Passive *Q*-switching and *Q*-switched mode-locking operations of 2 μm Tm:CLNGG laser with MoS_2 saturable absorber mirror

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With MoS_2 as saturable absorber, passive *Q*-switching and *Q*-switched mode-locking operations of a Tm-doped calcium lithium niobium gallium garnet (Tm:CLNGG) laser were experimentally demonstrated. The *Q*-switched laser emitted a maximum average output power of 62 mW and highest pulse energy of 0.72 μJ. *Q*-switched mode locking was also obtained in the experiment. The research results will open up applications of MoS_2 at the mid-infrared wavelength.

OCIS codes: (140.3540) Lasers, Q-switched; (140.3580) Lasers, solid-state.

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1. INTRODUCTION

Q-switched solid-state lasers have been attractive, as they can provide high peak power and pulse energy which can be used in material processing, medical treatment, scientific research, etc. Eye-safe 2 μm *Q*-switched solid-state lasers will be especially promising in surgery, ranging, and nonlinear optical frequency conversion. Among the *Q*-switching techniques, passive *Q* switching with a saturable absorber (SA) is a convenient and low-cost way to achieve *Q*-switching operation. Ultra-broadband SAs in particular can extend *Q*-switch operation wavelength to the mid-infrared band. So far, SAs such as semiconductor saturable absorber mirrors (SESAMs) [1–3], carbon nanotubes [4–7], graphene [8–17], and topological insulators [18,19] have been adopted for passive *Q*-switching and mode-locking operations. An excellent SA should possess the properties of moderate saturation intensity, high damage threshold, ultrafast recovery time, and broadband saturable absorption. However, SESAMs have the drawback of narrow operation bandwidth and complex fabrication process. Thus, there is great motivation to explore SAs with broadband saturable absorption and low-cost fabrication. Typically graphene, a two-dimensional zero-bandgap material, has been confirmed as an excellent broadband SA, but its weak absorption limits its modulation ability for light. Recently, a new type of two-dimensional material, transition metal dichalcogenides [20,21] have attracted extensive attention due to their characteristics of large modulation depth, broadband saturable absorption, and high nonlinear effects.

Monolayer or few-layer MoS_2 is one of the representative two-dimensional transition metal dichalcogenides, with a hexagonal structure of molybdenum atoms sandwiched between two layers of chalcogen atoms (S) [22]. MoS_2 has a thickness-dependent electronic band structure, which endows it with some new optical properties such as strong enhancement of photoluminescence in monolayer [23–25].

Few-layer MoS_2 as a broadband SA has been reported. First, the saturable absorption of MoS_2 dispersions with large population of single- and few-layer MoS_2 were measured at 800 nm [26]. Then, a mode-locked Yb-fiber laser with MoS_2 was realized by Zhang *et al.* [27]. Luo *et al.* demonstrated passively *Q*-switched fiber lasers at 1, 1.5, and 2 μm, respectively, by exploiting a few-layer MoS_2 polymer composite as SA [28]. Inspired by the introduction of quantum dots in semiconductor, Wang *et al.* developed a broadband MoS_2 SA by introducing suitable defects in the process of few-layer MoS_2 fabrication [29], which has broadband saturable absorption extending to 2.4 μm wavelength.

Tm-doped calcium lithium niobium gallium garnet (Tm:CLNGG) is a type of disordered crystal with significant inhomogeneous spectrum line broadening. Continuous wave (CW) mode-locking operations of Tm:CLNGG lasers with SESAM [2] and graphene [30] have been realized before. In this letter, we demonstrate the diode-pumped passive *Q*-switching and *Q*-switched mode-locking operations of a Tm:CLNGG laser by adopting a MoS_2 golden mirror as saturable absorber mirror (SAM). In the stable *Q*-switching state, the maximum pulse energy reached 720 nJ. In the *Q*-switched mode-locking state, the laser operated in harmonic mode locking with a repetition rate of 187 MHz.

