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The Pulse Intensity–Duration Conjecture: Evidence from free-electron lasers

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The recent remark by Mourou and Tajima, Science **331**, 41 (2011), on the intensity of the driver laser pulse and the duration of the created pulse that higher driver beam intensities are needed to reach shorter pulses of radiation remains a conjecture without clear theoretical reasoning so far. Here we discuss the observations leading to the conjecture and offer its extension to the case of relativistic electron bunches as the laser's radiating medium (free-electron laser). The idea is further extended towards the regime of vacuum non-linearities.

Subject Index A01, G20

1. Introduction

In recent years large-scale laser facilities, like the National Ignition Facility (NIF) [1], Laser Megajoule [2], and the Extreme Light Infrastructure (ELI) [3], delivering unprecedented power, have been of increasing interest. They are able to deliver energies on the kJ level in timeframes as short as 10 fs. These high power levels are not only of interest because of the corresponding high fields, but there also now emerges a case for them being the key to the creation of the shortest possible pulses.

This link between the driver intensity and the resulting pulse duration has been suggested by Mourou and Tajima [4,5] and can be stated as: "To decrease the achievable pulse duration, we must first increase the intensity of the driving laser." This is not the same as the converse trivial statement "to increase the achievable peak intensity of a pulse for a given energy, we must shorten pulse duration" [4,5].

In this paper we give an overview of the major steps in the development of solid-state lasers leading to the aforementioned conjecture (Sect. 2). In Sect. 3 we discuss electron beam non-linearities in terms of the free-electron laser (FEL) and find a scaling supporting the conjecture over several orders of magnitude. Finally we extend the discussion to the regime of vacuum non-linearities and their possible applications (Sect. 4).

2. Solid-state lasers

2.1. The non-linear regime

The first laser developed by Maiman [6] reached a pump intensity on the kW cm^{-2} level while producing pulses with durations in the microsecond region. The laser technique has then been significantly improved by the concept of Q-switching [7], increasing the internal intensity to the MW cm^{-2} level while allowing for the production of nanosecond pulses. The first demonstration of mode-locking in ruby [8], relying on the fast transparency recovery of the dye, and therefore the switch to the non-linear bound electron regime, allowed further increase in the peak intensities to the range of GW cm^{-2} resulting in pulse durations in the picosecond regime. In addition, the combination of dye amplifying materials with the colliding pulse mode-locking technique allowed for the further reduction of pulse durations to 100 fs [9] and even 27 fs [10].

The switch from relying on the dye resonance as the mode-locking mechanism to the intensity-dependent index of refraction of the amplifier medium led to Kerr-lens mode-locking (KLM) [11]. The out-of-resonance operation requires higher driving intensities on the order of TW cm^{-2} leading to pulse durations about 10 fs [12–14]. In this regime the pulse duration and its associated, required bandwidth are limited by the bandwidth of the amplifying material.

To further reduce the pulse duration, an amplification mechanism providing a larger bandwidth was required. One solution was the use of gas-filled hollow fibers [15] in combination with chirped mirrors, allowing for a pulse duration of 4.5 fs [16] while only requiring an internal pulse intensity of 10^{14} W cm^{-2}. Another approach relying on molecular phase modulation in gases resulted in a pulse duration of 3.8 fs at an internal intensity of only 10^{12} W cm^{-2}[17].

A further broadening of the bandwidth could be reached by using high harmonic generation (HHG) [18] in a gas jet. Here, a compressed beam is focused into a gas jet creating harmonics up to the cut-off frequency [19,20]. Using this technique the pulse duration could be reduced down to 100 as [21,22] and even 80 as [23].

2.2. The relativistic regime

To reach even shorter pulse durations one had to switch to the relativistic regime where electrons moving in the laser field become relativistic during their oscillation. Therefore, the required laser intensity for a wavelength of 1 μm is greater than 10^{18} W cm^{-2}, which is possible when using chirped pulse amplification (CPA) [24] and optical parametric chirped pulse amplification (OPCPA) [25]. In this regime a laser impinging on a surface causes the surface to oscillate in and out at relativistic velocity causing a periodic modulation of the impinging light resulting in high harmonics [26,27]. This relativistic high harmonic generation opens up a much broader spectrum that, in contrast to non-relativistic HHG, is not limited by the cut-off frequency [26,28].

