Tunable narrowband terahertz emission from mastered laser–electron beam interaction

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Published online: 30 March 2008; doi:10.1038/nphys916

In the quest for sources of optical radiation in the terahertz domain, promising candidates are nonlinear optical processes occurring when an intense laser beam interacts with a material medium[1–5]. Besides conventional media (such as crystals), relativistic electrons also show striking nonlinear collective behaviours, which can lead to powerful laser-induced coherent emission[6,7], revealing huge potentials of these devices as terahertz sources[8]. However, up to now only broadband emissions were reported, and experimental control of their radiation properties, such as their spectra[9,10], remained an important challenge. Here, we demonstrate the possibility of mastering the coherent emission experimentally by producing tunable narrowband terahertz radiation. The interaction is made to occur between an electron beam and laser pulses possessing a longitudinal quasi-sinusoidal modulation, and the narrowband emission occurs in a region of quasi-uniform magnetic field. The process therefore strongly differs from classical synchrotron radiation experiments, where narrowband emission occurs inside a periodic magnetic field.

Terahertz generation in classical media has a long history[1]. The fundamental processes involved, in particular optical rectification, current transients in semiconductors and difference frequency mixing, have been extensively studied both theoretically and experimentally, some of them since the 1960s[3]. Now, nanojoule to microjoule energies can be obtained in broadband sources[1,4,5], and narrowband emission on the basis of laser pulse-shaping[11–15] is also a well studied technique. Besides, terahertz emission using laser–electron interaction (laser-induced charge-density modulation) is a very new field. First results on broadband terahertz emissions using short-pulse laser slicing in an undulator have been reported during the past two years[6,7,10,16,17]. Shortly before, Carr *et al.*[8] addressed the closey related question of power attainable by charge densities showing picosecond-scale modulations (very short electron bunches in this case). This research field needs now experimental investigations at the very fundamental level, concerning in particular the actual feasibility of interaction processes and their potentials, as well as their fundamental limits.

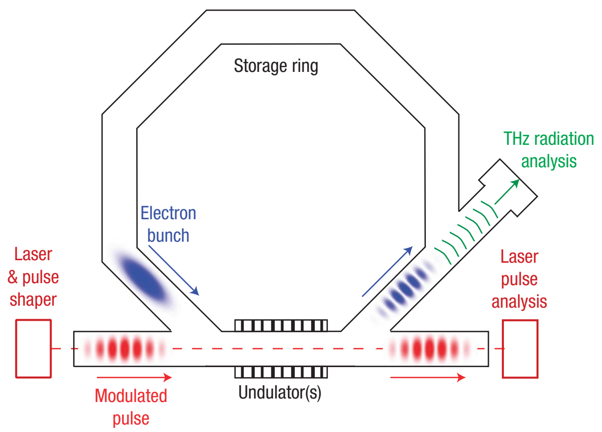

Figure 1 Principle of the experiment. A laser pulse (at 800 nm) is shaped with a longitudinally sinusoidal modulation, with a period in the picosecond range. The pulse interacts with the electron bunch of an accelerator (the UVSOR-II storage ring here), in a region of periodic magnetic field (an undulator tuned at the laser wavelength). The electron bunch is then deviated by a dipole magnet, and terahertz emission occurs with a process similar to classical laser-induced slicing coherent synchrotron radiation[6,7,10,16,17].

In laser–electron beam interaction, the technical arrangement presents similarities with terahertz emission experiments in classical materials. However, the physics strongly differs and involves complex evolutions of the electrons in phase space, specific to the so-called laser-induced slicing[6,7,10,16,17].