2. EXPERIMENT SETUP

The schematic of a passively *Q*-switched and *Q*-switched mode-locked Tm:CLNGG laser was shown in Fig. 1. An X-folded cavity was adopted in the experiment. The Tm:CLNGG crystal employed had a length of 9 mm and a cross-section of $4\ mm \times 4$ mm, with a Tm doping of 6%. The crystal was

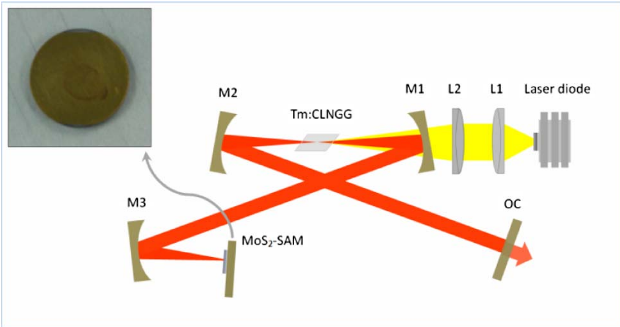

Fig. 1. Schematic of Tm:CLNGG laser setup with MoS_2 as SA. Inset: picture of the MoS_2 on golden-film mirror.

wrapped with indium foil and mounted in a water-cooled copper block. During the laser operation, the circulating water temperature was kept at 13°C. A single-emitter AlGaAs laser diode at about 790 nm was used as the pump source. The pump light was collimated by a doublet lens (L1) and then focused into the Tm:CLNGG crystal by another doublet lens (L2). Both lenses have the same focal length of 100 mm. The three plano-concave mirrors M1, M2, and M3 have the same radius of curvature of −100 mm, and were all highly reflectively coated for laser wavelength and antireflectively coated for pumping wavelength. A plano–plano mirror with 2% transmission was used as the output coupler. The MoS_2 SA was grown on a golden mirror with the pulsed laser deposition method as described in Ref. [23]. The laser mode size was ~40 μm in radius on the MoS_2 SA mirror. The inset of Fig. 1 shows the picture of the MoS_2 golden mirror. The large-area, few-layer MoS_2 can be clearly seen in the middle part of the golden mirror, and the modulation depth and nonsaturable loss were measured at 2 μm wavelength to be 7.2% and 10.0%, respectively.

3. RESULTS AND DISCUSSION

While the absorbed pumped power rose up to 2.19 W, the Q switching was initiated with a MoS_2 gold mirror as an end mirror. In the Q-switching operation, the maximum average output power was 62 mW with an absorbed pump power of 2.57 W. This low output power was attributed to a large linear loss of the MoS_2 mirror. The output laser had a round TM_{00}

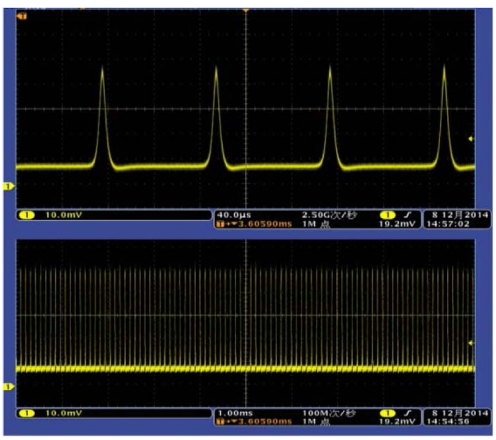

Fig. 2. Q-switching pulse train in 400 μs and 10 ms timescales, respectively.

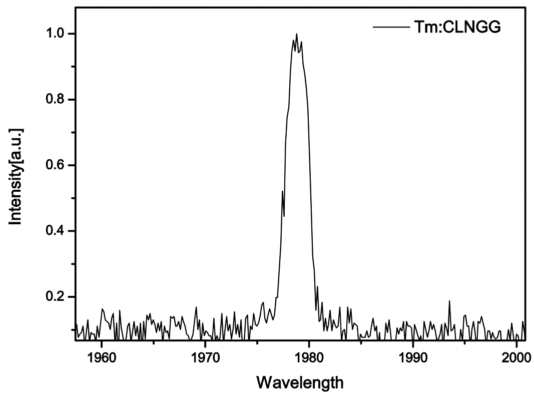

Fig. 3. Optical spectrum of the Q-switched Tm:CLNGG laser.

mode. With a high-speed detector (EOT, ET-5000) and a 500 MHz bandwidth oscilloscope (Tektronix, DPO3054), the typical Q-switched pulse train was recorded in 400 μs and 10 ms time scales, as shown in Fig. 2. It shows a clear and stable Q-switching operation. The optical spectrum of the Q-switched laser was measured with a mid-infrared optical spectrum analyzer (Ocean Optics, SIR5000) with a resolution of 0.22 nm, as shown in Fig. 3. The spectrum was centered at 1979 nm with a bandwidth of ~2.5 nm.

