Ultimate Energy Densities for Electromagnetic Pulses

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The ultimate electric and magnetic energy densities that can be attained by bandlimited electromagnetic pulses in free space are calculated using an ab initio quantized treatment, and the quantum states of electromagnetic fields that achieve the ultimate energy densities are derived. The ultimate energy densities also provide an experimentally accessible metric for the degree of localization of polychromatic photons.

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Ultrafast lasers have become an indispensable tool in a wide spectrum of science, including nonlinear optics [1, 2], metrology [3], laser fusion [4], biological imaging [5], biological surgery [6], and chemistry [7]. A key to the success of such lasers is the extremely high peak energy density that they can achieve, as the moderate energy of each laser pulse can be confined within femtoseconds or even attoseconds and focused to a micron-sized area. The high energy density strongly enhances light-matter interactions, and is especially crucial to the study of relativistic nonlinear optics [2]. Given the importance of an ultrahigh optical energy density in a broad range of applications, the limit to which one can focus a broadband optical pulse in three spatiotemporal dimensions and maximize the energy density is a fundamental problem.

Localization of electromagnetic pulses has been investigated both in the classical regime [8] and the quantum single-photon regime [9, 10]. While these seminal studies have contributed to our fundamental understanding of electromagnetic energy localization, all of their predictions have not yet been experimentally verified because of the difficulty in implementing their proposed electromagnetic pulse solutions. Most require a spectrum that contains arbitrarily high frequency components [8, 9] and can be exceedingly difficult to realize due to the finite laser gain bandwidth or finite transparency range of optical compo-
The use of a spontaneously emitting atom proposed in Ref. [10] couples the properties of the emitted photon to those of the atom and does not seem to be generalizable to other situations. Moreover, the quantum studies [9, 10] are mainly concerned with the decay of the energy density far away from the center of localization at an instant of time for one photon, and thus are not immediately relevant to the more practical problem of maximizing the energy density at the center of localization for a large number of bandlimited photons.

In this Letter, the ultimate electric and magnetic energy densities that can be attained by bandlimited electromagnetic pulses in free space are calculated using an *ab initio* quantized treatment, and the quantum states that achieve the ultimate densities are derived. By taking into account all degrees of freedom of electromagnetic fields and explicitly limiting the bandwidth of the pulses, our result overcomes all the shortcomings of the aforementioned studies and is more applicable to experimental situations. Measuring the energy densities at a fixed point is also considerably easier experimentally than measuring the decay of the energy density at an instant of time, so the maximum achievable energy densities can be used as an alternative and more accessible metric for the degree of localization of polychromatic photons. Most importantly, the ultimate energy densities impose a fundamental limit to which a bandlimited optical pulse can be focused spatially and temporally, so the presented result should prove useful for designing ultrafast optics experiments.

The procedure of calculating the maximum energy densities is similar to the one used to calculate the multiphoton absorption rate limit for monochromatic light in Ref. [11], except that here we generalize the procedure to polychromatic light, such that all degrees of freedom are taken into account and the treatment can be considered *ab initio*. We also calculate the maximum magnetic energy density and the corresponding quantum state, as the magnetic field can also play a significant role in relativistic nonlinear optics [2].

We first derive the ultimate electric energy density, since it is more important for most applications. Consider the quantized electric field operator in free space [12]:

\[
\hat{E}(r, t) = \frac{i}{(2\pi)^{3/2}} \sum_{\sigma} \int d^3k \left( \frac{\hbar \omega}{2\epsilon_0} \right)^{1/2} \epsilon(k, \sigma) \hat{a}(k, \sigma) e^{i(k \cdot r - \omega t)} + \text{H.c.,}
\]

