Synchrotron radiation from an accelerating light pulse

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Synchrotron radiation—namely, electromagnetic radiation produced by charges moving in a curved path—is regularly generated at large-scale facilities where giga–electron volt electrons move along kilometer-long circular paths. We use a metasurface to bend light and demonstrate synchrotron radiation produced by a subpicosecond pulse, which moves along a circular arc of radius 100 micrometers inside a nonlinear crystal. The emitted radiation, in the terahertz frequency range, results from the nonlinear polarization induced by the pulse. The generation of synchrotron radiation from a pulse revolving about a circular trajectory holds promise for the development of on-chip terahertz sources.

Synchrotron radiation (that is, the emission from a relativistic charge moving along a circular trajectory) was first observed in 1947 (1). Until the 1960s, synchrotrons were used to accelerate charged particles exclusively for experiments in particle physics, and radiation losses were studied primarily because they were an impediment to achieving high energies (2–4). Presently, large-scale synchrotrons that produce high-intensity x-rays are used in a wide range of scientific studies. Synchrotron radiation has also been produced at radio frequencies from accelerating polarization currents created by capacitor arrays (5) and is commonly observed in astrophysical phenomena such as radio galaxies and supernovae (6).

Here we present the observation of synchrotron radiation resulting from the acceleration of a light pulse (7–11). Our work distinguishes itself from previous studies involving accelerated beams in nonlinear media (12–14) in that the emphasis is placed on the identification of features exclusively associated with the centripetal acceleration of the primary beam. A metasurface (15–20) was used to guide a pulse of 800-nm central wavelength and 100-fs duration along a 100-μm-scale circular arc inside a lithium tantalate (LiTaO\textsubscript{3}) crystal. As the pulse mixes with itself through the second-order susceptibility of the crystal, the accelerating light pulse generates both second-harmonic and difference-frequency nonlinear polarizations. As for a charge moving under the influence of a centripetal force in vacuum, interference effects lead to a nonlinear polarization, which moves along a circular path and emits synchrotron radiation, as if acted upon a fictitious force (21).

We measure the emission from the difference-frequency polarization, which is in the terahertz range, over a scale of 100 μm.

In LiTaO\textsubscript{3}, the nonlinear polarization generated by a 800-nm pulse propagates at approximately three times the phase velocity of light at terahertz frequencies, below those of the lowest-lying transverse optical (TO) phonons of A\textsubscript{1} and E symmetry (22). As a result, synchrotron radiation at these frequencies interweaves strongly with Cherenkov radiation (23–26), and as such, its spectrum has features that are fundamentally different from those of charges circulating at subluminal speeds. Because the polarization field defines the local dipole density, the general properties of the radiation produced by our pulses relate to those of emission resulting from the motion of a single dipole or, more simply, from a charge \( q \) that moves at a constant speed \( v \) about a circular path. Assuming that the charge makes an infinite number of revolutions, the power emitted into the \( m \)th harmonic of the revolution frequency, \( \omega_0 = v/R \), is given by (26)

\[
P_m = \frac{q^2 v m \omega_0^2}{c^3} \left[ 2 J_2(m(u) + \frac{n}{2} \left(1 - c^2/v^2 \right) J_2(m(u) dx \right)
\]

where \( R \) is the radius of the circle; \( J_{2m} \) and \( J_{2m}' \) are, respectively, the Bessel function of order \( 2m \) and its derivative; \( c \) is the speed of light in vacuum; \( n \) is the index of refraction at the \( m \)th harmonic and \( u = 2\pi m v/c \).

Here \( \omega_0 \) is the revolution frequency, \( \omega_0 = v/R \), and \( \omega_0 \) is the maximum frequency. Figure 1 shows the calculated discrete power spectra for emission from a charge at both subluminal and superluminal speeds in a medium with constant \( n = 6.3 \). The radiated power increases with frequency when \( v > c/n \), whereas for \( v < c/n \), the emission spectrum exhibits a cutoff at a critical frequency \( \omega_C = 3\omega_0/(2 - n^2 c^2 v^2)^{1/2} \), beyond which the radiation emitted is negligible (this is more clearly seen on a linear scale) (28). In our experiments, the highest synchrotron frequency attainable is determined by the bandwidth of the pump.

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\textbf{Fig. 1. Radiated power: Comparison between subluminal and superluminal speeds.} Calculated power radiated by a charge moving in a circle of radius \( R = 50 \mu m \) at speeds below and above \( c/n \), in a medium with constant \( n = 6.3 \) (note the logarithmic scale) is shown. Emission is at integer multiples of the fundamental frequency \( \omega_0 = v/R \). At subluminal speeds, the emitted power drops rapidly for frequencies greater than the critical angular frequency \( \omega_C \). (Inset) Results for \( v = 3c/n \) and three different radii (linear scale). For speeds \( v >> c/n \), the power emitted into harmonics behaves linearly with slope given by \( (q^2 v^3/c^3)(1 - c^2/v^2)^{1/2} \). The curves and lines are guides for the eye. a.u., arbitrary units.