In the stable Q-switching operation, the dependence of repetition rate, pulse duration, average output power, and pulse

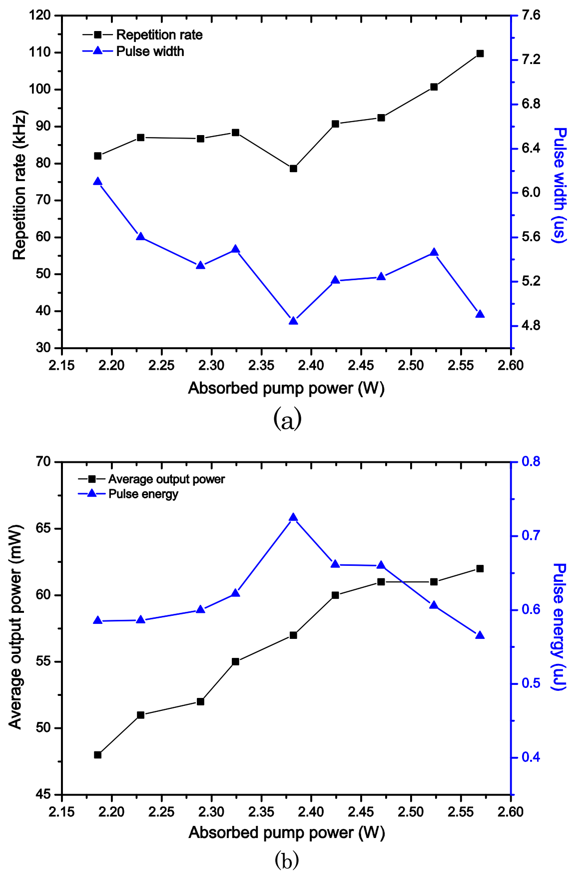

Fig. 4. (a) Q-switched pulse repetition rate and pulse duration versus the absorbed pump power. (b) Average output power and pulse energy versus the absorbed pump power.

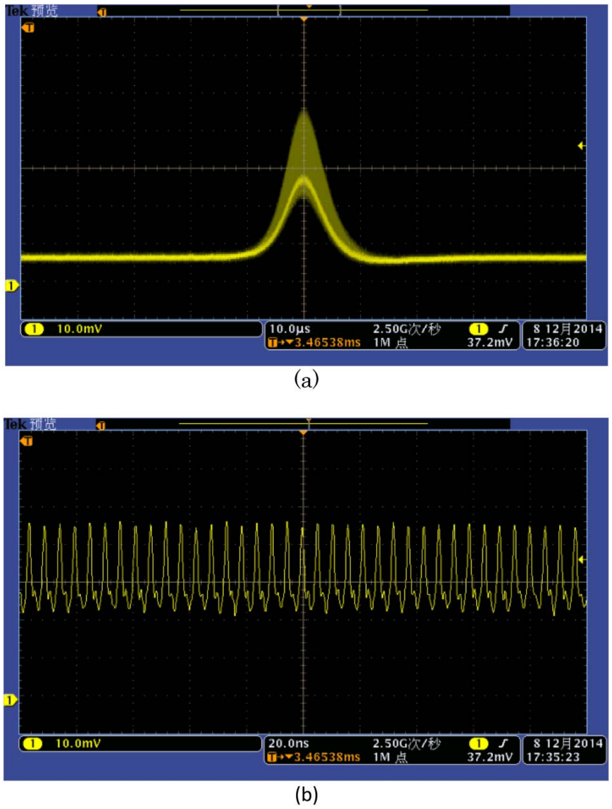

Fig. 5. (a) Envelope of Q-switched mode locking in 100 μs time scale. (b) Mode-locking pulse train within the Q-switching envelope in 200 ns time scale.

energy of the laser on absorbed pump power was shown in Fig. 4. The maximum average output power was 62 mW and the highest repetition rate was 110 kHz under the maximum absorbed pump power of 2.57 W. While the absorbed pump power was beyond 2.42 W, the output laser tended to saturate [Fig. 4(b)]. However, the repetition rate continuously increased with pump power, and thus the pulse energy began to decrease beyond 2.42 W of absorber pump power. The maximum single-pulse energy was 720 nJ at the absorbed pump power of 2.38 W, and the shortest pulse duration was 4.84 μs. When the pump power was beyond 2.57 W, the Q-switching state disappeared. However, after the pump power decreased to 2.57 W, the Q-switching state recovered. During the experiment, no damage of MoS_2 was observed.