where \( \sigma \) denotes the two transverse polarizations, \( \epsilon(k, \sigma) \) is the unit electric-field polarization vector, \( \omega = ck = c(k_x^2 + k_y^2 + k_z^2)^{1/2} \) is the frequency, \( \hat{a}(k, \sigma) \) is the annihilation operator satisfying the commutation relation \([\hat{a}(k, \sigma), \hat{a}^\dagger(k', \sigma')] = \delta^3(k - k') \delta_{\sigma\sigma'}, \) and H.c. is the Hermitian conjugate. To impose a limit on the bandwidth, it is necessary to describe
the optical modes in terms of the frequency variable. This can be done by changing the momentum-space coordinates \((k_x, k_y, k_z)\) to normalized spherical coordinates \((\Omega, \theta, \phi)\):

\[
\omega = \omega_0 \Omega, \quad k_x = k_0 \Omega \sin \theta \cos \phi, \quad k_y = k_0 \Omega \sin \theta \sin \phi, \quad k_z = k_0 \Omega \cos \theta, \\
dk_x dk_y dk_z = d\Omega d\theta d\phi \left(k_0^3 \Omega^2 \sin \theta\right), \quad \hat{a}(k, \sigma) = \hat{a}(\Omega, \theta, \phi, \sigma) \left(k_0^3 \Omega^2 \sin \theta\right)^{-1/2}. \tag{2}
\]

where \(\omega_0\) is a normalization frequency, \(k_0 \equiv \omega_0/c \equiv 2\pi/\lambda_0\), and the annihilation operator has been renormalized so that its commutator is \([\hat{a}(\Omega, \theta, \phi, \sigma), \hat{a}^\dagger(\Omega', \theta', \phi', \sigma')] = \delta(\Omega - \Omega') \delta(\theta - \theta') \delta(\phi - \phi') \delta_{\sigma \sigma'}\). The positive-frequency electric field becomes

\[
\hat{E}^+(r, t) = i \left(\frac{\hbar \omega_0}{2e_0 \lambda_0^3}\right)^{1/2} \int_0^\infty d\Omega \int_0^\pi d\theta \int_0^{2\pi} d\phi \left(\Omega^3 \sin \theta\right)^{1/2} \varepsilon(\theta, \phi, \sigma) \hat{a}(\Omega, \theta, \phi, \sigma) e^{i k \cdot r - i \omega t}. \tag{3}
\]

In the continuous Fock space representation \[12\], the \(N\)-photon momentum eigenstate is given by

\[
|\Omega_1, \theta_1, \phi_1, \sigma_1, \ldots, \Omega_N, \theta_N, \phi_N, \sigma_N \rangle \equiv \frac{1}{\sqrt{N!}} \prod_{n=1}^{N} \hat{a}^\dagger(\Omega_n, \theta_n, \phi_n, \sigma_n) |0\rangle, \tag{4}
\]

and the identity operator is

\[
\hat{1} = \sum_{N=0}^{\infty} |N\rangle \langle N|, \tag{5}
\]

\[
|N\rangle \langle N| = \sum_{\sigma_1, \ldots, \sigma_N} \int d\Omega_1 d\theta_1 d\phi_1 \ldots d\Omega_N d\theta_N d\phi_N \\
x |\Omega_1, \theta_1, \phi_1, \sigma_1, \ldots, \Omega_N, \theta_N, \phi_N, \sigma_N \rangle \langle \Omega_1, \theta_1, \phi_1, \sigma_1, \ldots, \Omega_N, \theta_N, \phi_N, \sigma_N|.
\tag{6}
\]

An arbitrary quantum state of electromagnetic fields can thus be expressed as

\[
|\Psi\rangle = \sum_{N=0}^{\infty} C_N |N\rangle, \quad C_N \equiv \langle N | \Psi \rangle, \tag{7}
\]

and a Fock state as

\[
|N\rangle = \sum_{\sigma_1, \ldots, \sigma_N} \int d\Omega_1 d\theta_1 d\phi_1 \ldots d\Omega_N d\theta_N d\phi_N \Phi_N(\Omega_1, \theta_1, \phi_1, \sigma_1, \ldots, \Omega_N, \theta_N, \phi_N, \sigma_N) \\
x |\Omega_1, \theta_1, \phi_1, \sigma_1, \ldots, \Omega_N, \theta_N, \phi_N, \sigma_N \rangle.
\tag{8}
\]
where

\[ \Phi_N(\Omega_1, \theta_1, \phi_1, \sigma_1, \ldots, \Omega_N, \theta_N, \phi_N, \sigma_N) \equiv \langle \Omega_1, \theta_1, \phi_1, \sigma_1, \ldots, \Omega_N, \theta_N, \phi_N, \sigma_N | N \rangle \]  

