Letter

Mode-locking based on zero-area pulse formation in a laser with a coherent absorber

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Abstract

We experimentally observe mode-locking in a continuous narrowband tunable dye laser with molecular iodine absorber cells, whose transitions have long phase relaxation time $T_2$. We show that mode-locking arises due to the coherent interaction of light with the absorbing medium leading to Rabi oscillations, so that zero-area ($0\pi$) pulses in the absorber are formed. Such mode-locking regimes are different from most typical passive mode-locking mechanisms where saturation plays a major role.

Keywords: coherent optical effects, mode-locking, Rabi oscillations, self-induced transparency, ultra-short pulses, on pulses

(Some figures may appear in colour only in the online journal)

1. Introduction

Mode-locked lasers are principal sources of short light pulses with a high repetition rate \cite{1, 2}. The most commonly used method to create such pulses inside a cavity is passive mode-locking. Typically passive-mode-locked lasers consist of two sections, one containing a gain medium and the other one a nonlinearly absorbing medium. The physical mechanism of mode-locking in such lasers is based on the effect of the saturation of absorption in the absorber and saturation of gain in the amplifier. Recently, another mechanism of passive mode-locking was proposed, where the absorber and gain media operate not in the regime of saturation but in the coherent regime of strong coupling of light with the medium \cite{3–10}.

A coherent light–matter interaction regime arises when the light pulse duration is shorter than the medium polarization relaxation time $T_2$ \cite{11–13}. In coherent physics the interaction of short pulses with a resonant medium is described using the pulse area defined as \cite{11–13}

$$\Phi(t,z) = \frac{d_{12}}{\hbar} \int_{-\infty}^{t} A(t',z) \mathrm{d}t',$$ (1)

with $A(t,z)$ being the pulse envelope and $d_{12}$ the transition dipole moment.

The presence of phase memory leads to unusual phenomena in the propagation of the pulse which cannot be observed when the regime of light–matter interaction is incoherent \cite{3–25}. For example, a pulse can propagate without losses in
the regime of self-induced transparency (SIT) (a pulse with area $2\pi$ [11]), or as a stable $\pi$ pulse [12]. The key idea behind mode-locking based on coherent effects is to use a configuration allowing the same pulse to be a $2\pi$-pulse in the absorber and a $\pi$-pulse in the amplifier [3–10]. In this way, since a $2\pi$-pulse is a stable soliton solution, the absorber damps any background perturbations making the regime stable. At the same time, a $\pi$-pulse configuration in the amplifier allows one to take energy from it, thus leading to a pulse shortening. Despite their great potential for delivering pulses up to a single-cycle duration [6, 7], lasers with such a coherent mode-locking mechanism have not yet been realized experimentally. One of the problems is to obtain coherent interaction inside the cavity for the parameters required for mode-locking. Although coherent effects have been observed in plenty of optical systems [11–29], achieving intracavity coherent effects combined with mode-locking is not a trivial task. However, they have been observed in single-section gas lasers [16], semiconductor structures [17–19] and quantum cascade lasers in the THz frequency range [21]. Furthermore, in a recent paper [22], the signatures of the SIT regime in a rubidium 78Rb gas cell placed in the cavity of a mode-locked laser were demonstrated experimentally. However, the SIT regime in [22] was not related to the mode-locking. In fact, the quality of the SESAM-induced mode-locking degraded in the presence of the SIT. Despite this, the experiment in [22] clearly demonstrated the feasibility of SIT inside the laser cavity. On the other hand, the possibility of coherent regime onset in SESAM absorbers has been predicted theoretically [23] but never tested experimentally. Besides, a SIT pulse is not the only pulse shape which propagates without losses in absorbing resonant media. Another possibility is so-called zero-area pulses ($0\pi$-pulses), which have been predicted theoretically and observed experimentally [24–26] and recently theoretically shown to be also useful in coherent mode-locking [10]. $0\pi$-pulses occur when short pulses (shorter than $T_2$) with a small area propagate in a resonant medium. In this case, the initial part of the pulse excites the resonant polarization, which then irradiates the electric field (‘coherent optical ringing’), which has a phase shift with respect to the incident pulse. As a result, the pulse envelope contains a characteristic tail at the trailing edge. The envelope becomes sign-alternating and the pulse area approaches zero. Importantly, such $0\pi$-pulses are governed by the area theorem of McCall and Hahn [11]:

$$\frac{d\Phi(t,z)}{dz} = -\frac{\alpha_0}{2} \sin \Phi,$$

which for $\Phi \ll 1$ predicts an exponential decay of the pulse area $\Phi(t,z) = \Phi_0 \exp(-0.5\alpha_0 z)$ ($\alpha_0$ is the absorption coefficient per unit length). However, the pulse envelope does not decay and thus the medium remains transparent for these pulses. An area theorem-based theoretical description of CML lasers was developed recently in [10]. In particular, zero-area pulse formation was predicted. The possibility of $0\pi$-pulse generation in laser with a coherent absorber was also discussed in [27].

