

REVIEW OPEN ACCESS

Review on Laser Initiation of Energetic Materials

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ABSTRACT

Laser initiation of energetic materials has been an interesting and promising research area due to its safety and reliability features and its great potential for initiation device miniaturisation. This article has critically reviewed its research in terms of the effects of essential initiation parameters, including laser wavelength, optical absorption and laser power on its performance. It aims to show the research and development of laser ignition initiation applications, in particular explosive devices. In addition, this article may also give guidance and recommendations for future research and development in laser initiation applications to energetic materials.

1 | Introduction

Explosive devices have been used in many applications, including well perforation in oil and gas industries, accurate demolition of buildings in the construction industry, blasting of rock in mining and quarrying industries and release system applications in the space industry. Such devices are also used in various ammunition and ordnance for defence applications. These devices are traditionally initiated using a three-step explosive train, as shown in Figure 1. The booster usually consists of a sensitive secondary explosive such as low-density pentaerythritol tetranitrate (PETN). The main charge consists of a low-sensitive secondary explosive such as cyclotrimethylenetrinitramine (RDX), which is reasonably safe to handle, store and transport. However, the detonators mostly consist of a sensitive primary explosive and a metal bridge wire, as shown in Figure 2 [1]. These components raise multiple safety and environmental concerns in addition to the corrosion and surface degradation of the metal components. Primary explosives are normally lead azide with Figures of Insensitiveness (FoI) of 20 (much more sensitive to unplanned initiation than RDX with an FoI of 80) [2]. The primary explosives are easy to initiate but unsafe due to the risk of accidental or premature initiation,

particularly under extreme temperature and pressure conditions, for example, in well perforation [3–5]. Furthermore, they are hazardous to health and environment due to the toxicity of the heavy metals that are restricted by legislation such as REACH (i.e., European Union regulation concerning the Registration, Evaluation, Authorisation and restriction of Chemicals). Exploding Bridge Wire Detonators (EBW) and Exploding Foil Initiators (EFI) [6] (or Slapper Foil type in Figure 2) that are often used for both military and non-military applications do not use primary explosives and are therefore safer than other types of detonators. However, the metal wires used for the detonators are subject to electromagnetic interference (EMT) and stray electrical energy and may lead to misfires of the devices by possible sources such as lightning and radio frequency (RF) radiation originating in radio communication or mobile wireless devices. These interferences may cause disastrous consequences to both life and property and must be avoided during explosive operations, for example, in well perforation, by suspending routine well-site operations such as radar, radio, cellular communications, cathodic protection and welding, and stopping the operations in severe weather conditions. Therefore, removing both the primary explosive and metal component from a detonator will offer a cleaner,

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reliable detonator with the immunity to EMT and stray electrical energy.

Similarly for defence applications, laser initiation can be used for fuzes in weapon and missile systems that initiate the booster and main explosive charges. A conventional fuze [7] normally contains a safety and arming unit (SAU) to ensure safe initiation and to protect users from premature or accidental initiation by providing the required safety features such as serial locks, mechanical shutters and immunity from inadvertent aiming and firing by stray electromagnetic and electrostatic stimuli. Laser initiation not only removes the metal parts and sensitive primary explosives in the initiators but will also remove the need for complex mechanical safety and arming systems. Laser initiation systems may be directly constructed in-line with a laser shutter and without the need for any safety concerns. Furthermore, modern weapon systems usually have many electric explosive actuators or devices that require cable looms and safety control units. Replacing these with laser initiation using miniature laser diodes and light fibre optics or opto-pyrotechnics will significantly reduce the overall system weight and remove the significant hazard from lightning and EMT.

