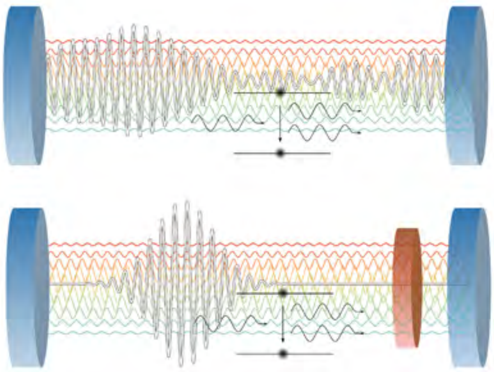
MODE-LOCKING OF LASERS

Franco PRATI[1], Auro M. PEREGO[2], Stephane BARLAND[3] and Germán J. DE VALCÁRCEL[4,*]
[1] Università dell'Insubria, via Valleggio 11, 22100 Como, Italy
[2] Aston Institute of Photonic Technologies, Aston University, Birmingham B4 7ET, United Kingdom
[3] Université Côte d'Azur - CNRS, INPHYNI, Nice, France.
[4] Universitat de València, Dr. Moliner 50, 46100 Burjassot, Spain
***german.valcarcel@uv.es**



Mode-locked lasers are the fundamental source of ultrashort and ultraintense light pulses and a principal pathway for three Nobel Prize technologies: optical frequency combs, chirped pulse amplification, and attosecond light pulses. They are essential in routine and high-level applications such as refractive surgery, spectroscopy, or metrology. Here, we overview the mode-locking working principles and point its main applications out.

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The ubiquity of mode-locked lasers in labs, operating theatres, and industrial processes owes to the extraordinary features of the light pulses they generate, which span the most diverse durations –tens of ps to tens of fs– and repetition rates –MHz to GHz–, reaching peak intensities orders of magnitude higher than the typical monochromatic laser emission. These features stem from synchronisation, a very general physical mechanism of nonlinear dynamical systems whereby a large number of oscillators lock their phases to a common value. Synchronisation can be a spontaneous phenomenon like neural oscillation, or imposed from the outside by a periodic stimulus like the regular heart beatings by a pacemaker.

A typical laser consists of an optical cavity formed by highly reflective mirrors containing a gain element, fabricated from semiconductors, doped crystals or optical fibres, to cite a few. The cavity has a set of well-defined *resonances* (*modes*), so only waves with given frequencies can oscillate within the device. In general, these frequencies are equidistant by the so-called *free spectral range*, equal to the inverse of the time it takes light to travel through the cavity, called the *cavity roundtrip time*. The gain medium can amplify light within a given bandwidth of frequencies. Typically, this bandwidth is much larger than the cavity free spectral range, so many modes can potentially oscillate simultaneously. In the basic operational regime, however, the cavity mode with higher gain wins the competition against the rest for extracting the energy stored inside the gain medium, and the laser emits a (quasi-) monochromatic light beam with constant intensity in time. That is the single-line or single-mode regime. However, the introduction of additional components inside the laser cavity can allow simultaneous and synchronised oscillation of many modes by coherently redistributing the intracavity energy, resulting in a powerful and stable light pulse that travels back and forth inside the cavity. Mode-locking (ML) refers to the variety of techniques for generating light pulses much shorter than the cavity roundtrip time in lasers [1–3], also in optical parametric oscillators, relying on the synchronisation of many modes. This differs from the Q-switching technique which is essentially a single-mode phenomenon resulting in very long pulsations compared with the cavity roundtrip time [1]. Note that any ML pulse contains many optical cycles. Thus, the pulses are characterised by their carrier wave and envelope.

A mode-locked laser emits a periodic train of pulses, which usually comes from a single intracavity pulse. That pulse periodically hits the so-called output coupler (e. g. a semi-transparent mirror), so consecutive pulses in the train are highly coherent with each other, only affected by the small noise component in one roundtrip, which is essential for interferometry. This single pulse regime is called *fundamental* ML. It is possible to have multiple pulses circulating in the cavity too, a regime dubbed *harmonic* ML.

From a historical perspective, it is remarkable that the first indications of laser ML were reported in 1962, just 2 years after the first operation of a laser itself. Since then, the ML laser activity is a success story that continues seemingly unabated [3]. The basic analytical theories of laser ML were firmly established by Dirk J. Kuizenga and Anthony E. Siegman, Geoffrey H. C. New, and Hermann A. Haus in the first half of the 1970's. Notably, Haus developed the master equation framework for different scenarios starting from 1975 [2]. The master equation is a partial differential equation for the pulse envelope. It describes not only the pulse shape, but also the conditions for pulse emergence and, especially, its stability, which is a major concern in *nonlinear dynamical systems* like the laser.

