

*Invited paper***All-solid-state cavity-dumped sub-5-fs laser****A. Baltuška<sup>1</sup>, Z. Wei<sup>1,\*</sup>, M.S. Pshenichnikov<sup>1</sup>, D.A. Wiersma<sup>1</sup>, Róbert Szipőcs<sup>2</sup>**<sup>1</sup> Ultrafast Laser and Spectroscopy Laboratory, Department of Chemistry, University of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands (Fax: +31-50-3634441, E-mail: D.A.Wiersma@chem.rug.nl)<sup>2</sup> Research Institute for Solid State Physics, P.O. Box 49, H-1525 Budapest, Hungary (E-mail: rsz@power.szfk.kfki.hu)

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**Abstract.** We discuss in detail a compact all-solid-state laser delivering sub-5-fs, 2-MW pulses at repetition rates up to 1 MHz. The shortest pulse generated thus far measures only 4.6 fs. The laser system employed is based on a cavity-dumped Ti:sapphire oscillator whose output is chirped in a single-mode fiber. The resulting white-light continuum is compressed in a novel high-throughput prism chirped-mirror Gires–Tournois interferometer pulse compressor. The temporal and spectral phase of the sub-5-fs pulses are deduced from the collinear fringe-resolved autocorrelation and optical spectrum. The derived pulse shape agrees well with the one retrieved from the measured group delay of the continuum and calculated characteristics of the pulse compressor.

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Ever since pulsed lasers were invented there has been a race towards shorter optical pulses [1]. Next to the fact that the breaking of any record is a challenge, a major scientific driving force came from dynamical studies showing that ultrashort pulses were essential to the exploration of elementary processes in chemistry, photobiology, and physics. For instance, the primary step in bond-breaking reactions (femtochemistry) [2], the rate of electron-transfer in photosynthetic reaction centres [3, 4], and the time scales of relaxation processes in condensed phase [5, 6] could only be time-resolved with femtosecond excitation pulses. On the road towards femtoseconds pulse generation a better grasp of the underlying physics proved to be essential. A milestone here was the development of the colliding pulse modelocked (CPM) laser [7]. When the importance of a careful balance between the group delay and dispersion on pulse formation was recognized [8, 9], sub-100-fs optical pulses became feasible [10, 11]. Further development ultimately led to the prism-controlled CPM laser [12, 13], which delivered pulses of  $\sim 30$  fs. It was this CPM laser that laid the foundation for many ground-breaking experiments in the past

decade, from the observation of wave-packet motion in chemical reactions [14] to the exploration of carrier dynamics in semiconductors [15, 16].

Another crucial invention for ultrashort optical pulse generation was the technique of fiber pulse compression [17]. A relatively long pulse is injected into a single-mode fiber and via the combined action of self-phase-modulation [18] and dispersion becomes spectrally broadened, carrying an almost linear chirp [19]. This spectrally and temporally broadened pulse is subsequently compressed by a pair of gratings [20–22], prisms [10, 23], or their combination [24, 25] to a much shorter pulse. The compressor's action is to retard in a well-defined manner the frequencies of the pulse that have advanced. Pulse compression of the amplified output of the CPM laser culminated in the generation of optical pulses of 6 fs (assuming a hyperbolic secant pulse profile) in 1987 [25]. The electric field of such a pulse exhibits only 4.5 oscillations at its FWHM.<sup>1</sup> With these ultrashort pulses, photon echoes in solution could be studied for the first time [26–28], and their large spectral widths turned out to be very useful for pump–probe experiments in photobiology [29, 30].

A new era in ultrafast laser technology began with the development of the fs modelocked Ti:sapphire laser [31], which routinely generates pulses of about 10–15 fs [32–37]. In addition, this laser exhibits low amplitude noise and is extremely reliable. Not surprisingly, in the past five years Ti:sapphire-based lasers have replaced the CPM lasers in many laboratories as new ultrafast light sources. Sub-10-fs pulse formation from a Ti:sapphire laser also looked promising, as the fluorescence bandwidth of the lasing material [38] allows for pulses as short as 4 fs. However, despite much work, the shortest pulses generated by this laser to date are  $\sim 7$ –8 fs in duration [39–44] and according to current understanding this seems to be the practical limit [42, 45, 46].

With the development of a 13-fs cavity-dumped laser, pulse compression was shown to be a viable route towards

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<sup>1</sup>The frequently cited number of 3 oscillations [6, 25] refers to the duration of the intensity envelope, which, in contrast to the electric field, contains no oscillations at the optical frequency.