymbol{c}$ is the speed of light, m, e are the electron mass and charge, r_0 is the radius of the equilibrium orbit of electron, and U_0 is the depth of effective electron potential. For a hydrogen atom in a spherically symmetric s-state at the lowest energy level the effective potential $\boldsymbol{U}_0 = \boldsymbol{I}_i$, where $\boldsymbol{I}_i$=13.6 eV is the ionization potential from the ground state. Since there is no reliable and unambiguous information about the values of the atomic radii, let us here assume, for certainty, the value $\boldsymbol{r}_0 = \boldsymbol{r}_B$, where $\boldsymbol{r}_B$ = 52.9 pm is the Bohr radius. Note, that the allowable region of the possible radius of hydrogen atom could range from the Bohr's radius to the Van der Waals' radius ~ 120 pm. As far as we know, no one has made direct measurements of the effective radius of a hydrogen atom. As we will see below, the value of the radius of a hydrogen atom can be estimated from the observed spectra of dipole radiation generated by a short pulse of electromagnetic radiation. Thus, the latter could be an excellent tool of the direct characterization of atoms and molecules.

For a given pulse of electromagnetic radiation, one can find a solution to the Eq. (1) that satisfies the initial conditions

$$\boldsymbol{r}(\boldsymbol{0}) = \boldsymbol{r}_0, \quad \dot{\boldsymbol{r}}(\boldsymbol{0}) = \boldsymbol{0}. \tag{2}$$

As a result of electron displacement, the atomic dipole moment changes

$$\boldsymbol{d}(\boldsymbol{t}) = -\boldsymbol{e}\boldsymbol{\delta}\boldsymbol{r}(\boldsymbol{t}) = -\boldsymbol{e}(\boldsymbol{r}(\boldsymbol{t}) - \boldsymbol{r}_0) \tag{3}$$

and electromagnetic radiation with an electric field is induced

$$\boldsymbol{E}_{\boldsymbol{d}}(\boldsymbol{t}) \propto \ddot{\boldsymbol{d}}(\boldsymbol{t}), \tag{4}$$

with polarization coinciding with the initial polarization of the incident electromagnetic wave.

Let's assume that the induced atomic dipole emission is observed by a sufficiently broadband spectrograph. The spectral power density of this re-emitted radiation is

$$\boldsymbol{S}(\boldsymbol{\omega}) \propto \boldsymbol{a}^2(\boldsymbol{\omega}) + \boldsymbol{b}^2(\boldsymbol{\omega}), \tag{5}$$

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where

$$a(\omega) = \frac{1}{\pi}\int_{-\infty}^{\infty} E_d(t)\cos(\omega t)dt, \tag{6}$$

and

$$b(\omega) = \frac{1}{\pi}\int_{-\infty}^{\infty} E_d(t)\sin(\omega t)dt \tag{7}$$

are the components of the direct Fourier transform [6].

As was shown in [4], the solution of Eq. (1) becomes very close to the solution described by the classical Lorentz oscillator for decreasing incident radiation intensity, with the exception of higher harmonics. That is, the spectrum contains the main harmonic at the laser frequency ω_L and the radiation component at the "pinging" frequency $\omega' \approx \omega_0$.

3. Results of simulations and discussion

The results of simulation of the re-emission produced by a forced atomic oscillator described by the Eq. (1) is shown in the Figs. 1–3 for a pulse shape given by the Eq. (8) and for laser wavelength λ_L =532 nm.

$$\frac{I(t)}{I_0} = \begin{cases} \cos^2(|t|/t_p), & |t|/t_p \le \pi/2 \\ 0, & |t|/t_p > \pi/2 \end{cases}, \tag{8}$$

