Excitation of atoms and ions in plasmas by ultra-short electromagnetic pulses

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Abstract. The problem of atoms and ions diagnostics in rarefied and dense plasmas by ultra-short laser pulses (USP) is under consideration. The application of USP provides: 1) excitation from ground states due to their carrier frequency high enough, 2) penetration into optically dense media due to short pulses duration. The excitation from ground atomic states increases sharply populations of excited atomic states in contrast with standard laser induced fluorescence spectroscopy based on radiative transitions between excited atomic states. New broadening parameter in radiation absorption, namely inverse pulse duration time 1/τ appears in addition to standard line-shape width in the profile G(ω). The Lyman-beta absorption spectra for USP are calculated for Holtsmark static broadening mechanism. Excitation of highly charged H-like ions in hot plasmas is described by both Gaussian shapes for Doppler broadening and pulse spectrum resulting in analytical absorption line-shape. USP penetration into optically thick media and corresponding excitation probability are calculated. It is shown a great effect of USP duration on excitation probabilities in optically thick media. The typical situations for plasma diagnostics by USP are discussed in details.

1. Introduction
The standard line broadening theory deals with the investigations of emitted or absorbed atomic/ions spectra on the frequency detuning from unperturbed frequency of specific atomic transition. So the line shapes are described by their dependences on the dimensionless ratio of frequency shift to the typical line widths depending on specific broadening mechanism (Doppler, Stark, collisions, etc). There are several problems in investigations of atomic spectra under plasma conditions. The first one is connected with small populations of excited atomic levels resulting in difficulties of registration signals of laser induced fluorescence (LIF). Really the most laser experiments deal with local excitations of atomic system from one to another excited atomic energy states where the transition frequency belongs to visible spectral range. At the same time the population difference between excited states are not so different to increase the difference sharply under the action of laser pulse. So the fluorescence signal is blended strongly by plasma background radiation. The second problem is connected with penetration of laser radiation inside optically dense medium when one would try to excite atomic systems from ground states.

Both problems pointed above can be solved by application of ultrashort laser pulses for plasma diagnostics by laser radiation. Really modern technology of ultrashort pulses generations provides the
movement of carrier laser frequency in VUV and even soft X-ray spectral range, see for example reference [1] where pulse duration is of order of 67 as for the carrier frequency near 200 eV. It opens possibilities to excite atomic systems not only from excited but also from ground atomic states. As far as the populations of ground states are 3-4 order of magnitude larger than excited states that means a sharp increase of atomic states populations by their excitations from ground states. The second problem of large optical thickness for radiative transitions from ground states is also simplified by application of ultrashort laser pulses. Such simplification is achieved due to broad spectra of the ultrashort pulses which make it possible to avoid strong absorption in the far wings of the broadening spectral lines. Both problems will be considered in details in present paper.

So in the case of ultrashort pulses the new parameter appears in the absorption line spectra connected with the pulse duration $\tau$ in addition with standard broadening parameters connected with standard line shapes. The connection with the new parameters follows from the general expression for the absorption probability of the ultrashort laser pulse given by the equation [2]:

$$W(\tau) = \frac{c}{4\pi^2} \int_0^\infty \frac{\sigma(\omega')|E(\omega', \omega, \tau)|^2}{h\omega'} d\omega',$$  \hspace{1cm} (1)

here absorption cross section is given by the formula

$$\sigma(\omega') = \frac{2\pi^2 e^2}{mc} f_0 G(\omega'),$$

$G(\omega')$ is standard line shape of a spectral line, $f_0$ is oscillator strength, $E(\omega', \omega, \tau)$ is the Fourier transform of USP electric field which for Gaussian envelope takes the form

$$|E(\omega')|^2 \approx \frac{\pi}{2} E_0^2 \exp \left\{ - \left( \omega - \omega' \right)^2 \tau^2 \right\},$$  \hspace{1cm} (2)

here $\omega$, $\tau$ are the carrier frequency and the duration of an electromagnetic pulse, $E_0$ is the amplitude of the electric field in a pulse.

2. Transition probabilities from ground atomic states for different broadening mechanisms

We will consider below the most famous mechanisms of spectral line broadening by plasmas, namely Holtsmark and Doppler broadening.