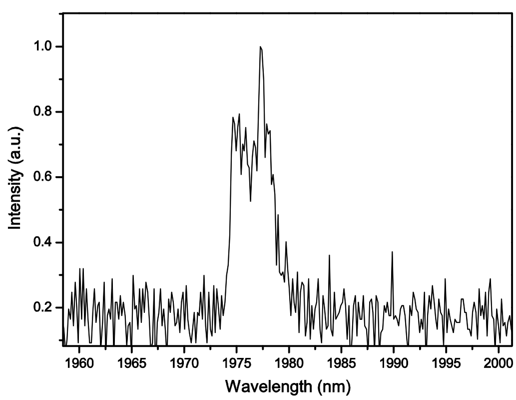

Fig. 6. Spectrum of the Q-switched mode-locked laser.

By carefully optimizing the laser spot position on the MoS_2 SAM and aligning the laser cavity, Q-switched mode-locking operation was realized at the average output power of ~60 mW. The Q-switched envelope and the mode-locked pulse train within the envelope were shown in Figs. 5(a) and 5(b), respectively. The mode-locked pulses have a period of 5.35 ns, which is exactly half of the cavity roundtrip time of 10.7 ns. This means there was harmonic mode-locking operation, induced by the high nonlinear effects of MoS_2, as reported in mode-locked fiber lasers with MoS_2 [31].

The output spectrum of the Q-switched mode-locked laser was shown in Fig. 6. The spectrum was centered at 1977 nm with a width of ~4.4 nm. Compared to the spectrum in the Q-switching state in Fig. 4, the spectrum width in the Q-switched mode-locking state became broader. It can be understood that the Q-switched mode-locked pulses have higher peak power and thus induced larger nonlinear phase, which results in spectrum broadening. However, CW mode locking in the laser was not obtained due to too large modulation depth of MoS_2.

4. CONCLUSION

In conclusion, we have experimentally demonstrated passive Q-switching and Q-switched mode-locking operations in a Tm:CLNGG laser with a MoS_2 golden mirror as the SAM. In the stable Q-switching operation, the maximum pulse energy reached 720 nJ. In the Q-switched mode-locking state, harmonic mode-locked pulses with a repetition rate of 187 MHz were observed. These results indicate that few-layer MoS_2 is a potential SA at the mid-infrared wavelength.

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REFERENCES

1. U. Keller, K. J. Weingarten, F. X. Kartner, D. Kopf, B. Braun, I. D. Jung, R. Fluck, C. Honninger, N. Matuschek, and J. Aus der Au, "Semiconductor saturable absorber mirrors (SESAM's) for femtosecond to nanosecond pulse generation in solid-state lasers," IEEE J. Sel. Top. Quantum Electron. **2**, 435–453 (1996).
2. J. Ma, G. Q. Xie, W. L. Gao, P. Yuan, L. J. Qian, H. H. Yu, H. J. Zhang, and J. Y. Wang, "Diode-pumped mode-locked femtosecond Tm:CLNGG disordered crystal laser," Opt. Lett. **37**, 1376–1378 (2012).
3. J. F. Li, H. Y. Luo, Y. L. He, Y. Liu, L. Zhang, K. M. Zhou, A. G. Rozhin, and S. K. Turistyn, "Semiconductor saturable absorber mirror passively Q-switched 2.97 μm fluoride fiber laser," Laser Phys. Lett. **11**, 065102 (2014).
4. S. Yamashita, Y. Inoue, S. Maruyama, Y. Murakami, H. Yaguchi, M. Jablonski, and S. Y. Set, "Saturable absorbers incorporating carbon nanotubes directly synthesized onto substrates and fibers and their application to mode-locked fiber lasers," Opt. Lett. **29**, 1581–1583 (2004).
5. J. Liu, Y. Q. Li, L. H. Zheng, L. B. Su, J. Xu, and Y. G. Wang, "Passive Q-switched mode locking of a diode-pumped Tm:SSO laser near 2 μm," Laser Phys. Lett. **10**, 105812 (2013).
6. X. T. Chen, S. Z. Zhao, J. Zhao, K. J. Yang, G. Q. Li, D. C. Li, W. C. Qiao, T. Li, H. J. Zhang, T. L. Feng, X. D. Xu, L. H. Zheng, J. Xu, Y. G. Wang, and Y. S. Wang, "Sub-100 ns passively Q-switched Nd:LuAG laser with multi-walled carbon nanotube," Opt. Laser Technol. **64**, 7–10 (2014).
7. Y. Q. Li, J. Liu, H. T. Zhu, L. H. Zheng, L. B. Su, J. Xu, and Y. G. Wang, "Performance of diode-pumped Tm^{3+}:Sc_2SiO_5 crystal