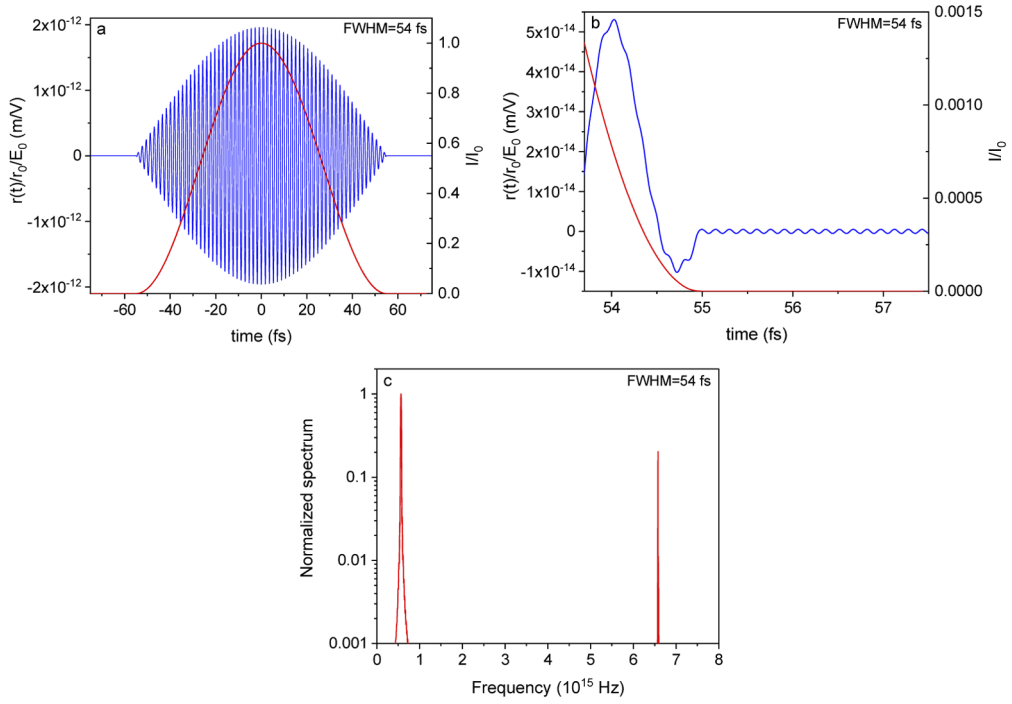

Fig. 1. (a) - Time dependent relative displacement of the forced electron oscillations divided by the amplitude of the laser electric field and the laser pulse shape; (b) - Same as in (a) shown during the later time; (c) normalized spectral power density of the response of hydrogen atom.

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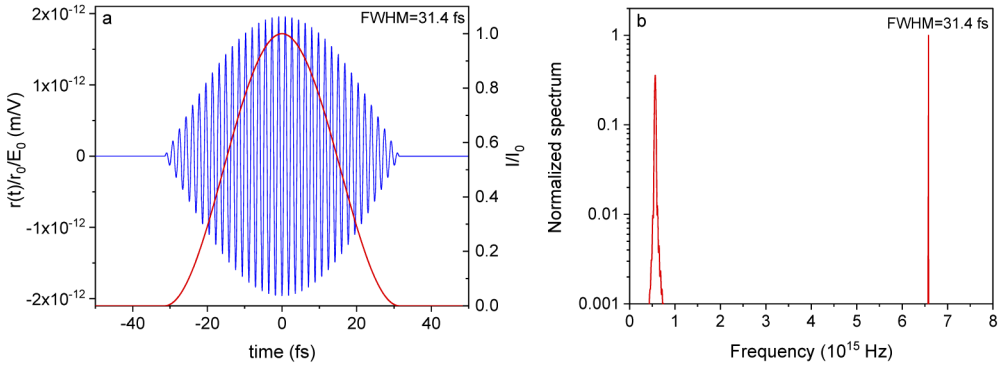

Fig. 2. (a) - Time dependent relative displacement of the forced electron oscillations divided by the amplitude of the laser electric field and the laser pulse shape for FWHM 31.4 fs and (b) normalized spectral power density of the response of hydrogen atom.