The concept of the relativistic mirror above can be extended by focusing a pulse down to a spotsize of λ^2. In this so-called λ^3-regime [29] the relativistic mirror gets deformed by the impinging Gaussian beam. Particle-in-cell (PIC) simulations show that the motion of the mirror compresses the sub-cycle pulse and guides it in a specific direction. The resulting pulse duration scales as

$$\tau = \frac{600\,\text{as}}{a_0}, \tag{1}$$

with a_0 being the normalized vector potential of the laser. Assuming an impinging laser intensity of 10^{22} W cm^{-2} over the regime where this setup is possible, this could result in a compressed pulse

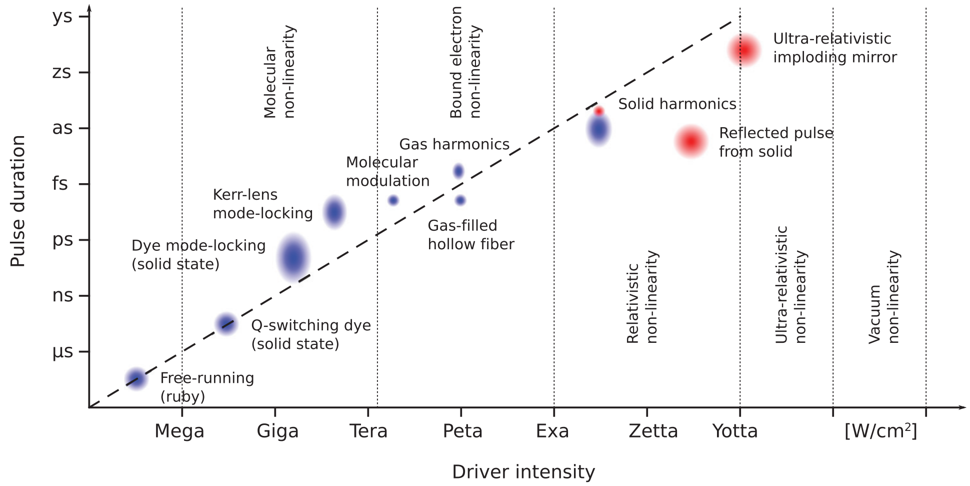

Fig. 1. Visualization of the Pulse Intensity–Duration Conjecture suggested by Mourou and Tajima [4,5]. The pulse duration and the driver intensity show an inverse linear dependence covering 15 orders of magnitude for experimental results (blue) and 18 orders of magnitude when including theoretical and numerical results (red).

duration of a few attoseconds. The authors of this concept also simulated the creation of thin electron sheets with a γ of a few tens and a duration of attoseconds. These electron sheets could be used to produce coherent beams of X-rays or even γ-rays by Thomson scattering. Another approach suggested by [30] is the so-called “relativistic flying mirror,” also based on a thin electron sheet that could be used for pulse compression.

2.3. *The ultra-relativistic regime*

To further compress the laser pulse, even higher frequencies and therefore higher mirror densities are needed to ensure a coherent reflection resulting, for example, in γ-rays. The required densities are of the order of $10^{27}\,\mathrm{cm}^{-3}$ and could be reached by an imploding flying mirror. An implosion reducing the mirror size by a factor of ten in each dimension would already allow a thousand-fold increase of the density. This might be possible by using pulse energies in the MJ range with ultra-relativistic intensities of $10^{24}\,\mathrm{W\,cm}^{-2}$ focused on a partial shell of a concave spherical target. This concept of the ultra-relativistic flying mirror [31] in combination with the imploding target is capable of reflecting a coherent 10 keV pulse into a γ-ray with only 100 yoctoseconds (ys) pulse duration.

2.4. *The conjecture*

Gathering all these discussed data points in one diagram (Fig. 1) leads to a remarkable result: in a double logarithmic plot we find an inverse linear relation between the intensity of the driving pulse and the duration of the created pulse covering 18 orders of magnitude. Excluding theoretically possible but not yet tested concepts still yields a relationship covering 15 orders of magnitude. One has to note that this relationship is only found upon optimization for the shortest possible pulse duration for each driver intensity. However, due to the different physical principles dominating the behavior in the various regimes no analytical model has been offered so far for the conjecture covering all these orders of magnitude.

3. Electron beam non-linearities

The lasers discussed up to now were all driven by solid-state lasers and associated specific setups. We now wish to consider lasers whose emission mechanism is based on relativistic electron bunches as the radiating medium. In other words we will examine if the stiffness of the relativistic electron bunch contributes to the pulse duration of this type of laser. The advantage of this regime is the wide parameter range that can be covered by varying the electron parameters, allowing for a single theory covering several orders of magnitude in the intensity and pulse duration.