Laser initiation is where optical energy at a selected wavelength is delivered to directly initiate a low-sensitive or insensitive secondary explosive and eliminate the above problems. An initiator based on this mechanism would not require a primary explosive and a metal wire and therefore provide clean and reliable technology. This technology would be able to replace existing initiation methods and provide a new generation of initiation technology in energetics for the medium to long term. Research has been carried out world-wide as early as the 60s' that exploits the advantages of this technology and investigates its relevant parameters for laser initiation development [8–10]. The earliest research carried out by Brish et al. in 1966 [8] studied laser-excited initiation of energetic materials, including lead azide and PETN, using a Q-switched Nd:Glass laser, which successfully initiated prompt detonation. Early research was carried out by Jungst et al. [11] in studying laser initiation of explosive and pyrotechnic components by using a continuous wave (CW) diode laser, where the laser induced a flame in the materials. Here, the laser initiation threshold of Ti/KClO₄ and the inorganic explosive, 2-(5-cyanotetrazolato) pentaamine cobalt (III) perchlorate (CP), were investigated. There were also some published studies that had reviewed the progress and benefits achieved from using laser initiation technology for explosives. Ronney [12] carried out a review on the comparison between the initiation by electrical mechanisms and laser sources. He claimed that there were some advantages (other than those mentioned above) of using laser initiation such as the ability to choose the location(s) and timing of initiation events. de Yong et al. [13] had an earlier review that critically examined the state of technology of laser initiation of explosives, pyrotechnics and propellants and concluded that the only possible approach to laser initiation of explosives in a fuze is via a laser-driven flyer plate, because the cost and size of the prototypes are too great for incorporation in most practical systems. A latter review by Bourne [14] on laser ignition of explosives claimed several advantages of laser initiation over electric initiation such as immunity to electrical effects and chemical stability. Most recently, Deng et al. [15] carried out a critical review on the performance and progress of

photothermal materials for laser initiation applications. In their review, the designs of photothermal materials with optimised performance were discussed. This may provide useful advice on the choice of photothermal energetic materials for laser initiation applications such as carbon or metallic nanoparticle-modified energetic materials.

This article will provide a review on the latest experimental research published on laser initiation for explosive applications. Here, a successful laser ignition is defined as self-sustained combustion that is induced by laser irradiation. This article only focuses on the effects of essential laser initiation parameters, including laser wavelength and the optical absorption of energetic materials and laser power and energy on the initiation performance. The future potential research required for laser ignition technology is also covered. This article may also offer a specific and useful guidance on the requirement and selection of the essential laser parameters for optimum laser initiation performance.

2 | Effects of Laser Wavelength on Ignition

Wide and extensive research in laser initiation of energetic materials at various laser wavelengths has been carried out. Early research by Brish et al. [16] carried out an experimental study to investigate the possible mechanism of laser-excited detonation of secondary explosives, including PETN and lead azide, at two different laser wavelengths, 1060 and 694.3 nm, using pulsed neodymium glass and ruby lasers. The result from the research showed that the dependence of critical laser energy density on beam diameter for the two laser wavelengths was preserved and consistent with the results of measurements of the reflection coefficient at these wavelengths. The similarity of critical laser energy densities for these wavelengths was attributed to the similarly high amount of the incident light reflected (about 80%) by the explosive surface for both wavelengths that lie in the region of weak absorption of the explosives. The analysis of their experimental results supported the thermal mechanism of the laser-excited detonation, and it was concluded that the pulsed laser energy was consumed in heating the explosives to form hot spots, rapidly introduced into the explosives and converted into shock-wave energy, which excited the prompt detonation. A thermal model for a three-dimensional reactive energetic material was studied by Langer et al. [17], where they simulated the formation and development of hot spots in energetic materials. Earlier, Field et al. [18] carried out a comprehensive study on hot-spot initiation of energetic materials and concluded that hot-spot production took place in a variety of sample forms, including single crystals, powder layers, pressed pellets, gel and polymer-bounded explosives (PBXs). Zinchenko et al. [19] studied the optical characteristics of trotyl, PETN, RDX and octogen (HMX) using a laser at wavelength of 632.8 nm. Using experimental measuring and theoretical modelling of the coefficients of reflection, transmission and absorption, they determined the distribution of absorbed light energy density with depth in plane layers of the explosive samples in order to understand the physics of laser-induced initiation. Deng et al. [15] concluded that for laser initiation applications of energetic materials, like explosives and pyrotechnics, materials, which should have good optical absorption and opto-thermal conversion

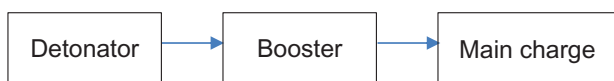


FIGURE 1 | An explosive train.

properties are required. Optical absorption of light by materials depends significantly on the light wavelength in addition to material properties such as density (or concentration in liquid form). Such dependence follows the Beer–Lambert law [20] as expressed by Equation (1) for the intensity of the incident light, I_0 , on a material surface and transmitted light, I , from the material:

$$\log \left(\frac{I_0}{I} \right) = A = \epsilon lc \quad (1)$$

where

- A is the absorbance.
- ϵ is the absorptivity of the attenuating species which is wavelength dependant.
- l is the optical path length through the species.
- c is the concentration of the species.