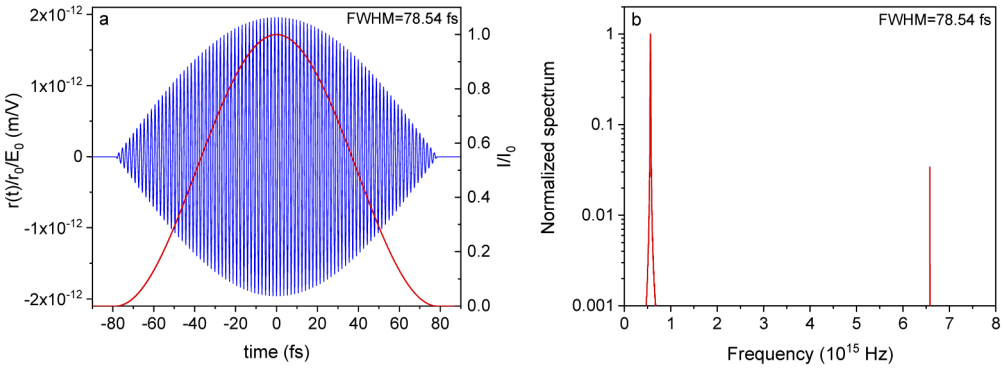

Fig. 3. (a) - Time dependent relative displacement of the forced electron oscillations divided by the amplitude of the laser electric field and the laser pulse shape for FWHM 78.54 fs and (b) normalized spectral power density of the response of hydrogen atom.

The corresponding reduced values of the dynamics of the amplitude of the perturbing electric field:

$$\boldsymbol{E_a(t)/E_0 = (I(t)/I_0)^{1/2}}. \tag{9}$$

Here $\boldsymbol{I_0}$ and $\boldsymbol{E_0}$ are the amplitudes of the radiation intensity and electric field, respectively.

An example of calculation for a pulse with a FWHM of 54 fs that corresponds to the parameter $\boldsymbol{t_p}$ = 35 fs is shown in Fig. 1.

The presence of the "pinging" frequency is clearly demonstrated in the Fig. 1(b). After the end of the laser pulse depicted by the red line, the forced oscillation ends at approximately 55-th femtosecond and after that the electron continues small amplitude oscillation with the frequency of "pinging" $\boldsymbol{\omega' \approx \omega_0 \approx 2\pi \cdot 6.59 \cdot 10^{15}}$ **rad/s**. The spectral analysis shown in the Fig. 1(c) demonstrates the power content of the "pinging" frequency in the spectral power density distribution.

With an increase in the steepness of the pulse fronts, the relative amplitude of the **"**pinging" component in the re-radiation spectrum at the frequency $\boldsymbol{\omega'}$ increases and it could become comparable or even exceed the spectral power density component of scattered light at the laser frequency $\boldsymbol{\omega_L}$. In confirmation of the above, Figs. 2 and 3 show examples of calculations of shorter (FWHM 31.4 fs) and longer (FWHM 78.54 fs) pulses with parameters $\boldsymbol{t_p}$ = 20 fs and $\boldsymbol{t_p}$ = 50 fs, respectively.

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In the case of a low intensity of the incident radiation, when the nonlinearity of forced electron motion and related to that generation of higher harmonics can be neglected [5], we can assume that the dipole moment has two components corresponding to oscillations at the "driving" and "pinging" frequencies:

$$\boldsymbol{d}(\boldsymbol{t}) = -\boldsymbol{e}\boldsymbol{\delta r}(\boldsymbol{t}) \approx -\boldsymbol{e}(\boldsymbol{\delta r}_{\omega_L}(\boldsymbol{t}) + \boldsymbol{\delta r}_{\omega'}(\boldsymbol{t})) = \boldsymbol{d}_{\omega_L}(\boldsymbol{t}) + \boldsymbol{d}_{\omega'}(\boldsymbol{t}). \tag{10}$$

We can easily perform analysis of the limiting case when the laser is turned on instantaneously and the forced radiation is computed for a linear oscillator with the natural frequency ω_0 and the damping constant γ [7]. As follows from our previous work [4]

$$\omega_0 = (2U_0/mr_0^2)^{1/2}, \tag{11}$$

$$\gamma = \frac{\xi}{m}\omega_0^2 = \frac{e^2\omega_0^2}{6\pi\varepsilon_0 mc^3},\ 1/\mathrm{s}. \tag{12}$$