Such an example is given by the free-electron laser. Its light generation process is based on a collective instability arising when an electron beam propagates through the periodic magnetic field of an undulator and interacts with a copropagating light wave. The collective instability leads to microbunching, i.e. the formation of a substructure in the electron beam on the scale of the radiated wavelength λ, allowing for coherent emission. Since the emission process is based on the formation of this substructure in the electron bunch, the bandwidth is governed by the electron beam intensity I_{beam} and the formation efficiency of the collective instability characterized by the Pierce parameter ρ [32].

The shortest possible duration, i.e. the inverse bandwidth, of a single radiation spike is given by [33]

$$\tau = \frac{\sqrt{\pi}}{\sigma_\omega}, \tag{2}$$

using the bandwidth σ_ω defined as [34]

$$\sigma_\omega(z) = \sqrt{\frac{3\sqrt{3}\rho}{k_u z}}\omega_l, \tag{3}$$

with $k_u = 2\pi/\lambda_u$ the undulator wavenumber of an undulator with period length λ_u, z the longitudinal position inside the undulator, and ω_l the resonant frequency.

To minimize the free parameters, we limit our discussion to a system operating with a fully modulated electron beam, i.e. at saturation $z_{\text{sat}} \approx \lambda_u/\rho$ [35], allowing for maximum coherence of the emitted light. In addition, we restrict our discussion to the “natural” beam size of the setup, i.e. the constant matched beam size of the undulator, and assume the setup parameters to be optimized for a short pulse duration. Using the dependence of the Pierce parameter on the current and the normalized energy $\rho \propto I^{1/3}\gamma^{-1}$, and the proportionality between the resonant frequency and the normalized energy $\omega_l \propto \gamma^2$, the resulting pulse duration scales as

$$\tau \propto I^{-1/3}\gamma^{-1}, \tag{4}$$

with I being the electron beam current and γ the normalized electron energy. Here the general relationship that the pulse duration drops with increasing electron energy and current, which are directly proportional to the electron beam intensity

$$I_{\text{beam}} = \frac{\gamma m_e c^2 I}{2\pi\sigma_r^2 e} \propto \gamma I, \tag{5}$$

can already be seen. Here, m_e is the electron mass, e the elementary charge, c the speed of light, and σ_r the beam radius. This proportionality is a first hint that an FEL shows a scaling supporting the Conjecture.

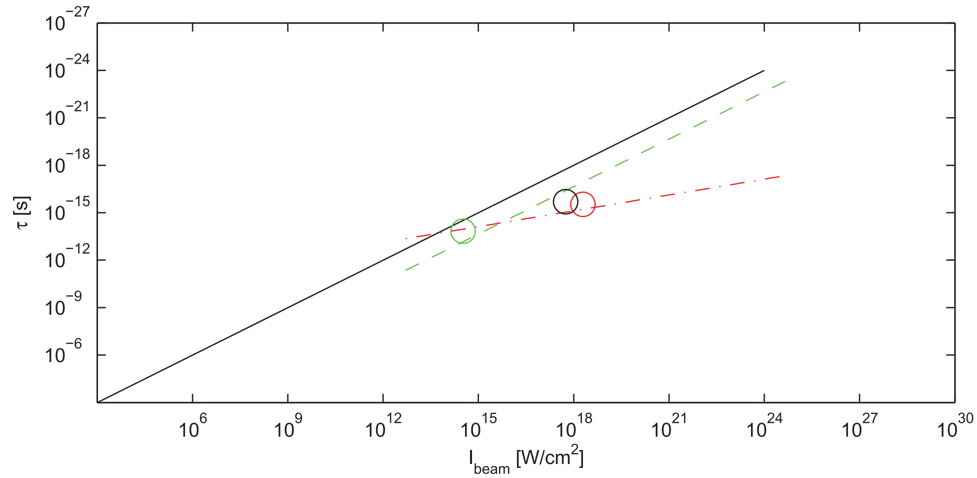

Fig. 2. Comparison of the Conjecture suggested by Mourou and Tajima (solid black line), the here-derived dependence of the pulse duration on the beam intensity for a fixed current (dashed green line) and a fixed energy (dash-dotted red line), and the data points of LCLS (red circle), FLASH (green circle), and the X-FEL (black circle).