The principle of our experiment is illustrated in Fig. 1. A laser pulse with a quasi-sinusoidal envelope interacts with the electron bunch of a storage ring, in a region of periodic magnetic field (an undulator tuned at the laser wavelength). In this first step (Fig. 2a,b), the electrons mainly experience a fast energy modulation at the optical scale (not resolved in Fig. 2),

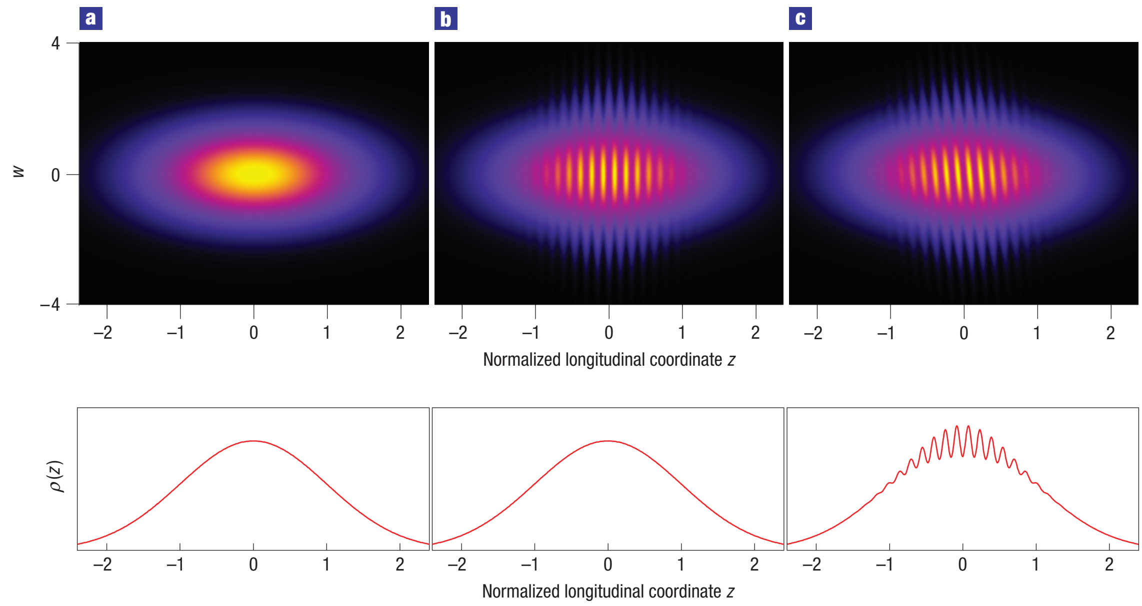

Figure 2 Illustration of the narrowband terahertz-generation process. Electron-bunch phase-space distributions (upper images) are represented with their associated longitudinal charge density distributions $\rho(z)$. z and w are the electrons' longitudinal position and energy deviation respectively, expressed in units of their r.m.s. values before interaction. **a**, Distribution before interaction. **b**, Just after interaction with the laser in the undulator, modulations are visible in phase space, but do not noticeably affect the charge distribution. **c**, After visiting the next magnetic field section, the distribution is 'tilted'. $\rho(z)$ is then longitudinally modulated, and strongly radiates. A fast oscillation, not visible at this scale, is also present (see the theoretical section for details).

whose amplitude is quasi-sinusoidally modulated at a terahertz frequency. Then, the electrons pass through a bending magnet, where they experience different trajectories. As a result, the phase-space distribution is 'tilted', as shown in Fig. 2, leading to a longitudinal charge modulation, which in turn induces strong terahertz emission. The emission frequency is equal to the laser-pulse modulation frequency.

Although the desire to realize experimentally such processes was reported some time ago through theoretical/numerical works by our group[9] and the LBNL laboratory[10], and although recent experiments with short (unmodulated) pulses were successful[6,7,10], experimental evidence of terahertz emission in the modulated case still remained an open question. A main factor probably stemmed from the initial project to use—instead of sinusoidal modulations—a series of laser pulses, which can be delicate to produce[9,10]. Here, a key point consisted of using a pulse-shaping technique widely used in solid-state terahertz sources[12–15], chirped pulse beating[11] (see the Methods section), to produce the pulses incident on the electron bunch. Compared with the set-ups envisaged for multiple-pulse generation[18], chirped pulse beating enables us to obtain a sinusoidal modulation with a widely scalable period T_m and pulse duration T_D using a simple (and robust) set-up, compatible with accelerator environments. The pulse duration could be varied from a few picoseconds to 100 ps, and we examined the terahertz emission for a wide range of modulation periods. The pulse repetition rate was 1 kHz and the incident energy was typically in the 0.1–0.5 mJ range.