In spite of the fact that the coherent effects are well-studied theoretically and experimentally to date, there has not been any experimental evidence of CML so far. In this paper, we demonstrate the possibility of passive coherent mode-locking in a narrowband dye laser when absorbing cells with molecular iodine vapors are placed in the cavity. As was preliminarily reported in [28], since molecular iodine vapors have very long coherence time $T_2$ in the range of hundreds of nanoseconds [30], the absorber should be in the coherent regime. Here we provide a solid experimental evidence that this is indeed the case and that coherent interaction with the iodine cell causes mode-locking. Besides, our experimental results corroborated with numerical simulations show that zero-area pulses ($0\pi$-pulses) are formed. The choice of molecular iodine vapor at room temperature as a candidate for a coherent absorber in our experiments is not accidental. It is a common object for observation of coherent interactions in the visible wavelength range such as photon echoes, transient nutations, etc [30]. A certain disadvantage of molecular iodine in our experiments is a high density of absorption lines in the generation region of the rhodamine 6G laser. Nevertheless, this allowed us to observe the mode-locking effect on various absorption lines.

### 2. Experimental results and discussion

As a basis for our experimental setup we used a narrowband CW dye laser as shown in figure 1. This continuous dye laser was assembled on the basis of a laser module ‘T and D scan laser system’. The pump was produced by a Verdi V10 laser. A linear 97 cm long cavity of the dye laser was defined by five mirrors L1–L5. As a dye jet (DJ) an ethylene glycol solution of the dye rhodamine 6G was used. It was placed between two spherical mirrors L3 and L4, and deployed in a horizontal plane at the Brewster angle relative to the direction of lasing. The pump radiation was focused onto the DJ using a mirror P1. To narrow the generation line a Lyot filter (LF) and two Fabry–Perot etalons E1, E2 were used to narrow the generation line. In addition, two I2 absorber cells were placed inside and outside the cavity. We used several variants of the setup, differing by the presence or absence of cells and selective elements in the cavity.
E1 led to a linewidth less than 5 pm. We also placed two cells with molecular iodine $I_2$ inside and outside of the cavity. The cells windows were deployed at the Brewster angle. The laser intensity was registered with a time resolution of about 0.5 ns provided by a high-speed photodiode and a digital oscilloscope. The registration system also included a power meter and a spectrometer of the laser control module controlled the operation of stepping motors associated with the intracavity filters. The spectra were recorded using a Fabry–Perot interferometer. The interferograms of the emission spectra were recorded by a digital camera. We start our consideration from the case where no selective elements and no cells were placed in the cavity. In this case, our laser setup generated a broad spectrum and no pulsations of intensity were observed. Second, we consider a configuration where a single iodine cell was placed in the laser cavity whereas the other cell was placed outside of the cavity, and no frequency selective elements were present in the setup. In this case, dips appeared in the output spectrum whereas no mode-locking was observed. In both cells, no luminescence was visible, despite the considerably high output power. This is a clear indicator that the lasing frequencies did not coincide with the frequencies of the absorption lines in the cell in this case. Hence, in this configuration no interaction of the laser radiation with the resonance lines of the molecular iodine took place. As the next step, an LF was placed in the laser cavity to narrow the spectrum. We tuned the LF to the spectral range between 585.0 and 585.3 nm. This range contained around 14 strong absorption lines as shown in figure 3 (blue line). In this modification of the setup, in contrast to the previous case, the observed output spectrum contained narrow absorption lines, which could be observed in the interferograms, as shown in figure 2 (lower panel). However, no luminescence in the cells and no mode-locking was observed. Unstable self-pulsing similar to that shown in figure 4(a) was observed. To ensure that the origin of these lines is the absorption in the cell and not the parasitic interference effects in the intracavity optical elements, we repeated the experiments after cooling the cell. Cooling facilitates the significant decrease of iodine pressure. To cool the cell we wrapped it in a wadding soaked with liquid nitrogen. Upon cooling the narrow absorption lines disappeared, and appeared again when heating the cell back to room temperature. Finally, we further narrowed the output spectral line by placing the Fabry–Perot etalon E1 in the cavity. Inclination of the etalon allowed tuning the frequency across the maximum of transmission of the LF. In this case, away from the absorption lines unstable self-pulsing (but not mode-locking) regimes were observed similar to the ones in the previous case (figure 4(a)). However, when the laser frequency was tuned to the absorption lines of iodine, mode-locking with long-term stability of the pulse shapes occurred (see figure 4(b)). Mode-locking regimes were observed and well reproduced at the following wavelengths: 580.020 nm, 585.136 nm, 585.155 nm, 585.179 nm, 585.181 nm, and 585.220 nm. We observed that the generation line was located at the position of the dip of the configuration without E1 and E2 (see figure 2, upper and lower panels). Also, in contrast to the previous variant of the setup, the mode-locking was always accompanied by strong luminescence in both cells (the intracavity and the extracavity one). When we cooled the intracavity cells in this setup, both mode-locking and luminescence in the intracavity cells disappeared. The appearance of a stable mode-locking regime at the frequencies corresponding to the resonances of the cells also shows clearly that the coherent interaction in the iodine cells plays a decisive role in the observed mode-locking mechanism. We also measured the absorption in the $I_2$ cells in dependence on wavelength at room temperature and compared it with the luminescence intensity in the cells (see figure 3). To test the absorption, the cells were illuminated by an independent tunable narrowband laser source that enabled us to clearly fix the absorption regions, which, as one can see from figure 3, coincided with the luminescence peaks.