ACTIVE ML OF LASERS

A well-known and relatively simple way to synchronise nonlinear oscillators whose frequencies are equally spaced is by external periodic modulation at a frequency close to that spacing. Historically this was the first ML technique. In this, so-called active ML, an intracavity light modulator, e. g. electro-optic, is subject to an AC voltage by a suitable RF driver, causing its periodic change in transmission (AM-ML; AM = amplitude modulation) or in optical path (FM-ML; FM = frequency modulation). When the driving frequency is carefully tuned to the cavity free spectral range, or to some multiple thereof, many longitudinal modes of the laser cavity can synchronise resulting in very short optical pulses.

The rationale for active ML pulses is readily apparent in both the frequency and time domain. In the frequency domain, a periodic modulation in time manifests in the appearance of new frequencies separated by the modulation frequency, as Fourier analysis tells us. Alternatively, the modulation coherently injects energy from each spectral component to its neighbours so all components eventually synchronise. In the time domain the picture depends on the type of active ML, but the result is similar in both cases. In short, pulses naturally pass through the modulator at times when the modulator introduces the minimum loss (AM-ML), hence experiencing the maximum net gain, or the minimum or maximum phase shift (FM-ML), hence experiencing no net frequency shift and remaining in resonance with the gain (see insert).

Kuizenga and Siegman developed a theory for active ML [1] assuming steady Gaussian pulses. This theory fairly generally predicts pulse durations on the ps scale and above, which compare favourably with experiments; going below ps is not effective with this technique, and we must resort to passive ML. Haus master equation for active ML [2] shows that such Gaussian pulses are stable. The theory assumes that gain is a slow variable, responding to the integrated effect of many successive pulses, and neglects any atomic coherent (quantum) effect [4]. Although Haus' theory is applicable in many cases, it is not generally so. Recently, we have proposed a theory valid for fast gain media in presence of coherent effects (the *coherent master equation:* CME) [5]. The CME is supported by experimental evidence and contains Haus master equation in the appropriate limit, so it can be considered the most complete theory for active ML.

The synchronisation of many cavity modes can be driven by an external clock (active ML) or can be an emergent, spontaneous phenomenon (passive ML), resulting in both cases in short and powerful laser pulses. ML may also result from quantum (atomic) coherent effects, the so-called Risken-Nummedal-Graham-Haken multimode instability [4,5], which we do not consider here.

In AM-ML the pulse crosses the modulator at the maximum transmission (least loss) instants, resulting in maximum net gain. The pulse shortens by the same time-dependent losses, which increase moving away from the pulse peak. This narrowing is balanced at some point by pulse broadening mechanisms like finite gain bandwidth and group-velocity dispersion. The modulation period ideally must match the cavity roundtrip time to ensure that the pulse meets the modulator in the same state roundtrip after roundtrip. In FM-ML, light pulses pass through the modulator in coincidence with a stationary, maximum or minimum, value of the introduced phase shift. Otherwise, the pulse would experience a linear in time phase variation (to the leading order) resulting in a given frequency shift like a Doppler shift, eventually bringing the pulse out of resonance. At a stationary point of the phase modulation the phase acquired by a light pulse varies quadratically in time, so there is no net spectral shift. However, laser pulses get *chirped*, as the "instantaneous frequency", *i.e.* the optical phase time derivative, varies linearly across the pulse. Chirped pulses occur as well, for different reasons, in passive ML, e. g. in fibre lasers.

In passive ML, a saturable absorber (SA) produces a similar result but through different physics that depends on whether the SA is fast or slow, as explained in the main text. Fast SAs are typically based on the optical Kerr nonlinearity in combination with linear elements like apertures and polarisers. The refractive index of a Kerr material varies proportionally to the light intensity. Owing to the transverse Gaussian