passively *Q*-switched 2 μm laser," Opt. Commun. **330**, 151–154 (2014).
8. Q. L. Bao, H. Zhang, Y. Wang, Z. H. Ni, Y. L. Yan, Z. X. Shen, K. P. Loch, and D. Y. Tang, "Atomic-layer graphene as a saturable absorber for ultrafast pulsed lasers," Adv. Funct. Mater. **19**, 3077–3083 (2009).
9. X. L. Zhang, X. Zhao, Z. B. Liu, Y. S. Liu, Y. S. Chen, and J. G. Tian, "Enhanced nonlinear optical properties of graphene-oligothiophene hybrid material," Opt. Express **17**, 23959–23964 (2009).
10. Z. P. Sun, T. Hasan, F. Torrisi, D. Popa, G. Privitera, F. Q. Wang, F. Bonaccorso, D. M. Basko, and A. C. Ferrari, "Graphene mode-locked ultrafast laser," ACS Nano **4**, 803–810 (2010).
11. X. L. Li, J. L. Xu, Y. Z. Wu, J. L. He, and X. P. Hao, "Large energy laser pulses with high repetition rate by graphene *Q*-switched solid-state laser," Opt. Express **19**, 9950–9955 (2011).
12. C. Feng, D. H. Liu, and J. Liu, "Graphene oxide saturable absorber on golden reflective film for Tm:YAP *Q*-switched mode-locking laser at 2 mum," J. Mod. Opt. **59**, 1825–1828 (2012).
13. V. R. Sorochenko, E. D. Obraztsova, P. S. Rusakov, and M. G. Rybin, "Nonlinear transmission of CO_2 laser radiation by graphene," Quantum Electron. **42**, 907–912 (2012).
14. G. Q. Xie, J. Ma, P. Lv, W. L. Gao, P. Yuan, L. J. Qian, H. H. Yu, H. J. Zhang, J. Y. Wang, and D. Y. Tang, "Graphene saturable absorber for *Q*-switching and mode locking at 2 μm wavelength [Invited]," Opt. Mater. Express **2**, 878–883 (2012).
15. J. Ma, G. Q. Xie, P. Lv, W. L. Gao, P. Yuan, L. J. Qian, U. Griebner, V. Petrov, H. H. Yu, H. J. Zhang, and J. Y. Wang, "Wavelength-versatile graphene-gold film saturable absorber mirror for ultra-broadband mode-locking of bulk lasers," Sci. Rep. **4**, 5016 (2014).
16. H. W. Chu, S. Z. Zhao, T. Li, K. J. Yang, G. Q. Li, D. Li, J. Zhao, W. C. Qiao, J. Q. Xu, and Y. Hang, "Dual-wavelength passively *Q*-switched Nd, Mg:$LiTaO_3$ laser with a monolayer graphene as saturable absorber," IEEE J. Sel. Top. Quantum Electron. **21**, 1600705 (2015).
17. C. J. Jin, Y. Bai, L. F. Li, T. Jiang, Z. Y. Ren, and J. T. Bai, "A single-frequency, graphene-based passively *Q*-switched Tm:YAP laser," Laser Phys. **25**, 015001 (2015).
18. H. H. Yu, H. Zhang, Y. C. Wang, C. J. Zhao, B. L. Wang, S. C. Wen, H. J. Zhang, and J. Y. Wang, "Topological insulator as an optical modulator for pulsed solid-state lasers," Laser Photon. Rev. **7**, L77–L83 (2013).
19. Z. Q. Luo, C. Liu, Y. Z. Huang, D. D. Wu, J. Y. Wu, H. Y. Xu, Z. P. Cai, Z. Q. Lin, L. P. Sun, and J. Weng, "Topological-insulator passively *Q*-switched double-clad fiber laser at 2 μm wavelength," IEEE J. Sel. Top. Quantum Electron. **20**, 0902708 (2014).