Note, that for hydrogen atom the characteristic damping time of electron natural oscillations is $\tau_\gamma = \gamma^{-1} \sim 7.5 \cdot 10^{-10}, s$. This time is more than six orders of magnitude larger than the period of natural oscillations, $T_{\omega_0} = \frac{2\pi}{\omega_0} \approx 1.52 \cdot 10^{-16}$ s and it is comparable to the typical pulse durations of a Q-switched lasers.

Previously we showed [4] that for a linear Lorentz oscillator, the "pinging" frequency of the re-radiation, ω', is less than the natural frequency of the oscillator, ω_0,

$$\omega' = \left(\omega_0^2 - \frac{1}{4}\gamma^2\right)^{1/2}, \tag{13}$$

Since $\gamma \ll \omega_0$, the "pinging" frequency, ω', is very close to the natural oscillation frequency ω_0. Given that $\omega' \neq \omega_0$ it is possible that the radiation of the atom at the pinging frequency will propagate in gas while being absorbed only slightly on the wings of the line resonance at the natural oscillation frequency, ω_0. The actual propagation length of the radiation of the atom at the pinging frequency could be estimated. However, in order to do so, a broad range of gas densities, gas types, laser pulse energies, laser pulse durations, and laser pulse irradiances should be considered. Such a set of calculations is outside of the scope of the current work, and we leave it for a future study.

For typical laser interaction conditions $\gamma \ll \omega_L, \omega'$ and $\omega_L \ll \omega'$, or $\omega_L \gg \omega'$. Then the total power of Rayleigh and "pinging" scattering, averaged over a time $\Delta t \geq max\left\{\frac{2\pi}{\omega_L}, \frac{2\pi}{\omega'}\right\}$, is

$$P_{tot} = P_{\omega_L} + P_{\omega'} = I_L\left(\sigma_{\omega_L} + \sigma_{\omega'}\right) \propto \langle \ddot{d}^2 \rangle = \langle \ddot{d}_{\omega_L}{}^2 \rangle + \langle \ddot{d}_{\omega'}{}^2 \rangle, \tag{14}$$

where I_L is the laser radiation intensity, and σ_{ω_L} and $\sigma_{\omega'}$ are the Rayleigh and "pinging" cross sections, correspondingly. From this consideration it follows that, the interpretation of the data of scattering diagnostics experiments that utilize short laser pulses with duration $\tau \leq \gamma^{-1}$ should take into account that the total scattering cross-section includes the cross section of laser light re-emission at the "pinging" frequency: $\sigma_t = \sigma_{\omega_L} + \sigma_{\omega'}$. The analysis of short laser pulse scattering experiments will lead to significant error if this total cross section is treated as solely due to the Rayleigh scattering determined from the measurements performed using long pulse or cw laser sources. Additionally, in the analysis of short laser pulse scattering experiments one should be mindful of the fact that the cross section of the "pinging" component is independent of the laser wavelength while the cross-section of the Rayleigh scattering is inversely proportional to the 4th power of the wavelength, or, expressed as a function of frequency:

$$\sigma_{\omega_L}/\sigma_{\omega'} \sim \omega_L^4/\omega'^4. \tag{15}$$

Thus, the possible error of not including pinging in the analysis of the laser Rayleigh scattering experiments for short laser pulses is higher for longer laser wavelengths.

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The diagnostics based on Rayleigh scattering use spectrally resolved measurements when the temperature of scattering medium is of interest. The shape of the spectral line of scattered laser light is used to determine temperature assuming Doppler broadening. Note that a narrow-band receiver tuned to the vicinity of the laser frequency ω_L would provide the correct Rayleigh scattering cross section, the value of which is not affected by "pinging" radiation. However, the density of scattered medium is usually measured using energy or power meters that provide spectrally integrated value. In this case, disregarding "pinging" component will lead to an error that would increase with decrease of the laser pulse duration. Recently, the demand is increasing to measure density of rarified medium when the scattered light has low intensity with large intensity background. This leads to increased use of short and ultrashort pulse lasers.