(9)

is the \( N \)-photon momentum-space probability amplitude, which must satisfy the normaliza-
tion condition:

\[
\sum_{\sigma_1, \ldots, \sigma_N} \int d\Omega_1 d\theta_1 d\phi_1 \cdots d\Omega_N d\theta_N d\phi_N \left| \Phi_N(\Omega_1, \theta_1, \phi_1, \sigma_1, \ldots, \Omega_N, \theta_N, \phi_N, \sigma_N) \right|^2 = 1, \tag{10}
\]

and the symmetrization condition:

\[
\Phi_N(\ldots, \Omega_n, \theta_n, \phi_n, \sigma_n, \ldots, \Omega_m, \theta_m, \phi_m, \sigma_m, \ldots) = \Phi_N(\ldots, \Omega_m, \theta_m, \phi_m, \sigma_m, \ldots, \Omega_n, \theta_n, \phi_n, \sigma_n, \ldots) \text{ for any } n \text{ and } m. \tag{11}
\]

To impose a limited bandwidth (\( \alpha \leq \Omega \leq \beta \)) on the electromagnetic fields, we require the probability of photons existing outside the bandwidth to vanish:

\[
\Phi_N(\Omega_1, \theta_1, \phi_1, \sigma_1, \ldots, \Omega_N, \theta_N, \phi_N, \sigma_N) = 0 \text{ for any } \Omega_n < \alpha \text{ or } \Omega_n > \beta. \tag{12}
\]

With the theoretical framework put forth, we now proceed to calculate a bound on the electric energy density (minus the zero-point energy density) given by

\[
U_e \equiv \left\langle \frac{\varepsilon_0}{2} \hat{\mathbf{E}} \cdot \hat{\mathbf{E}} \right\rangle = \left\langle \varepsilon_0 \mathbf{\hat{E}}^{(-)} \cdot \mathbf{\hat{E}}^{(+)} \right\rangle. \tag{13}
\]

A bound on the electric energy density is equivalent to a bound on the energy density for one component of the electric field:

\[
U'_e \equiv \left\langle \frac{\varepsilon_0}{2} \left( \mathbf{p} \cdot \mathbf{\hat{E}} \right)^2 \right\rangle = \varepsilon_0 \left\langle \left( \mathbf{p} \cdot \mathbf{\hat{E}}^{(-)} \right) \left( \mathbf{p} \cdot \mathbf{\hat{E}}^{(+)} \right) \right\rangle, \tag{14}
\]

where \( \mathbf{p} \) is an arbitrary real unit vector, because \( U_e \) and \( U'_e \) are equivalent if we choose \( \mathbf{p} \) to be parallel to the electric field. In terms of the momentum-space representation,

\[
U'_e = \frac{\hbar \omega_0}{2 \lambda_0^3} \sum_{N=0}^{\infty} |C_N|^2 N \sum_{\sigma_2, \ldots, \sigma_N} \int d\Omega_2 d\theta_2 d\phi_2 \cdots d\Omega_N d\theta_N d\phi_N
\times \left| \sum_{\sigma} \int_{\alpha}^{\beta} d\Omega \int_{0}^{\pi} d\theta \int_{0}^{2\pi} d\phi \left[ i \left( \Omega^3 \sin \theta \right)^{1/2} \mathbf{p} \cdot \mathbf{\varepsilon}(\theta, \phi, \sigma) e^{ik \mathbf{r} - i\omega t} \right] \right|^2 \Phi_N(\Omega, \theta, \phi, \sigma, \Omega_2, \theta_2, \phi_2, \sigma_2, \ldots, \Omega_N, \theta_N, \phi_N, \sigma_N)^2, \tag{15}
\]
where the symmetric property of $\Phi_N$ given by Eq. (11) is used. By virtue of the Schwarz’s inequality and the normalization condition given by Eq. (10),