Therefore, the wavelength of a laser plays a decisive role in its optical absorption capability within materials and their laser initiation properties. Absorption spectra of many energetic materials have been extensively studied and used for the choice of laser wavelengths in initiation applications. Cooper et al. [21] have carried out both experimental and theoretical studies on optical absorption of six explosive molecules, RDX, HMX, PETN, trinitrotoluene (TNT), triacetone triperoxide (TATP) and hexamethylene triperoxide diamine (HMTD). In their research, the optical absorption spectra in the ultraviolet (UV)–visible region of these explosives, which were diluted in acetonitrile, were measured, and calculation of these spectra was performed by utilising time-dependent density function theory (TD-DFT). From their spectra, see Figure 3, all six explosives exhibited strong absorption peaks at UV wavelengths, ~ 230 nm for RDX, HMX and TNT and ~ 190 nm for PETN, TATP and HMTD. A similar experimental study was carried out by Feagin et al. [22] using a Thermo Scientific UV/Vis spectrophotometer on five commonly used explosives, PETN, HMX, RDX, hexanitrostilbene (HNS) and hexanitrohexaazaisowurtzitane (CL-20). As shown in Figure 4, PETN, HMX and RDX have optical absorption peaks around the same wavelengths as the UV region (see Figure 3). The UV–Vis absorption spectrum of an insensitive explosive FOX-7 (1,1-diamino-2,2-dinitroethylene) was investigated by Šimková et al. [23]. Absorption bands at 280 and 350 nm were observed for FOX-7. Furthermore, these spectra show a weak optical absorption peak at wavelengths in the visible to the infrared (IR) region with increasing penetration depths. These optical absorptions as a function of wavelength clearly suggest that laser radiation at UV wavelengths would be favoured in the laser initiation of explosives in terms of more efficient optical energy conversion.

An experimental study was carried out by Ahmad et al. [24] on the comparison of laser ignition of sulphur/charcoal/potassium nitrate-based compositions at laser wavelengths of 500 nm (Ar-

ion laser) and 784 nm (diode laser). They concluded that lasers at the visible wavelength initiated a faster ignition than that at the near-IR (NIR) wavelength. Paisley [25] had studied the mechanism of prompt detonation by pulsed lasers at various wavelengths from 266 to 1.06 μm for secondary explosives, including PETN, HNS and HMX, together with graphited PETN, HNS and HMX. His study suggested that there were two competing initiating mechanisms for the pulsed laser-induced prompt detonation at UV and NIR wavelengths, these being photodissociation leading to molecular bond breaking and thermal decomposition by hot spots leading to detonation, respectively. The latter required confining the laser/explosive surface with an optical window. Östmark et al. [26] used a tuneable CO_2 laser at 9–11 μm to ignite the explosive RDX. They measured the effect of laser wavelength on the threshold ignition energy of explosives and showed that laser absorption in the material followed the Lambert–Beer law, and the absorption depth of the irradiated material was controlled mainly by the material's absorption at the wavelength. It was also concluded that the Lambert–Beer absorption in the solid phase of the explosive led to its decomposition into gaseous products. It was also concluded that at larger optical absorption depths (e.g., >25 μm), a larger volume of explosive is heated, ‘confining’ the decomposition and thus lowering the laser ignition energy. Using a CO_2 laser of high-irradiance (up to 800 W cm^{-2}) at a wavelength of 10.6 μm , Ali et al. [27] performed an experimental study with a simple dual ignition criteria model on laser ignition of HMX and triaminotrinitrobenzene (TATB) and reported the time to ignition as a function of laser irradiance, energy and ignition temperature. Rapid heating by laser irradiation caused thermal decomposition and consequently the ignitions. Thus, the optical absorption of a specific explosive at the wavelength of a laser plays an essential role in laser ignition. An earlier investigation into an interesting application of laser ignition was in large-calibre gun propulsion systems. This was conducted by Barrows et al. [28] which showed the importance of the material's optical absorption by coating or dispersing graphite on the sample surface. In their study, two high-energy Nd:glass lasers (Laser Photonics) served as ignition sources with variable pulsed laser energy of up to 30 J at a wavelength of 1050 nm.