Using Eqs. (4) and (5) the duration of the created photon pulse is related to the electron beam intensity as

$$\tau \propto \gamma^{-1} \propto I_{\mathrm{beam}}^{-1}, \tag{6}$$

when only varying the energy for a fixed current, due to the linear dependence of the bandwidth on the electron energy. On the other hand, another dependence emerges:

$$\tau \propto I^{-1/3} \propto I_{\mathrm{beam}}^{-1/3}, \tag{7}$$

for a fixed normalized energy and variation of the current. The exact dependence of the pulse duration on the setup parameters is derived in the Appendix.

Figure 2 shows the comparison of the scaling obtained from the analysis of systems driven by solid-state lasers, and the derived dependencies of the FEL pulse duration on the electron beam intensity. In the case of the fixed current the dependence is inverse linear, as in the Conjecture. This may be understood as follows: the assumption of a fixed current represents the situation of a medium that gets harder to bend due to the increasing energy, and results in shorter pulses. Now we see that the Conjecture emerges when we have related the pulse duration and its functional dependence to the energy of the electron beam (i.e., the γ factor), as the latter implies the stiffness of the medium. The slope of the red line in Fig. 2, i.e. the case of a fixed energy and variation of the current, can be related to a simultaneous change of the density of the medium and the driver intensity due to the increasing current (and particle number in the beam) resulting in a different scaling. This is reasonable, as the "spring constant" in this case is not directly scaled. This leads to the conclusion that the scaling of the Conjecture is indeed due to the rigidity of the pulse-generating matter, and that high driver intensities are needed either to reach these rigidities, as in the case of an FEL, or to excite more rigid non-linearities, as in the cases discussed in the original Conjecture.

Besides the different scaling, both cases support the Conjecture, since they lead to the same result: to reduce the pulse duration, one has to increase the driver intensity.

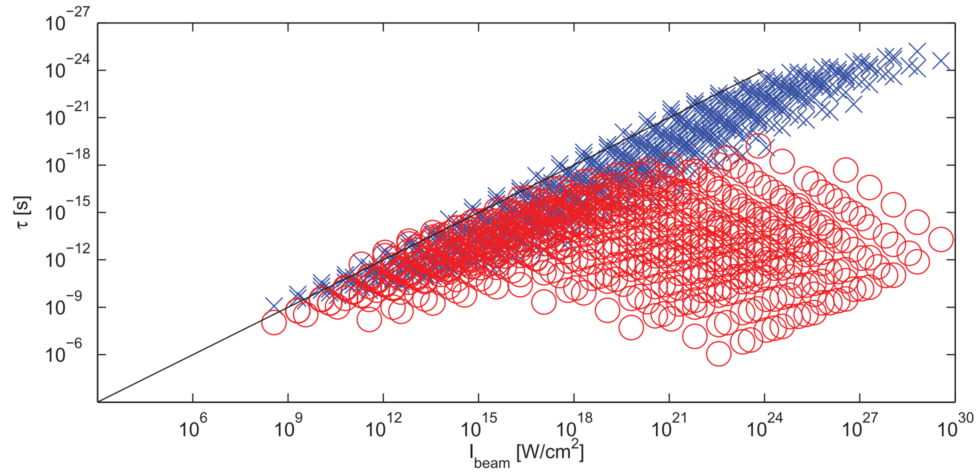

Fig. 3. Pulse durations for different beam intensities according to Eq. (4) (blue crosses), and according to Eqs. 2 and 3 calculated using the fitting formula of M. Xie [36] taking diffraction and emittance into account (red circles). The following parameters were varied: $\gamma = 10^1 \rightarrow 10^{12}$, $\lambda_u = 10^{-3} \rightarrow 10^0$ m, $K = 10^{-2} \rightarrow 10^1$, and $I = 10^2 \rightarrow 10^6$ A. The normalized emittance was set to $\epsilon_n = 10^{-6}$ m rad. The black line is again the Conjecture as a reference.

An interesting result of the Conjecture is the product of pulse duration and intensity, the coefficient of the graph. It is the constant of the Conjecture for solid-state lasers, with a value of 1 J cm^{-2}. Lower values of the product are regarded as more favorable (or more efficient), since in that case lower beam intensities are needed to reach shorter pulses.