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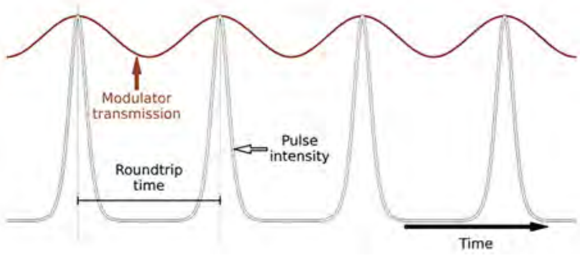
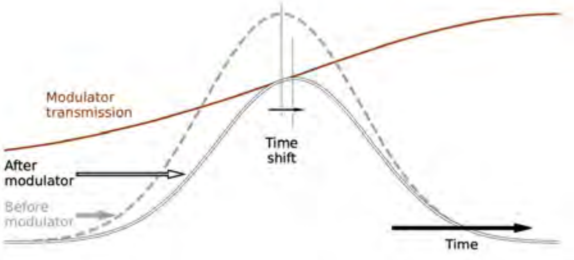
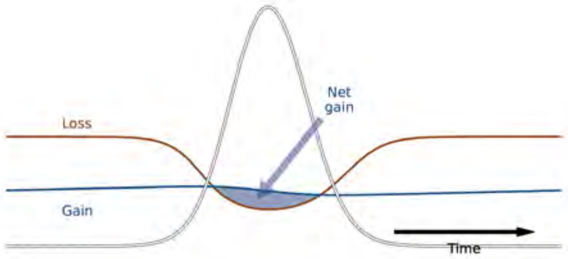
Active ML via amplitude modulation



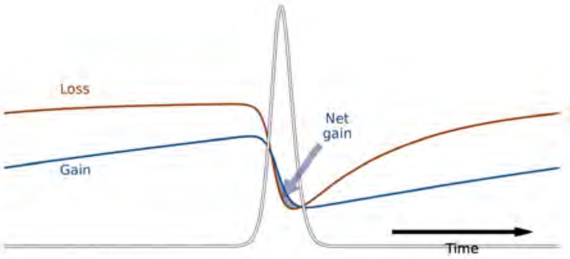
Active ML via amplitude modulation: a "wrong" pulse gets shifted to the "right" place



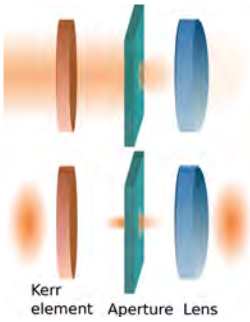
Passive ML with fast SA



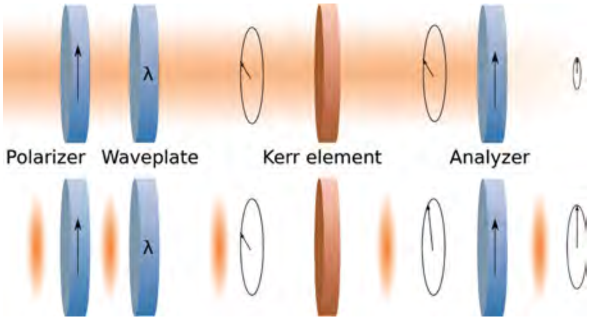
Passive ML with slow SA



Kerr-lens ML



Nonlinear polarisation rotation



● ● ● intensity profile of laser beams, the refractive index of an intracavity Kerr element is maximum at the cavity axis, acting as a convergent lens and resulting in intensity-dependent light focussing. If we put an aperture after the Kerr lens, losses decrease with increasing light intensity, and we get a SA. For the same rationale, pulses shorten as the pulse waist after the Kerr medium localises at the transverse plane containing the pulse peak, like in a diabolo. This so-called Kerr lens ML enables generation of 10-fs pulses with peak powers around the MW, already with commercial solid-state lasers like the ubiquitous Ti:sapphire laser. Another use of the Kerr effect in isotropic materials is the nonlinear rotation of the light polarisation. If this rotation takes place between a polariser and an analyser, the transmission of the device will depend on the light intensity. The relative orientation of the elements is chosen to maximise the losses for the less intense beams, yielding a SA. This method is particularly convenient for fibre lasers.

PASSIVE ML OF LASERS

The most common method of producing ML in a laser is currently passive ML. This is achieved by various means, but with the same rationale: inserting an element into the laser cavity that introduces *saturable* losses, which decrease with increasing light intensity. This element, called a *saturable absorber* (SA), can work by actually absorbing light via an optical transition ("natural" SA) or by exploiting optical nonlinearities ("artificial" SA) [2].

Unlike active ML, passive ML results in the spontaneous formation of ultrashort pulses. In passive ML the modes are "forced" to lock their phase, producing high-intensity pulses, because such a configuration minimises losses. Hence, intense pulses prevail over other forms of emission, according to a Darwinian mechanism of "survival of the fittest". The long-term success of this strategy requires that the pulse bleaches the SA as it passes through, while having sufficient gain, so periodic time windows are created, with period equal to the cavity roundtrip time, in which the net gain is positive. The intensity-dependent loss helps obtaining short pulses by penalising their low-intensity tails.