More comparative evaluations of laser ignition at various wavelengths were performed on RDX which is a common secondary explosive and has strong optical absorption [29] in the UV region of the EM spectrum. Renlund et al. [9] demonstrated that RDX can be directly detonated by a 24-ns pulse from a UV laser of 308-nm wavelength, but not by a 20-ns pulse from an IR laser of 1064-nm wavelength. Yan et al. [30, 31] studied laser ignition sensitivity of RDX crystal surface defects. Micro-explosions on the crystal surface were induced by a 6.4-ns pulse from a 355-nm UV laser with radiant energy densities of 12 J cm^{-2} . McGrane et al. [32] demonstrated in their study that a CW visible laser of 532-nm wavelength was able to ignite pure RDX with a long exposure time of 120 s at laser powers greater than 0.6 W. However, ignition did not occur with an IR laser of 1550 nm wavelength, although melting and decomposition did occur at a high laser power of 1.4 W.

As understood from the above research, explosives have shown their ease of ignition and detonation by lasers at UV wavelengths. However, UV lasers are bulky and expensive, which

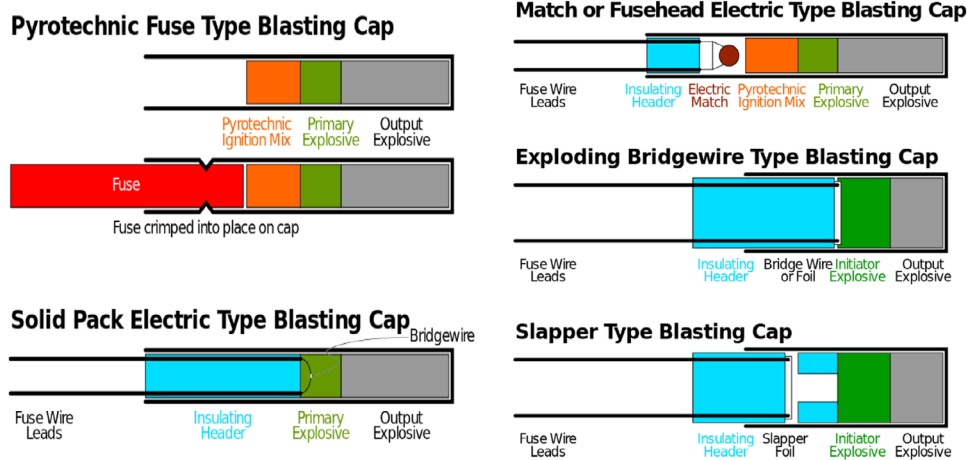


FIGURE 2 | Types of blasting detonator [1].

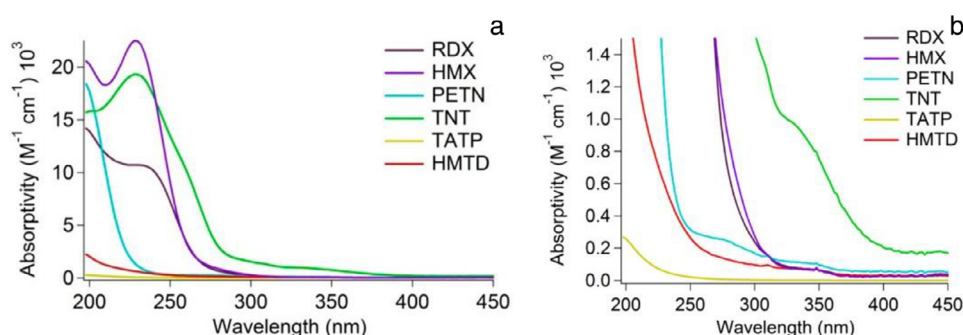


FIGURE 3 | Experimentally determined UV-Vis absorption spectra for six explosives: RDX (black), HMX (purple), PETN (cyan), TNT (green), TATP (yellow) and HMTD (red) in the full (a) and expanded (b) perspectives [21]. EMT, electromagnetic interference; HMTD, hexamethylene triperoxide diamine; PETN, pentaerythritol tetranitrate; RDX, cyclotrimethylenetrinitramine; TATP, triacetone triperoxide; TNT, trinitrotoluene.

make them unsuitable for detonator applications, whereas diode lasers, which operate in the IR-to-Vis (IR-Vis) region, are small, inexpensive and easy to integrate, thus much preferred for use as an initiation light source [33]. However, IR lasers lack sufficient optical absorption in the visible to IR wavelength bands; therefore, to be able to use these lasers, additives with good optical absorption are commonly added to explosives to aid initiation.