Let's assume that the incident radiation is plane-polarized. Due to the thermal motion of the atoms/molecules, each of the components of the scattered radiation is subject to doubled Doppler broadening. Full with half maximum (FWHM) of thermal Doppler broadening at the frequency of the disturbing field takes into account a double frequency shift and depends on the angle $\boldsymbol{\theta}$ between the initial direction of the laser radiation and the radiation scattered towards the detector [1]:

$$\Delta\nu_T = \frac{\omega_L}{\pi c}\left(\frac{8k_B T \ln 2}{M}\right)^{1/2} \sin\left(\frac{\theta}{2}\right), \tag{16}$$

where M is the mass of atoms/molecules, T is the gas temperature in the laser-gas interaction region, k_B is the Boltzmann constant. In the transient (pulsed) mode, the component of the dipole radiation at the "pinging" frequency has the same polarization as the main mode of Rayleigh scattering at the laser frequency ω_L. However, the FWHM of the Doppler broadening of the "pinging" component is independent of the observation angle $\boldsymbol{\theta}$ and is subject to a single Doppler frequency shift:

$$\Delta\nu'_T = \frac{\omega'}{\pi c}\left(\frac{k_B T \ln 2}{2M}\right)^{1/2}. \tag{17}$$

Thus, observation of the spectral characteristics of the "pinging" signal also allows one to obtain the same information about the gas temperature as Rayleigh scattering. However, since the "pinging" frequency is different and independent from the laser frequency, ω_L, there is no need to use a filter for the center of the laser emission line as in the case of so-called filtered Rayleigh scattering [1]. An additional benefit of the "pinging" scattering diagnostics is due to the uniqueness of the natural electron oscillation frequency for each type of atoms and molecules. Thus, using "pinging" one could obtain information on the composition of a multicomponent gas and, with appropriate normalization, information on the concentrations of individual components of the mixture.

As we demonstrated above (see Eq. (13)) the frequency of the "pinging" component of laser forced re-radiation is very close to the natural oscillation frequency of the optical electron, ω_0, that is determined by the effective potential, U_0, and the radius of the orbit, r_0, of the optical electron. For the considered case of s-state of the hydrogen atom and assuming the parameters stated above, the "pinging" dipole re-radiation has the wavelength of $\lambda' \approx 45.45$ nm and the quantum $\varepsilon' \approx 27.3$ eV. The energy of the re-radiated "pinging" quanta is approximately twice larger than the ionization potential of hydrogen. Then the ionization of the surrounding atoms with the "pinging" photons could be an additional mechanism for the generation of the seed electrons for the laser optical breakdown since it requires significantly lower laser intensities that are required by the Keldysh mechanism of multiphoton or tunnel ionization [8].

Note that the "pinging" radiation wavelength was obtained based on the accepted values of the effective potential, U_0, and the radius of the orbit, r_0. Experimental observation of 'pinging" radiation will answer the question of how valid the selected values of these parameters are.

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4. Conclusions

Our recently developed theoretical model of forced atomic oscillator predicts that a pulse of electromagnetic radiation with fronts that are faster than the characteristic time of atomic radiation decay, γ^{-1}, additionally to the re-emission of radiation at the forcing frequency known as the Rayleigh scattering, the re-emission will contain a component with the frequency that is slightly lower than the frequency of the natural oscillations of the optical electron on its stationary orbit. We call this newly predicted effect the "pinging" emission. If the "pinging" existence is experimentally verified, then the analysis of elastic scattering of short laser pulses on atoms and molecules must include the re-emission at the frequency that is close to the frequency of the natural oscillations of the optical electron that is independent of the irradiation frequency. Disregarding the component of scattering due to the "pinging" emission would lead to significant errors in determining the scattering cross-section.

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Disclosures

The authors declare no conflicts of interest.

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