We can study this for the FEL in the case of a fixed current, resulting in a product of pulse duration and beam intensity of

$$\tau I_{\text{beam}} = \frac{\pi^{1/3}}{2^{1/2}3^{3/4}} \frac{m_e c I_A^{1/3}}{e} \frac{I^{2/3}\left(1+\frac{K^2}{2}\right)}{\epsilon_n^{2/3}\,\mathrm{JJ}^{2/3}\,\lambda_u^{1/3}}, \tag{8}$$

with the Alfvén current $I_A \approx 17\,\text{kA}$, the undulator parameter K, the Bessel function dependent factor $\mathrm{JJ} = \mathrm{J}_0(\zeta) - \mathrm{J}_1(\zeta)$ with $\zeta = K^2/(4+2K^2)$, and the normalized emittance ϵ_n. The detailed derivation of Eq. (8) is shown in the Appendix. Using the parameters $I = 1$ kA, $\lambda_u = 1$ cm, $K = 1$, and $\epsilon_n = 1$ mm mrad, which are reasonable for current FELs, the product equals approximately 14 J cm^{-2}. In the case of an FEL the product may easily range from 1 J cm^{-2} to 100 J cm^{-2} by adjusting the setup parameters. The fractional power dependence of the product on the current is mainly due to the beam intensity; the scaling with the other parameters like λ_u, K, and ϵ_n are more complex and are a result of the competing effects between the pulse duration and the beam intensity.

Figure 3, where a wide parameter range has been scanned using the derived formulas, shows again that the Conjecture only emerges upon optimization for the shortest possible pulse duration. The blue domain shows the results according to the 1D theory while the red domain takes degrading effects, i.e. diffraction and emittance, into account by using the fitting formula of M. Xie [36]. The parameter scan shows that the upper envelope of the blue domain is represented by the Conjecture line and only degrading effects cause a deviation. However, if we optimize for the efficiency of lasing instead of the pulse shortness, the scaling line nearly goes to the envelope of the bottom of the blue region in Fig. 3.

4. Vacuum non-linearities

4.1. Scaling

Beyond the realms of the previously discussed non-linearities lie the vacuum non-linearities. An even stronger pulse compression could be obtained by using the self-phase modulation in vacuum. The bandwidth of the self-phase modulation scales as

$$\frac{\Delta\omega}{\omega} \approx \frac{Ln_2}{c}\frac{\mathrm{d}I}{\mathrm{d}t} \propto n_2\frac{E_n}{T^2}, \tag{9}$$

using the carrier frequency ω, the self-phase modulation bandwidth $\Delta\omega$, the non-linear index of refraction n_2, the propagation length L, the driving pulse energy E_n, and its duration T. This shows that, using high input intensities, significant non-linear effects are possible leading to a broad bandwidth and the possibility of extreme compression. This resembles a fact already known from the visible range where only with the availability of picosecond–femtosecond pulses were significant non-linear processes like self-phase modulation or Kerr-lens mode-locking [11] possible.

4.2. Streaking vacuum

Besides the production of extremely short pulses, the exploration of processes on short time scales is also of high interest. An interesting quantity, therefore, is the critical power, defining the limit for self-focusing. It is given by

$$P_{\mathrm{cr}} \approx \frac{\lambda^2}{2\pi n_0 n_2} \sim \mathrm{GW} \tag{10}$$

in the case of the χ_3 non-linearity,

$$P_{\mathrm{cr}} \approx \frac{mc^5}{e^2}\left(\frac{\omega}{\omega_p}\right)^2 \sim 17\left(\frac{\omega}{\omega_p}\right)^2 \mathrm{GW} \tag{11}$$

in the case of the relativistic plasma non-linearity, and

$$P_{\mathrm{cr}} \approx \frac{90}{28}\frac{cE_S^2\lambda^2}{\alpha} \sim 10^{15}\left(\frac{\lambda}{\lambda_{1\mu}}\right)^2 \mathrm{GW} \tag{12}$$

for the vacuum non-linearity. Here we used the Schwinger field E_S, the limit for electron–positron pair production in the vacuum [37–41], and the fine structure constant α. The critical power for self-focusing in vacuum is a factor α^{-6} higher than its equivalent in gas. On the other hand, the ratio between the Keldysh field and the Schwinger field is $E_K/E_S = \alpha^3$, resulting in an equivalent power ratio of α^6. While the Keldysh field is needed to overcome the potential energy of the Rydberg energy W_B over the Bohr radius a_B, the Schwinger field is needed to overcome the potential energy of $2mc^2 = \alpha^{-2}W_B$ over the distance of the Compton length αa_B.