3 | Effects of Optical Sensitisers in Explosives on Ignition

A variety of laser-absorption additives have widely been used as optical sensitisers in explosives for laser ignition applications, and significant improvements of ignition have been achieved. Carbon black (CB) is a commonly used optical sensitiser, thanks to its good optical absorption through the wavelengths ranging from 200 to 2500 nm as seen in Figure 5 [34]. CB can be added to explosives to assist and enhance optical absorption of the energetic materials in ignition applications.

A very early study of laser ignition of explosives was carried out by Barbarisi et al. [35] in 1969. He discovered that ignition by a laser could be improved with the addition of a black agent within the explosive. Since then, more research has been conducted to

enhance absorption of the laser in explosives in order to achieve successful ignition. In an early study, Kuznetsov et al. [36] ignited cylindrical samples of HMX mixed with 1% CB powder using a light flux. In their work, a comparative evaluation of the optical absorptions was made by measuring the reflection coefficients of samples of HMX and HMX + 1%CB. It was reported that by adding CB, the reflection coefficient of HMX reduced from 87% to 11% at a wavelength of 360 nm and from 93% to 12% at 1000 nm. This observation shows almost no wavelength selectivity for the CB additive. The required radiant fluxes to ignite the explosive for a specific pulse duration were $3.3 \times 10^5 \text{ W m}^{-2}$ for ignition of HMX + 1% CB and $29 \times 10^5 \text{ W m}^{-2}$ for HMX. CB has been proven to be an effective additive for optical sensitisation in laser ignition of energetic materials and the most widely studied, especially in secondary explosives and pyrotechnics. Paisley [25] used CB with secondary explosives PETN, HNS and HMX and achieved successful ignition by using lasers at wavelengths from 266 to 1.06 μm . Goveas et al. [37] used CB in their study as an additive in the secondary explosive HMX and showed that increasing the additive concentration reduced the required laser power for ignition of HMX. Similar studies carried out by Taskai et al. [38], Skocypec et al. [39] and Munger et al. [40] showed a significant increase in optical absorption in the explosives when CB was added and a reduction of the laser energy threshold for ignition of explosives.