$$U'_e \leq \frac{\hbar \omega_0}{2 \lambda_0^3} \sum_{N=0}^{\infty} |C_N|^2 N \sum_{\sigma_1, \ldots, \sigma_N} \int d\Omega_1 d\theta_1 d\phi_1 \ldots d\Omega_N d\theta_N d\phi_N \times |\Phi_N(\Omega_1, \theta_1, \phi_1, \sigma_1, \ldots, \Omega_N, \theta_N, \phi_N, \sigma_N)|^2 \times \sum_\sigma \int_\alpha^\beta d\Omega \int_0^\pi d\theta \int_0^{2\pi} d\phi (\Omega^3 \sin \theta) |p \cdot \varepsilon(\theta, \phi, \sigma)|^2$$

$$= \frac{\pi \langle N \rangle \hbar \omega_0}{3 \lambda_0^3} (\beta^4 - \alpha^4), \quad (16)$$

where $\langle N \rangle \equiv \sum_N |C_N|^2 N$ is the average photon number. As expected, the bound on $U'_e$ does not depend on $p$, and is therefore also applicable to the total electric energy density.

Defining the actual lower and upper frequencies as $\omega_1 = \alpha \omega_0$ and $\omega_2 = \beta \omega_0$, respectively, and the corresponding wavelengths as $\lambda_{1,2} = 2\pi c/\omega_{1,2}$, we obtain the central result of this Letter:

$$\langle : \frac{\varepsilon_0 c}{2} \hat{E} \cdot \hat{E} : \rangle \leq \frac{\pi}{3} \langle N \rangle \left( \frac{\hbar \omega_2}{\lambda_0^3} - \frac{\hbar \omega_1}{\lambda_0^3} \right). \quad (17)$$

This simple expression agrees with the intuition that the ultimate energy density is limited by the maximum energy of photons ($\langle N \rangle \hbar \omega_2$) divided by the smallest volume that the photons can occupy ($\lambda_0^3$).

In the limit of a small bandwidth compared to the center frequency, we can let $\Delta \omega \equiv \omega_2 - \omega_1$, $\omega_0 = (\omega_1 + \omega_2)/2$, $\Delta \omega \ll \omega_0$, and obtain

$$\langle : \frac{\varepsilon_0 c}{2} \hat{E} \cdot \hat{E} : \rangle \approx \frac{2}{3} \langle N \rangle \frac{\hbar \omega_0 \Delta \omega}{\lambda_0^3}, \quad (18)$$

which is a bound on the peak intensity in the slowly-varying envelope regime, and again agrees with the intuition that the highest intensity is achieved when the mean energy of the photons is focused to their minimum pulse width ($2\pi/\Delta \omega$) and beam size ($\lambda_0^2$). This approximate bound also agrees with that derived in Ref. [11], where the monochromatic approximation is made at the beginning. Beyond the slowly-varying envelope regime, the exact bound given by Eq. (17) depends on the upper frequency to the fourth power, underlying the importance of high-frequency components in maximizing the energy density, as they have a higher energy as well as a smaller localization volume.

The use of the Schwarz’s inequality is reminiscent of the matched filter concept in communication theory [13]. In Eq. (15), the $N$-photon amplitude can be regarded as the input
signal, and the expression in square brackets can be regarded as a filter transfer function in
the measurement of the electric field. An \( N \)-photon amplitude that achieves the Schwarz
upper bound is one that is linearly dependent on the square-bracketed expression, or in
other words, when the input signal matches the filter. Assuming a factorizable \( \Phi_N \), the
following \( N \)-photon amplitude that achieves the ultimate electric energy density can then
be obtained:

\[
\Phi_N = \prod_{n=1}^{N} f_e(\Omega_n, \theta_n, \phi_n, \sigma_n),
\]

\[
f_e = \begin{cases} 
- i C^{-1/2}(\Omega^3 \sin \theta)^{1/2} \mathbf{p} \cdot \varepsilon^*(\theta, \phi, \sigma)e^{-i k \cdot r_0 + i \omega t_0} & \text{for } \alpha \leq \Omega_n \leq \beta, \\
0 & \text{otherwise,}
\end{cases}
\]

\[
C = \frac{2\pi}{3} (\beta^4 - \alpha^4).
\]

(19)

This state produces the maximum electric energy density at \((r_0, t_0)\), with the electric field
at \((r_0, t_0)\) polarized along \( \mathbf{p} \).