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pulses. For speeds considerably greater than $c/n$, the radiated power increases substantially. For example, the power radiated at a given frequency for $v = 3c/n$ is several orders of magnitude larger than that at a nearby frequency for $v = 0.95c/n$. We emphasize the facts that these results assume a frequency-independent refractive index and that the spectra consist of discrete lines only if the motion is strictly periodic; a source undergoing a few cycles or a fraction of a cycle emits instead a continuous spectrum. This applies to conventional synchrotron radiation for which the source is a single charge, as well as to a nonlinear polarization moving along a circular path. Calculated spectra for an electron revolving in a medium with constant $n = 6.3$ and $v = 3c/n$ for three different radii are shown in the inset of Fig. 1. The values for the electron velocity and light speed are similar to those in our experiments on LiTaO$_3$. The fact that the radiation involves a few discrete frequencies suggests that light-produced synchrotron terahertz radiation could be useful as a means to obtain on-chip continuous-wave terahertz sources. Furthermore, synchrotron emission from a nonlinear accelerating polarization can serve as a new method for generating coherent light at any frequency and in any medium at which conventional phase-matching with the nonlinear polarization is poor, such as at frequencies immediately above the Reststrahlen band in crystals such as ZnTe or GaP, or at second-harmonic frequencies. Within this context, we note a report of unusually phase-matched second-harmonic generation using Bessel beams (29).
We designed a plasmonic nanoantenna meta-
surface (17, 18) to accelerate light of wavelength
800 nm along a circular arc of radius of cur-
vature equal to 100 μm, inside the LiTaO3 crystal.
Unlike conventional methods for accelerating
light, which rely on spatial light modulators (30),
metasurfaces have the ability to bend light at
arbitrarily large angles from the normal to a
planar interface and thereby produce trajec-
tories of very small radii inside a material (31).
The nanoantennas scatter roughly 5% of the
incident power into the accelerating beam, which is
cross-polarized relative to the incident
polarization. Approximately 50% of the incident
beam power is transmitted unperturbed and
~20% is reflected. To reduce the spatial overlap
between the unperturbed and accelerating beams,
the nanoantennas were arranged to produce an
accelerating beam when illuminated at a 45°
angle of incidence. The design process is
described in detail in (31) and summarized in
the supplementary materials (32). A two-dimensional
(2D) plot showing the calculated incident and
scattered intensities for continuous-wave illu-
mination at 800 nm is shown in Fig. 2A. Calcula-
tions of the accelerating pulse produced by
the metasurface when illuminated with a pulse
from an ultrafast laser are presented in (32).

The 500-μm–by–500-μm metasurface was fab-
ricated onto a 1-mm-thick LiTaO3 substrate by
using electron-beam lithography and lift-off
techniques. Prior to fabrication, all four of the
1-mm-wide sides of the substrate were polished
to optical quality to eliminate Rayleigh scattering.
Details are discussed in (32). A schematic
describing the positioning of the metasurface
and a scanning electron microscope image of
the nanoantennas are shown in Fig. 2B.

The accelerating pulse was generated by il-
uminating the metasurface with a pulse of
800-nm central wavelength and duration of 100 fs
from a regeneratively amplified Ti:sapphire oscillator
operating at the repetition rate of 250 kHz.
We performed a spatially resolved pump-probe experiment in the differential transmission geometry to detect the terahertz radiation generated by the accelerating pulse. The experimental setup is shown in Fig. 2C. The pulse is polarized parallel to the optical (z) axis while the electric field of the accelerating pulse lies in the xy plane and rotates slightly as the pulse propagates. The component of the nonlinear polarization, \( P_{NL} \), relevant to our experiments is that along the optical axis \( P_{NL} z = \frac{1}{2} \left| E_p \right|^2 \), here, \( E_p \) is the field of the pump, and \( \chi^{(2)} \) is the corresponding element of the second-order nonlinear susceptibility. The probe beam directed to the sample was focused to a spot with a full width at half maximum (FWHM) of 10 μm in the plane of the accelerating beam trajectory. The probe’s focusing lens was mounted on a motorized translation stage, allowing for the translation of the focal spot throughout the acceleration plane. To calibrate the position of the probe, the accelerating and focused probe beams were first imaged together using a standard imaging setup. The transmitted probe was then directed to a balanced photodetector at which its average intensity was subtracted from that of the reference beam. The focal spot was translated throughout a 400-μm–by–400-μm area of the accelerating beam plane in steps of 10 μm. At each position, the arrival time of the probe pulse was delayed in steps of 53.4 fs, and the corresponding differential intensity trace was recorded. The output of a completed measurement is an image sequence showing the propagation of the time-derivatives of the 800-nm pump-pulse intensity and terahertz synchrotron field. We did not measure the power spectrum of the terahertz radiation. As noted earlier and as opposed to the discrete emission resulting from repeated rotations (Fig. 1), we expect it to be a continuum because the accelerating pulse moves just a fraction of one cycle. Further details concerning the experiment and the processing of the data are presented in the supplementary materials (32).