Experiments quickly revealed strong tunable terahertz emission when the period T_m was in the 1–2 ps range. First experiments were carried out using pulses with relatively short durations and high peak powers (≈2 ps, 0.5 mJ), to start with conditions close to the already mastered laser slicing[6,7,16,17]. Then we chose to increase the pulse-train duration to 60 ps using a grating stretcher (see the Methods section). This increased strongly the number

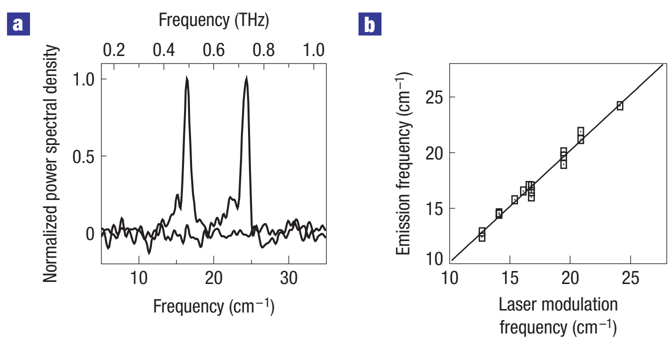

Figure 3 Typical narrowband terahertz emission spectra and tunability curve. **a**, Two terahertz spectra obtained with a 60-ps-long pulse and two positions of the Michelson interferometer corresponding to modulation frequencies of the laser pulse of 16 cm^{-1} and 24 cm^{-1} respectively. The pulse energy at the undulator is of the order of 130 μJ. **b**, Peak emission frequency versus laser-pulse modulation frequency (obtained with pulses of 2 ps and 60 ps). The full line is the 45° line.

of modulation periods, and enabled us to reduce drastically the spectrum width of the emitted terahertz radiation from ≈4 cm^{-1} to ≈1 cm^{-1}. Typical terahertz spectra obtained in these conditions are shown in Fig. 3a. Compared with conventional laser-induced slicing coherent synchrotron radiation (CSR), it is important to note that the effect was obtained with a peak power dramatically lower, because of the larger pulse durations used here (60 ps instead of typically 50 fs–2 ps in classical CSR[6,7,16]), and because of the energy loss introduced by the standard-quality gratings used in the pulse-shaper.

Tunability is possible simply by adjusting one Michelson retroreflector position (see the Methods section). In addition to

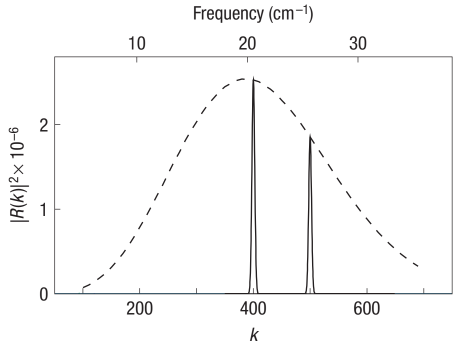

Figure 4 Calculated efficiency versus terahertz modulation frequency. The two peaks (full lines) are spectra associated with modulation frequencies of $k_m = 400$ and $k_m = 500$ respectively. The dashed curve represents the peak value versus modulation frequency. The r_{ij} parameters are the typical ones for the present UVSOR-II experiment: $r_{51} = 5.3 \times 10^{-4}$, $r_{52} = 2.1 \times 10^{-3}$ and $r_{56} = 2.9 \times 10^{-3}$, and the bunch r.m.s. length is 3.1 cm. Laser parameters are $w_0 = 0.3$, $\sigma_L = 0.3$ and $\phi = 0$.