As an emergent, spontaneous dynamic process, the detailed mechanism leading to the formation of passive ML pulses is highly dependent on the recovery times of SA and gain. While gain is usually slow, having a relaxation time longer than the cavity round-trip time and thus pulse duration, the SA can be slow or fast, with a recovery time much shorter than the pulse duration. Natural SAs are typically slow and are made from different materials, such as semiconductors –most notably the SESAM [3]– carbon nanotubes or dyes, to name a few. On the contrary, artificial SAs are inherently fast, as they are based on the instantaneous Kerr effect typical of glasses and optical fibres.

With a fast SA there is a big difference in response time with the gain, which results in a weak saturation of the latter in correspondence with the passage of the pulse. On the contrary, the fast SA strongly saturates, making the losses momentarily less than gain (see insert). With a slow SA, the relaxation times of absorber and amplifier are comparable, and the process is more involved. The main difference with the fast SA case is that now the dynamic saturations of both the SA and the gain participate clearly in the mechanism of ML, so in this case, both must be easily saturable by the typical pulse intensity. A main practical difference between passive ML with a slow and a fast SA is that the latter, in many cases, is not self-starting (pulses do not emerge from noise). That happens because the instantaneous Kerr response is too weak for weak pulses, and a slow SA is sometimes used in addition to initiate the process. Despite these differences, the common essence of passive ML with a fast and a slow SA is the symbiotic coexistence of a short periodic ● ● ●

In the ultrafast (fs) domain, passive ML of fibre lasers with semiconductor SAs or solid-state lasers with Kerr-lens ML are in general preferred but today semiconductor mode-locked lasers are being designed to operate as monolithic, robust and integrable optical frequency combs for spectroscopy field applications among others.

window of positive net gain with the ML pulse. In mode-locked fibre lasers, the Kerr effect plays a prominent role, and, in many cases, optical soliton effects govern the final state of the device [6]. Theories of passive ML with a fast and slow SA were first established by Haus [2], predicting hyperbolic secant pulses under certain approximations. However, their status with respect to experiments is far from the case of active ML, and theories are under progress that solve these problems [7].

CONCLUSION

Building on the approaches described above, laser ML is now a well-established ensemble of techniques for the generation of high power or ultrashort light pulses for applications ranging from routine tasks to the most extreme regimes of light-matter interaction research. In each of these directions, different implementations offer specific advantages and some form of ML is often at the root of more complex light generation devices. One key application is of course that of optical frequency combs as pioneered by Nobel Prize recipients Theodor W. Hänsch and John L. Hall, where the periodic emission of ultrafast mode-locked laser pulses leads to a broad ensemble of coherent, discrete, equally spaced optical spectral lines, opening new horizons in high resolution spectroscopy, distance ranging, optical clocks, metrology, exoplanets search and optical communications to name just a few. In conjunction with the coherent chirped amplification approach of Nobel laureates Gérard Mourou and Donna Strickland, mode-locked laser pulses are also at the heart of Europe's Extreme Light Infrastructure, one of the most powerful light sources in the world which brings lasers to nuclear and high energy physics. In ultrafast spectroscopy the ultrashort pulses produced by mode-locked lasers allow to study dynamic processes on ps to fs timescales. Such timescales can be lowered to the attosecond using high harmonic generation processes, as pioneered experimentally by the 2023 Nobel Prize awardees Pierre Agostini, Ferenc Krausz and Anne L'Huillier.

Passive ML also finds applications in high-resolution imaging techniques such as optical coherence tomography and nonlinear microscopy. Additionally, these lasers are employed in precision surgeries where the delivery of ultrashort pulses is crucial. Passive ML is utilized in material processing applications, including micromachining and surface ablation. The precise control over the laser pulses enables minimal thermal damage and high-quality processing. In the ultrafast (fs) domain, passive ML of fibre lasers with semiconductor SAs or solid-state lasers with Kerr-lens ML are in general preferred but today semiconductor mode-locked lasers are being designed to operate as monolithic, robust and integrable optical frequency combs for spectroscopy field applications among others.

In contrast to passive ML which leads to ultrafast optics, active ML can lead to more robust and broader tuning of the pulse emission period thanks to the externally controllable clock. For this reason, hybrid systems which feature both a saturable loss element for passive ML and a periodic external forcing are particularly useful when synchronous interfacing with electronics is required as for instance in multiphoton imaging or telecommunication signal reshaping. ●

ACKNOWLEDGMENTS

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