2.1. Holtsmark static broadening

Holtsmark broadening of spectral lines is due to the Stark effect on the atomic structure under the action of static ion plasma microfield. It typical value for the linear Stark effect is equal to the broadening by the averaged value of electric field determined by ion density by the well known relationship

$$\Delta = C_{st} N^{2/3}$$

where $C_{st}$ is the Stark constant for a specific radiative transitions in neutral hydrogen or hydrogen-like ions, $N$ being the ion density.
Well known line shape in static theory is described by Holtsmark distribution function $H(\beta)$, $\beta = F/F_0$.

$$H(\beta) = \frac{2}{\pi} \int_0^\infty x \cdot \sin(\beta x) \cdot \exp\left(-x^{3/2}\right) dx$$

Substituting the Holtsmark line shape into general equation (1) we obtain the absorption line shape for USP in Holtsmark plasma microfield:

$$W_{Hol} \approx \frac{\sqrt{\pi}}{4} f_0 E_0^2 \frac{\tau}{\omega_0 \Delta} J_{Hol}(\Delta \tau, \omega - \omega_0)$$

$$J_{Hol}(\alpha, \beta) \approx \int_0^\infty x \exp\left(-x^{3/2} - x^2/4\alpha^2\right) \left[x \frac{\cos(\beta x)}{2\alpha^2} + \beta \sin(\beta x)\right] dx,$$

here the parameters are equal to

$$\alpha = \Delta \tau, \quad \beta = (\omega - \omega_0)/\Delta$$

The general absorption line shapes for different pulse durations is presented on figure 1 for Ly-beta line.

![Figure 1](image)

**Figure 1.** Ly\(_{\beta}\) excitation probability vs dimensionless frequency shifts $\beta$ for different values of pulse duration parameter $\alpha = \Delta \tau$: solid line $- \alpha = 0.4$, dotted line $- \alpha = 0.5$, dashed line $- \alpha = 1$, dotted-dashed line $\alpha = 10$.

One can see that the well-known dips in the line centers disappear with the decrease of pulse duration.
The specific example of the Hydrogen Ly-beta line shape in the plasma with standard parameters $N_e=10^{18}$ cm$^{-3}$ and electron temperature $T_e=1$ eV is presented on figure 2 for different pulse durations (cycles number $N$).

![Figure 2](image)

**Figure 2.** Excitation probability spectrum for various pulse duration: solid line – $N=500$ cycles, dotted line – $N=300$ cycles, dashed line – $N=200$ cycles of carrier frequency.

The effect of pulse duration on the absorption probabilities is shown on figure 3 where the probability $W$ is presented as a function of pulse duration. It is clear that for the long pulse durations the probability follows to the linear dependence on pulse duration corresponding to the standard representation for the absorption per unit time.
Figure 3. Dependence of $Ly_\beta$ excitation probability on pulse duration (cycles number $N$) for different detuning of carrier frequency. Solid line $-\beta=0$, dotted line $-\beta=0.5$, dashed line $-\beta=1$, dotted-dashed line $-\beta=2$.

2.2. Doppler broadening

Doppler broadening mechanism is typical for highly charged ions in high temperature plasmas. The line shapes of H-like ions in their radiation transitions to the ground states depend also on the thin structure of H-like ions satisfying following relationships:

$$\Delta f_\omega \approx \frac{\alpha^2 Z^4}{4n^3},$$

(4)

where $\alpha \approx 1/137$ is the fine structure constant, $n$ is the principal quantum number of the upper level.

The Doppler broadening is of specific interest for USP absorption shapes due to the identical dependencies between Fourier components of Doppler line shape and Gaussian line shape of the USP electric field. It results in analytical expression for the absorption line shape following from the general equation (1) [3]:

$$W_{n_j} \approx \frac{\pi}{8} f_{n_j} \frac{E_0^2}{\omega_{n_j} \Delta \omega_D^2} F(y, \rho_{n_j}), \quad F(y, \rho) = \frac{y^2}{\sqrt{1+y^2}} \exp\left\{-\rho^2 \frac{y^2}{1+y^2}\right\}$$

(5)

Where the following parameters are introduced:

$$y = \sqrt{2} \Delta \omega_D \tau, \quad \rho_{n_j} = \frac{\omega - \omega_{n_j}}{\sqrt{2} \Delta \omega_D}$$

(6)

here $\omega_{n_j}$ is the own frequency of the transition $1s \rightarrow np_{j}$.
It is obvious that the total probability of excitation of the $np$-state is

$$W_n = W_{n,1/2} + W_{n,3/2}.$$  \hspace{1cm} (7)

Hereafter we will analyze the probability of excitation of a hydrogen-like ion normalized to the squared amplitude of the electric field strength $E_0^2$ as a function of the duration and the carrier frequency of a pulse: $\tilde{W} = W/E_0^2$.