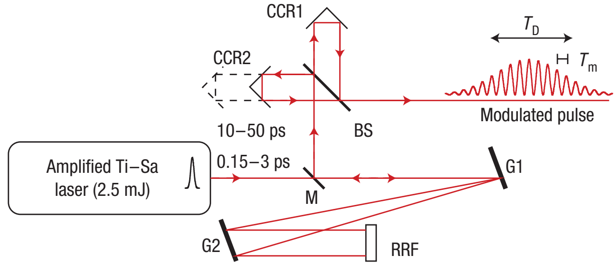

Figure 5 Details of the pulse shaper. G1, G2, 1,200 lines mm^{-1} gratings; M, 45° mirror; RRF, roof retro-reflector; CCR1, CCR2, high-precision corner-cube retroreflectors. Focusing of the modulated pulse is carried out by a lens with 5 m focal length, placed ≈4 m after the pulse-shaping system.

potential applications, this enables us to check the consistency between the internal modulation frequency of the laser pulse, and the emitted peak frequency (they should be theoretically equal). Consistency was found at various pulse durations and beam currents, a typical curve being represented in Fig. 3b. All results hence confirm that the process involved in the narrowband terahertz emission corresponds to the modulated-pulse-induced CSR. A detectable peak was found from modulation frequencies in the 12–25 cm^{-1} range.

From a theoretical point of view, the terahertz emission wavelength range can be estimated from a rather simple approach[10,19–21]. The radiated terahertz field is closely related to the longitudinal electron distribution $\rho(z)$, which acts as a source in the process. In particular it is possible to get the following approximation for the spectrum, that is, the Fourier transform $R(k)$ of the longitudinal charge distribution $\rho(z)$ valid in the terahertz region:

$$R(k) \approx (2\pi)^{-1/2} e^{-(r_{51}^2 + r_{52}^2 + r_{56}^2)k^2/2} \times \int_{-\infty}^{+\infty} e^{-ikz} e^{-z^2/2} J_0(k r_{56} a(z))\, dz, \quad (1)$$

where the r_{ij} are normalized coefficients associated with the transport matrix elements R_{ij} (ref. 19). J_0 is the Bessel function of zero order. For the present purpose, we have examined the response to a gaussian pulse of r.m.s. width σ_L, which induces a variation w_0 of the energy multiplied by a sinusoidal modulation:

$$a(z) = w_0 e^{-(z/2\sigma_L)^2} \cos(k_m z/2 + \phi), \quad (2)$$

where k_m and ϕ represent the wavenumber and phase of the intensity modulation, respectively. As expected and in agreement with experimental data, numerical integration reveals that, at small values of w_0, the response is mainly a single peak at k_m (Fig. 4). The efficiency curve (at moderate w_0) is bell shaped. The peak efficiency is found to tend asymptotically to a finite value (20 cm^{-1} for our experimental parameters) in the small-w_0 limit (in practice $w_0 = 0.3$ or smaller), and this maximum shifts towards lower frequencies when w_0 increases. Asymptotic expressions for the spectrum are beyond the scope of this paper, and will deserve further work. The objective of the present modelling is mainly to provide a simple way through equation (1), to anticipate the accessible terahertz emission range for a set of machine parameters or to design a system emitting at a desired terahertz target wavelength.

The feasibility of the process naturally motivates us to enter two research directions. The first concerns the optimization of terahertz emission (power, bandwidth and so on), and the identification of technical and fundamental limits. For the moment, the typical arrangement presented here—which was destined to feasibility studies and not optimized for high power generation—provided a brilliance comparable to those of commercial sources such as the Teraview (in the nJ/cm^{-1} range) at currents of the order of 20 mA. However, straightforward improvements are expected to increase the emitted power and brilliance. Key points of the powers reachable by optimized set-ups have been discussed in recent papers (see in particular ref. 8), and these topics will hence not be addressed here in detail. However, increases by orders of magnitude are expected to be obtained by elementary optimizations of overlap, incident power and current density in the storage ring, in particular because of the quadratic scaling of terahertz power with laser intensity and bunch charge density. A second, more fundamental interest concerns the use of this type of experiment as a ‘probe’ to investigate the electron beam nonlinear dynamics and instabilities. Spatiotemporal instabilities are indeed of importance because they limit the operation of storage rings at high current. Theories predict the occurrence of instabilities through the growing of unstable modes in a way similar to pattern-forming systems[22], and responses to sinusoidal perturbations are a fundamental point of the instability processes. However, direct experimental tests of these theories still represent big challenges, because phase-space evolutions in a storage ring are usually not observable directly in real time, and also because methods for selectively perturbating short wavenumbers were lacking up to now. Since the type of experiment described here gives the possibility to imprint periodic wave patterns inside the electron-bunch phase space, and to follow phase-space modulations by monitoring the emitted terahertz radiation, we think that this opens the way to new direct tests of current models, for example by providing information on growth/relaxation rates of perturbations versus wavenumber[20,21].