Besides, within the resolution limit of our wavelength meter they also coincided with the wavelengths at which it was possible to observe mode-locking. The latter observation is very important, since sufficiently high absorption was observed in the cells at the resonances. Because of this, one would expect that it should have led to a drop in power of generation in the...
mode-locking regime when the laser frequency was tuned to one of the cell absorption lines. However, our measurements show that the output power did not have any notable drop near the absorption lines. This could be the case only when, as already mentioned, the passage of pulses was not accompanied by significant absorption.

To further clarify the role of absorption in molecular iodine additional experiments were carried out. As the spectrum of I$_2$ contained a large number of absorption lines, one can suppose that the cells played only an auxiliary role as mode selectors, which introduced frequency-dependent losses and thus effectively narrowed the cavity resonance. To check if this possibility took place we artificially narrowed the emission spectrum using the Fabry–Perot element E1 and a thick glass plate E2, but without cells in the cavity. In this case self-pulsating regimes were indeed observed in the cavity (see figure 4(a)). However, these regimes did not have long-term stability in contrast to the mode-locking regimes in the presence of cells (see figure 4(b)). The next important feature of the observed mode-locking was the double-peak pulse shape which is very typical for $\pi$-pulses (see figure 5). Besides, the relative intensities of the sub-peaks in the double-peak pulse experienced slow periodic modulation, which manifested in the modulation of the envelope in figure 4(b).

To corroborate our experimental findings we also performed numerical simulations, which showed rather good agreement with the experiment. For the numerical modeling a system of Maxwell–Bloch equations for the two-level medium described in [8–10] was used. Numerical simulations of these equations showed that the presence of the absorber with the dephasing time $T_2 > \tau$ (cavity roundtrip time) did not significantly influence the average output power of the laser and only slightly reduced its value, which is in agreement with the experimental results. For the laser without an absorber section, due to the fact that the active medium occupied a small part of the cavity, we obtained either CW solutions or self-pulsating regimes. The placing of a coherent absorber in the cavity led to a significant modification of the

![Figure 4](image1.png)  
**Figure 4.** The oscillograms of the output laser intensity measured for 20 $\mu$s. (a) A self-pulsing regime of the laser intensity without the cells and (b) a mode-locking regime in the case where the iodine cell was present in the cavity.

![Figure 5](image2.png)  
**Figure 5.** An oscillogram (time scale 2.5 ns div$^{-1}$) of mode-locked pulses in the experimental setup. The measured contrast (the ratio between the maximal and minimal intensity values) is around 40:1.
dynamics. Pulses which were formed in the absorber had zero area (that is, they were \(0\pi\)-pulses) and thus passed through the absorber without losses. As is usual for \(0\pi\)-pulses, they stored their energy in the medium and then retrieved it with the phase shift \(\pi\) [24–26].

A typical example of a two-peak \(0\pi\)-pulse obtained in our numerical simulations is shown in figure 6. In particular, in figure 6(a) the field envelope is shown and in figure 6(b) the intensity is shown. This is to be compared with figure 5 where an example of an experimentally obtained oscillogram of mode-locked pulses is presented. In a pulse with zero area, the envelope changes its sign as shown in figure 6(a). Therefore, the intensity should be double-peaked and even touch the zero line as in figure 6(b). However, in the experiment, the dip in intensity is not so pronounced and does not touch zero. We explain this by the insufficient temporal resolution of the recording system which we estimate to be around 0.5 ns. To corroborate this estimation, we plot in figure 6(c) a convolution of the intensity with the function \(H(t) = e^{-t/\delta}\) with \(\delta = 0.5\) ns. The convolution models the finite response of the detecting system and the convoluted intensity shows a reasonable agreement with the experiment (figure 5). Furthermore, we were able to reproduce experimentally the observed slow pulsation dynamics mentioned above (not shown). This agreement with the experiment supports both the coherent character of the mode-locking regime and the \(0\pi\) shape of the pulses in our experiment. Finally, we note that an envelope which crosses zero should lead to a tiny spectral dip at the absorber resonance. This dip is however unobservable with the spectral resolution available.

3. Conclusions

In conclusion, we observed mode-locking regimes in a cavity with a molecular iodine cell as an absorber. Despite the complicated level structure of \(I_2\) we were able to lock, using additional spectral filtering, the particular iodine lines. We have demonstrated that mode-locking appears due to the coherent character of the interaction of light with iodine. The observed dynamics of the mode-locked pulses indicate that they do have zero area in the absorber. These findings were supported by numerical simulations, which are in good agreement with our experiment.

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