The absorption spectra of H-like ion are shown on figure 4 with account of thin structure. It follows that the dip between fine structure components disappeared with the decrease of pulse duration.

![Figure 4](image_url)

**Figure 4.** Normalized spectrum of excitation of the transition in a hydrogen-like ion with the nucleus charge $Z = 13$ by pulses of different durations: solid curve - $\tau = 0.24$ fs, dotted curve - $\tau = 0.48$ fs, dashed curve - $\tau = 2.4$ fs.

The effect of pulse duration on absorption probabilities is shown on figure 5 for different ion charges for carrier frequency equal to: $\omega = (\omega_{j=1/2} + \omega_{j=3/2})/2$. It is seen a strong dependence of the function $\tilde{W}(\tau)$ on the ion charge.
3. Absorption probabilities for USP in optically dense media

Let us consider the effect of pulse duration on the UPS penetration into optically thick media. The optical thickness is described by the standard equation:

\[ \Lambda(L) \approx \kappa_0 L \approx 2\pi N_0 \kappa^2 L \frac{A}{\Gamma} \]  

(8)

here \( \Delta \) is line-width \( (\Delta \approx v\omega_c/c - \text{Doppler}; \Delta \approx N v \sigma - \text{Lorenz}) \), \( \omega_c \) is the central frequency of transition, \( \omega \) is carrier frequency of the pulse, \( \beta = (w - \omega) / \Delta \), \( w \) is current frequency, \( \delta = (\omega_c - \omega) / \Delta \), \( \alpha = \Delta \tau \).

By substitution of the optical thickness (8) into general expression for the reduction of the pulse intensity in the dense media one can arrive to the expression of the electric field intensity in the dense media as following:

\[ E^2(\Lambda, \alpha, \beta, \delta) = \alpha^2 \exp \left\{ - \left( \beta + \delta \right)^2 \alpha^2 - \Lambda J_{L,D}(\beta) \right\}, \]  

(9)

here

\[ J_D(\beta) = \exp \left(-\beta^2/2\right) / \sqrt{2\pi}, \quad J_L(\beta) = 1 / \left[ \pi \left(1 + \beta^2\right) \right]. \]

The general results for the spectra of field squared in the dense media are illustrated by figure 6.
Figure 6. Spectra of field squared inside dense medium for Lorenz lineshape for \( \alpha=1, \delta=0 \) and different optical thickness \( \Lambda \): solid line – \( \Lambda=100 \), dotted line – \( \Lambda=30 \), dashed line – \( \Lambda=10 \).

It is simple to obtain general expression for the USP absorption probability as a function of media optical depth following equation (8) and general expression (1). The result has the form:

\[
W(\Lambda,\alpha,\delta) \propto \alpha^2 \int \exp \left[ -\left( \beta + \delta \right)^2 + \alpha J_{L.D}(\beta) J_{L.D}(\beta) \right] d\beta
\]

(10)

The dependence of the USP absorption probability on the medium optical depths is presented on figure 7 for different pulse durations.

Figure 7. Excitation probability vs optical thickness for \( \delta=0 \) and different pulse duration parameter \( \alpha=\Delta \tau \): solid line – \( \alpha = 2 \), dotted line – \( \alpha = 1 \), dotted-dashed line – \( \alpha = 0.02 \), dashed line – \( \alpha = 0.1 \).
It is clear seen a strong effect of pulse duration on the absorption probability due to its penetration into the media.

4. Conclusion
General scheme of USP absorption spectroscopy looks as following: (1) excitation from ground atomic states by USP can be considered as ground state population transfer to the excited state (practically redefinition of ground state); (2) typical situation: large optical thickness for radiative transitions from ground states and transparency for ones between excited states; (3) the excitation from ground states by USP is accompanied by electron collisions excitation transfer between excited atomic states, the effect providing increase of radiation emission in optically thin spectral lines corresponding to transitions between excited states. Combined actions of USP and electron collisions results in the transfer of effective ground state to the excited atomic levels resulting in sharp increase of latter populations. It is especially clear for atomic systems in thermodynamic equilibrium. Here the Boltzman exponential factor and, as a result, levels populations increase sharply after transfer of the ground energy level to the excited one under the action of USP.

References
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