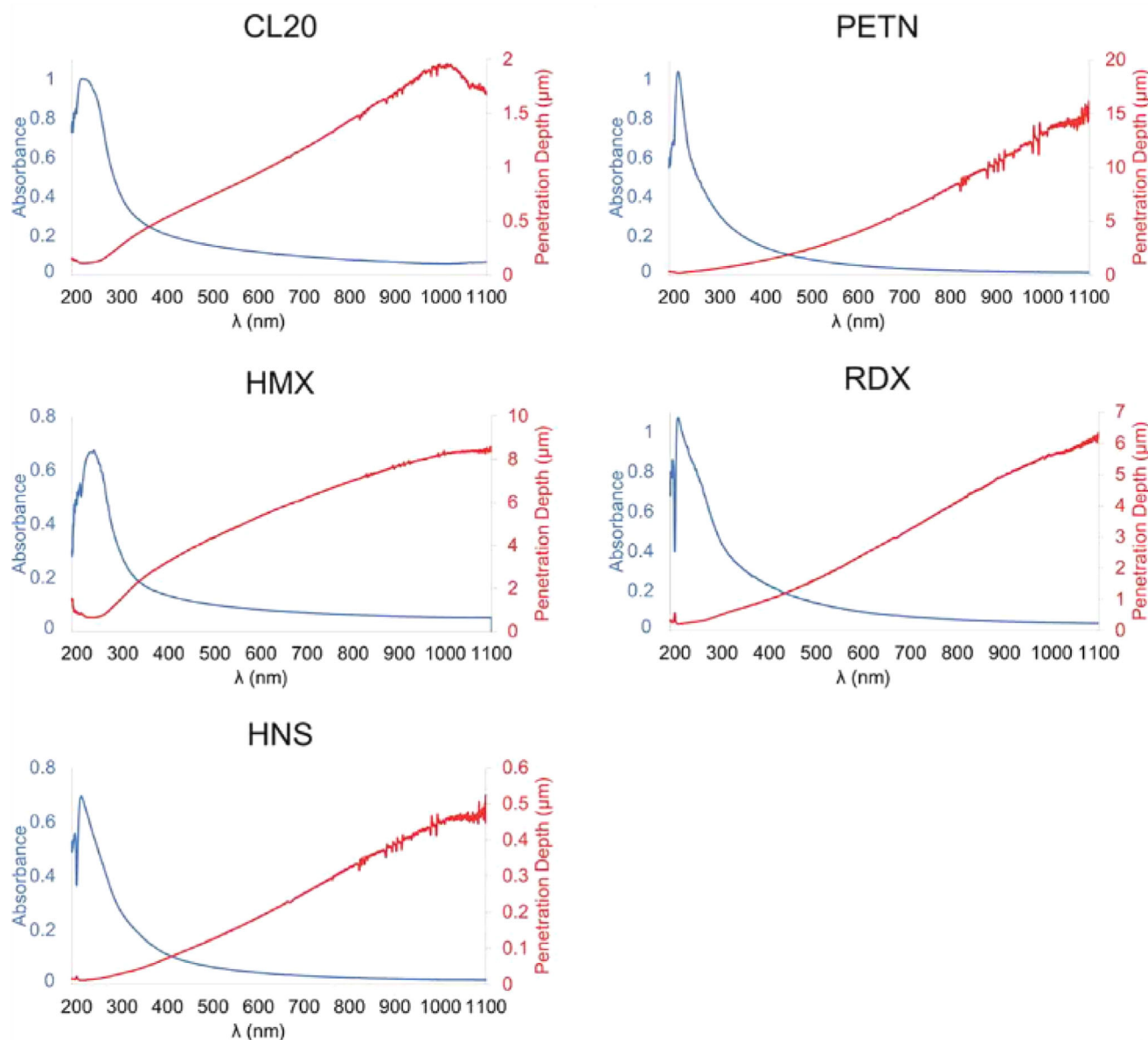


FIGURE 4 | The absorbance and penetration depth of investigated HE powders as a function of wavelength [22]. CL-20, hexanitrohexaazaisowurtz-itane; HNS, hexanitrostilbene; PETN, pentaerythritol tetranitrate; RDX, cyclotrimethylenetrinitramine.

More recently, Fang et al. [41] have carried out research on optical sensitisation of low-sensitive secondary explosive HNS IV by mixing it with CB and a laser-absorbing dye using the same mixing concentration of 5 wt.% for both additives. The optically sensitised HNS IV samples were easily ignited (i.e., only combustions were induced in open air) by a diode laser of 808 nm wavelength with a minimum laser power output of 4 W; the ignition delay times at laser power of 45 W were 2.1 ms for HNS IV/dye and 1.2 ms for HNS IV/CB. The samples of pure HNS IV without sensitisation were not ignitable at a maximum laser power of 45 W. This result shows that an enhancement of laser ignitability of sensitised HNS IV was greater than 10-fold and that CB and the dye had similar behaviours as an optical sensitizer for HNS IV. Fang et al. [42] also carried out research on laser ignition of the insensitive explosive FOX-7 and achieved significant enhancement of laser ignitability of FOX-7 when mixed with 5 wt.% CB using a diode laser at a wavelength

of 974 nm. The minimum laser power required to ignite optically sensitised FOX-7 was below 10 W, and ignition was achieved as short as 70 μs when using laser power of 40 W. Their latest study [43] has investigated the methods of mixing FOX-7 with CB. They report that a ground mixing method provides a more uniform dispersion of CB in the mixed samples, as shown in Figure 6a,b. This results in a lower laser ignition threshold than the tumble mixing method.

Other laser-absorbing additives have been investigated as optical sensitizers in laser ignition applications. Khanef, Mitrofanov and Dolgachev et al. [44–47] investigated the application of a copper oxide (CuO) film to organic explosive materials. They experimentally evaluated the effect of the absorption coefficient of a CuO film on the laser ignition delay of PETN. The ignition of explosives, including PETN, RDX, HMX and TATB with a CuO film, was achieved by using a pulsed fibre laser. In their studies,