To apply the above result to the classical regime, let

\[
C_N = e^{-\langle N \rangle/2} \frac{\langle N \rangle^{N/2}}{\sqrt{N!}}.
\]

(20)

Together with a factorizable \( \Phi_N \) in Eq. (19), the quantum state becomes a coherent state in
the continuous mode representation [12], and the mean electric field is then given by

\[
E(r, t) = i \left( \frac{\langle N \rangle \hbar \omega_0}{2\epsilon_0 \lambda_0^3} \right)^{1/2} \int_{\alpha}^{\beta} d\Omega \int_{0}^{\pi} d\theta \int_{0}^{2\pi} d\phi
\]

\[
\times (\Omega^3 \sin \theta)^{1/2} \varepsilon(\theta, \phi, \sigma) f_e(\Omega, \theta, \phi, \sigma)e^{i k \cdot r - i \omega t} + \text{H.c.},
\]

(21)

which, incidentally, must be an exact solution of the Maxwell equations. The Fourier transform of \( E(r, t) \) is proportional to \( \omega^3 \), and the classical power spectrum is then proportional
to \( \omega^6 \) within the allowed frequency band.

Consider now the magnetic field operator:

\[
\hat{B}(r, t) = \frac{i}{(2\pi)^{3/2}} \sum_{\sigma} \int d^3 k \left( \frac{\mu_0 \hbar \omega}{2} \right)^{1/2} \kappa \times \varepsilon(k, \sigma) \hat{a}(k, \sigma)e^{i k \cdot r - i \omega t} + \text{H.c.},
\]

(22)

where \( \kappa \equiv k/k \). While the total magnetic energy must be the same as the total electric
energy for photons in free space, it is not difficult to show that the magnetic energy density at
\((r_0, t_0)\) is zero where the electric energy density is maximum for the state given by Eqs. (19).
To maximize the magnetic energy density instead, we can simply apply the same procedure as above to the magnetic energy density, which turns out to obey the same bound as the electric one:

$$
\left\langle \frac{1}{2\mu_0} \vec{B} \cdot \vec{B} \right\rangle \leq \frac{\pi}{3} \langle N \rangle \left( \frac{\hbar\omega_2}{\lambda^3_2} - \frac{\hbar\omega_1}{\lambda^3_1} \right).
$$

(23)

The quantum state with the ultimate magnetic energy density is also similar to the electric case,

$$
\Phi_N = \prod_{n=1}^{N} f_b(\Omega_n, \theta_n, \phi_n, \sigma_n),
$$

$$
f_b = \begin{cases} 
-ic^{-1/2}(\Omega^3 \sin \theta)^{1/2} \mathbf{m} \cdot [\mathbf{k} \times \mathbf{e}^*(\theta, \phi, \sigma)] e^{-i c k \cdot r_0 + i\omega t_0} & \text{for } \alpha \leq \Omega_n \leq \beta, \\
0 & \text{otherwise},
\end{cases}
$$

(24)

where \( \mathbf{m} \) is the unit vector of the magnetic field at \((\mathbf{r}_0, t_0)\).

The ultimate electric and magnetic energy densities may be challenging to achieve experimentally, as they require a power spectrum proportional to \( \omega^6 \) within the allowed band, spatial focusing in all directions with a specific angular spectrum, and polarization control. That said, the results set forth impose fundamental limits to which the energy densities can reach regardless of technological advances in the control of electromagnetic fields, and therefore should prove useful for designing ultrafast optics experiments.

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