METHODS

The laser pulses are produced by a usual commercial sapphire–titanium laser chain (coherent Mira 900-F and Legend F-HE), delivering 2.5 mJ pulses at 1 kHz, which can be compressed down to 130 fs. To stretch and modulate these pulses, we used the system of chirped pulse beating[11] represented in Fig. 5. A stretcher based on a grating pair (standard 1,200 lines mm^{-1}, size 50 mm, 70 and 80% efficiency) induces a strong negative chirp and increases strongly the pulse duration (in the 5–100 ps range here). Then, two copies of the pulse are made to interfere with an adjustable delay τ in a Michelson interferometer. This leads to a pulse with a quasi-sinusoidal temporal modulation whose frequency is proportional to τ (see ref. 11 for details).

As an important point, the Michelson interferometer is provided with two precision corner-cube retro-reflectors (parallelism < 1 arcsec), to keep a good overlap after the large propagation distance (of the order of 10 m) to the undulator. The stretching is provided by the grating pair for long-pulse operation. For the experiments with shorter pulse widths (typically <5 ps) we did not use this grating pair, and simply adjusted the amplifier's internal grating-pair distance. With this set-up, the total duration T_D is adjustable in the 150 fs–100 ps range, and for a given pulse duration the modulation period T_m is typically adjustable between 150 fs and T_D.

The terahertz emission is analysed at the beam line BL6B (ref. 23) using an in-vacuum Martin-Puplett Fourier transform far-infrared spectrometer, which enables the recording of spectra in the 2–55 cm^{-1} range with a resolution of 0.5 cm^{-1} and is detected by an InSb bolometer (QMC, QFI/2).The pulse processing is made using a gated integrator (SRS250) to reduce background from normal synchrotron radiation. The gated integrator is triggered by the same 1 kHz oscillator as the Ti:Sa laser and the width was chosen around 1 μs, which corresponds to the response time of the InSb bolometer. The UVSOR-II storage ring was operated in single bunch mode at 600 MeV, with a bunch duration of ≈80 ps r.m.s. The experiment was carried out at various currents in the 0–40 mA range (that is, with a bunch charge in the 0–7 nC range), to avoid beam instabilities and spontaneous CSR[24].

Received 9 October 2007; accepted 19 February 2008; published 30 March 2008.

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Acknowledgements

We would like to thank M.-E. Couprie for stimulating discussions and all the UVSOR staff for their help. This work was supported by the Joint Studies Programme (2005–2007) of the Institute for Molecular Science and the JSPS Fellowship Programme for Research in Japan (grant S06215), by Grants-in-Aid for Scientific Research B15360038 and B15360039 from JSPS and by the research foundation for Opt-Science and Technology. CERLA is supported by the French Ministère chargé de la Recherche, the Région Nord-Pas de Calais and FEDER.

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Author contributions

The main contribution of M.K. was in project planning. Modulated-pulse experiments and data analysis were carried out by M.H., M.S., C.E., C.S. and S.B. Analytical and numerical work was done by Y.T., M.S., M.H., C.S. and S.B. The main contributions of A.M., S.K., T.T. and T.H. were in the realization and operation of systems that were crucial in the experiment: the accelerator (A.M.), beamlines (S.K. and T.T.) and laser (T.H.).