20. Q. H. Wang, K. Kalantar-Zadeh, A. Kis, J. N. Coleman, and M. S. Strano, "Electronics and optoelectronics of two-dimensional transition metal dichalcogenides," Nat. Nanotechnol. **7**, 699–712 (2012).
21. C. Ataca, H. Şahin, and S. Ciraci, "Stable, single-layer MX2 transition-metal oxides and dichalcogenides in a honeycomb-like structure," J. Phys. Chem. C **116**, 8983–8999 (2012).
22. B. Radisavljevic, A. Radenovic, J. Brivio, V. Giacometti, and A. Kis, "Single-layer MoS_2 transistors," Nat. Nanotechnol. **6**, 147–150 (2011).
23. K. F. Mak, C. Lee, J. Hone, J. Shan, and T. F. Heinz, "Atomically thin MoS_2: a new direct-gap semiconductor," Phys. Rev. Lett. **105**, 136805 (2010).
24. A. Carvalho, R. M. Ribeiro, and A. H. C. Neto, "Band nesting and the optical response of two-dimensional semiconducting transition metal dichalcogenides," Phys. Rev. B **88**, 115205 (2013).
25. A. Splendiani, L. Sun, Y. B. Zhang, T. S. Li, J. Kim, C. Y. Chim, G. Galli, and F. Wang, "Emerging photoluminescence in monolayer MoS_2," Nano Lett. **10**, 1271–1275 (2010).
26. K. P. Wang, J. Wang, J. T. Fan, M. Lotya, A. O'Neill, D. Fox, Y. Y. Feng, X. Y. Zhang, B. X. Jiang, Q. Z. Zhao, H. Z. Zhang, J. N. Coleman, L. Zhang, and W. J. Blau, "Ultrafast saturable absorption of two-dimensional MoS_2 nanosheets," ACS Nano **7**, 9260–9267 (2013).
27. H. Zhang, S. B. Lu, J. Zheng, J. Du, S. C. Wen, D. Y. Tang, and K. P. Loh, "Molybdenum disulfide (MoS_2) as a broadband saturable absorber for ultra-fast photonics," Opt. Express **22**, 7249–7260 (2014).
28. Z. Q. Luo, Y. Z. Huang, M. Zhong, Y. Y. Li, J. Y. Wu, B. Xu, H. Y. Xu, Z. P. Cai, J. Peng, and J. Weng, "1-, 1.5-, and 2-μm fiber lasers *Q*-switched by a broadband few-layer MoS_2 saturable absorber," J. Lightwave Technol. **32**, 4077–4084 (2014).
29. S. X. Wang, H. H. Yu, H. J. Zhang, A. Z. Wang, Z. M. W. Zhao, Y. X. Chen, L. M. Mei, and J. Y. Wang, "Broadband few-layer MoS_2 saturable absorbers," Adv. Mater. **26**, 3538–3544 (2014).
30. J. Ma, G. Q. Xie, P. Lv, W. L. Gao, P. Yuan, L. J. Qian, H. H. Yu, H. J. Zhang, J. Y. Wang, and D. Y. Tang, "Graphene mode-locked femtosecond laser at 2 μm wavelength," Opt. Lett. **37**, 2085–2087 (2012).
31. M. Liu, X. W. Zheng, Y. L. Qi, H. Liu, A. P. Luo, Z. C. Luo, W. C. Xu, C. J. Zhao, and H. Zhang, "Microfiber-based few-layer MoS_2 saturable absorber for 2.5 GHz passively harmonic mode-locked fiber laser," Opt. Express **22**, 22841–22846 (2014).