Figure 3 displays calculated 2D plots of the accelerating pump pulse alone, at different po-
positions along the beam trajectory, as well as an experimental image of the accelerating beam trajectory obtained from the pump-probe measurements. The main results of this work are presented in Fig. 4. Panels A to D show plots of the measured time derivatives of the terahertz electric field and pump intensity for four different arrival times of the probe. In all cases, the unperturbed incident pulse, which passes through the metasurface, was digitally removed. Consistent with results reported in (33), this pulse generates a negligible amount of terahertz radiation at its edges, which was also removed from the data. Panels E to H of Fig. 4 show the corresponding theoretical simulations. For comparison purposes, results of calculations for the radiation emitted by a single dipole are displayed in panels I to L.

The theoretical difference-frequency synchro-
tron electric field was obtained by numerically
solving the following scalar, inhomogeneous wave equation for each terahertz frequency component

\[
\left( \frac{\partial^2}{\partial t^2} + \frac{\partial^2}{\partial z^2} + \frac{\Omega^2}{c^2} \right) E_S = \frac{1}{c^2} \frac{\partial}{\partial t} \left( \chi^{(2)} |E_p|^2 \right) / \Omega \tag{2}
\]

Here, \( \Omega \) is the terahertz frequency and \( f(t) \) denotes the Fourier transform of \( f(t) \) at \( \Omega \). \( E_S \) is the electric field of the 800-nm accelerating pulse; and \( \chi^{(2)} \) is the nonlinear susceptibility of the metasurface. The theoretical plots in Fig. 4, E to H, were generated by taking the time de-ivatives of the solution to Eq. 2 and the calculated accelerating pulse intensity. The 2D matrix for each plot was then convolved with a Gaussian function of 10-μm FWHM to account for the size of the probe’s focal spot. The complete image sequence showing the time evolution of the pump pulse and synchrotron field is presented in (32).

The simulations reproduce most of the features observed in the experiments, and there is reasonably good quantitative agreement between the two sets of results shown, respectively, in panels E to H and A to D of Fig. 4. Consistent with the optical properties of LiTaO3, the 800-nm pulse propagates at a speed approximately three times that of the terahertz synchrotron field. As mentioned earlier, the superluminal nature of the propagation leads to new features in the radiation pattern, which distinguish themselves from those of ordinary, subluminal synchrotron
Henstridge opposed to a single one (two (inner and outer) radiation branches as measured terahertz fields show the presence of radiation. Specifically, both the theoretical and measured terahertz fields show the presence of two (inner and outer) radiation branches as opposed to a single one (28). As the early time panels A and E show, these branches meet in a form that strongly resembles the characteristic shock wave of Cherenkov radiation. By the time the pulse passes through the midpoint of the circular trajectory, the fields have developed prominent curvatures in both branches, which are readily apparent for \( t \geq 1.87 \) ps. Such a distorted, curvilinear Cherenkov pattern is also evident in the single-dipole results. Another key feature of superluminal speeds is the formation of a cusp in the inner radiation branch (34, 35), which is clearly observed in the calculations for the single dipole. Note that the cusp is well developed by the time the dipole makes one-half of a revolution. The 800-nm pulse in the experiment completes between one-quarter and one-half of a revolution. Consistent with the single-dipole data, the motion of our pulses produces a small cusp in the inner radiation branch that can be seen in Fig. 4 at \( t = 2.94 \) ps near \((x, z) = (175 \mu m, 75 \mu m)\) and at \( t = 4.01 \) ps near \((x, z) = (200 \mu m, 100 \mu m)\).

REFERENCES AND NOTES
32. Materials, methods, and supplementary media are available online.

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supervised the project; M.H. designed and carried out the ultrafast optical measurements, performed the calculations, and prepared the LiTaO3 substrates; C.P. designed the metasurface with input from A.G.; D.W. fabricated the metasurface with contributions from V.M.S. and A.B.; and M.H. wrote the manuscript with help from R.M. All authors reviewed and commented on the manuscript.

Competing Interests: We declare no competing interests.

Data and materials availability: All data are presented in the manuscript and supplementary materials.

SUPPLEMENTARY MATERIALS
www.sciencemag.org/content/362/6413/439/suppl/DC1
Materials and Methods

Supplementary Text
Fig. S1
Movie S1
References (36–38)
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Shrinking synchrotron generation
The generation of synchrotron radiation is typically achieved by accelerating charges in large magnetic fields. Synchrotron facilities are usually the realm of large international organizations. Henstridge et al. show that the interaction of a femtosecond light pulse moving in an arc on a specially designed metasurface can also generate synchrotron radiation. In this case, the synchrotron radiation at terahertz frequencies was produced by the nonlinear polarization induced by the light pulse. The results hold promise for the development of powerful on-chip light sources.
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