Another important parameter is the Keldysh parameter [42]

$$\gamma_K = \omega\frac{\sqrt{2m_e V_I}}{eE}, \tag{13}$$

using the carrier frequency ω, the ionization energy of the system V_I, and the field strength of the laser E. When the Keldysh parameter is lower than unity ionization is dominated by tunnel ionization and can be treated as non-perturbative, while a Keldysh parameter greater than unity indicates a regime where the ionization is dominated by multi-photon processes. An equivalent parameter can be defined for the vacuum,

$$\gamma_{V\sigma} = \frac{m_\sigma \omega c}{eE} = \frac{1}{a_0}. \tag{14}$$

Here, σ is either e indicating electron processes or q when quark processes are considered [5]. However, for the latter more study is necessary as the process is debated [43]. Similar to the Keldysh parameter this parameter indicates whether non-perturbative $\gamma_{V\sigma} < 1$ or perturbative quantum electrodynamics (QED) $\gamma_{V\sigma} > 1$ is applicable.

This indicates the parameter range allowing for the creation of electron–positron pairs from vacuum with high but still realistically achievable laser intensities by combining a gamma photon with a strong laser field in vacuum [38–40]. This would open up the possibility of vacuum streaking with lasers with zeptosecond time resolution [44].

5. Conclusion

We have studied the underlying mechanism between the laser intensity and resulting pulse length. The suggested Conjecture, we find, is based on the rigidity of the medium that compresses the pulse. Because the rigidity is the key for compression, the stiffer the medium (or setup) is, the shorter the obtained pulse is. We have demonstrated this principle by adopting a setup using an electron beam as the compression medium, i.e. an FEL. This examination in fact confirms the Conjecture for the case of electron beam non-linearities. The higher the energy of the electron beam, the more intense the beam energy is. In turn, the higher the beam intensity is, the shorter the FEL pulse length is, according to Eq. (6). One can extend the road for shorter pulses by adopting the stiffest medium of all, the vacuum (QED vacuum).

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Appendix A. Pulse duration of a free-electron laser at saturation

For the case of a free-electron laser the dependence of the pulse duration at saturation on the setup parameters can be calculated as follows:

The exact scaling of the bandwidth is obtained by evaluating the bandwidth (3) assuming saturation of the amplification process. The saturation distance is approximately given by $z_{\text{sat}} \approx \lambda_u/\rho$ [35] with the undulatory period length λ_u and the Pierce parameter ρ [32]. The Pierce parameter is defined as

$$\rho = \frac{1}{2\gamma}\left[\frac{I}{I_A}\left(\frac{K\,\mathrm{JJ}\lambda_u}{\sqrt{2}2\pi\sigma_r}\right)^2\right]^{1/3}, \tag{A1}$$

using the normalized electron energy γ, the current I, the Alfvén current $I_A \approx 17\,\text{kA}$, the undulator parameter K, the Bessel function dependent factor $\mathrm{JJ} = \mathrm{J}_0(\zeta) - \mathrm{J}_1(\zeta)$ with $\zeta = K^2/(4 + 2K^2)$, and the electron beam size σ_r. Inserting this and the resonance frequency of the FEL $\omega_l = 2\omega_u\gamma^2/(1 + K^2/2)$, with $\omega_u = 2\pi c/\lambda_u$ being the undulator frequency, in the bandwidth equation yields

$$\sigma_\omega = \sqrt{6\sqrt{3}\pi}\left[\frac{I}{I_A}\left(\frac{K\mathrm{JJ}\lambda_u}{\sqrt{2}2\pi\sigma_r}\right)^2\right]^{1/3}\frac{c\gamma}{\lambda_u(1+\frac{K^2}{2})}. \tag{A2}$$

Using the matched beam size of a twofold focusing undulator [45] that can be seen as the natural beam size of the system

$$\sigma_r = \sqrt{\frac{2\epsilon_n}{Kk_u}}, \tag{A3}$$

with the normalized rms emittance ϵ_n, the pulse duration is given by

$$\tau = \frac{1}{\sqrt{6\sqrt{3}}}\left[\frac{I_A}{I}\left(\frac{2\sqrt{2\pi\epsilon_n}}{K^{3/2}\mathrm{JJ}\sqrt{\lambda_u}}\right)^2\right]^{1/3}\frac{\lambda_u(1+\frac{K^2}{2})}{c\gamma}. \tag{A4}$$

The product of the so-determined pulse duration and the beam intensity (5) is the coefficient of the Conjecture (8).

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