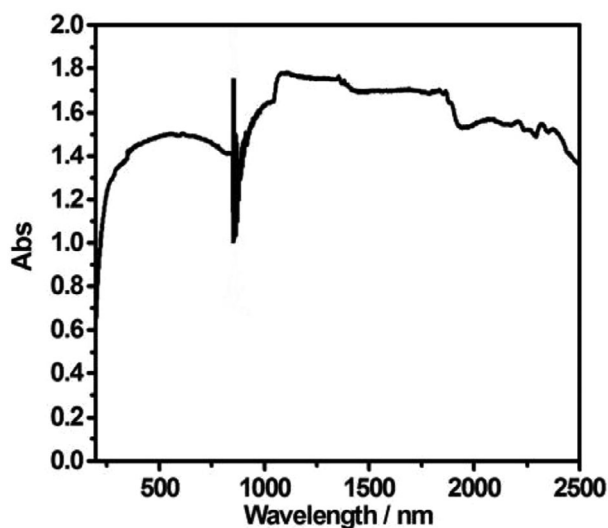


FIGURE 5 | UV-Vis-NIR absorption spectrum of carbon black powder [34].

the CuO film was obtained by pressing CuO particles of micron size to a density close to that of a single crystal. This film acted as a laser-absorbing material with good thermal conductivity which resulted in the ignition of explosives due to thermal mechanisms.

Metallic nanoparticles are also very interesting additives; they have good optical absorption and photothermal properties due to their surface plasmonic resonance (SPR) [48–53]. These plasmonic nanoparticles have a specific optical absorption band which is dependent on the particles shape and size [48]. Such features of metallic nanoparticles have been exploited for laser-induced heat generation in applications such as solar cells [54], photonic devices [55] and extensively in biomedical applications, that is, human oesophagogastric adenocarcinoma [56], cancer thermal therapy [57, 58] and biomedical applications [59, 60]. These applications mostly used gold nanoparticles (GNPs) which acted as heaters induced by lasers. On the basis of this photothermal mechanism of plasmonic metallic nanoparticles as electromagnetic absorbers [61], metallic nanoparticles have been used in the application of laser ignition of explosives.

Most recently, Fang et al. [62] have experimentally investigated the feasibility of using gold nanorods (GNRs) of 10 nm in diameter and 40 nm in length to optically sensitise an insensitive explosive FOX-7 by coating the powdered FOX-7 with commercially available GNRs, as shown in Figure 7. In their research, a CW diode laser at the wavelength of 808 nm was used to excite the SPR of the GNRs which has a maximum optical absorption at this wavelength, as shown in Figure 8. Such optically sensitised FOX-7 (pressed pellet) was easily ignited by a minimum laser power of around 8 W and laser beam size of ~1 mm, whereas pure FOX-7 (without the GNRs coating) was unable to ignite with a laser power of up to 45 W (the maximum power available for the study). As a typical example of the successful application of GNRs-sensitised FOX-7, a laser initiator was designed in this study and shown in Figure 9. This device was evaluated by firing tests where laser initiation of FOX-7/GNRs resulted in the metallic disc rupturing and impacting a pressure sensor. The impact pressure generated by the flying metallic disc was sufficient to detonate a secondary explosive like PETN or HMX.

Using a CW diode laser of 808 nm wavelength, experimental studies were carried out by Fang and Churchyard et al. [63, 64] to investigate the laser ignitability of RDX which was recrystallised with GNRs. Here, the GNRs were embedded inside the RDX crystals which significantly enhanced the laser ignitability of RDX and reduced the laser ignition threshold power to as little as 1 W (or fluence $\sim 2.83 \text{ J cm}^{-2}$) with a delay of $\sim 28 \text{ ms}$, indicating that the threshold laser power requirement for the ignition of RDX crystals was lowered by at least 45-fold [63]. This embedding method for optical sensitisation of RDX crystals was found to be superior in terms of optical sensitisation and heating efficiency within the crystals compared to the coating by simple mixing. This result should, therefore, facilitate the development of a miniature low-power laser initiator. In their later experimental study [64], two different sizes of GNRs were used as optical sensitisers which were embedded inside RDX crystals, the smaller being $10 \times 40 \text{ nm}$ and the larger $40 \times 160 \text{ nm}$; both GNRs had the same diameter-to-length ratio of 1:4 and SPR wavelength of 808 nm. The research showed that the size of GNRs does not dramatically change the ignition delay time of the RDX crystals. However, the smaller GNRs reduced the power threshold, given a constant size, for laser ignition by 25% compared to large GNRs; this is due to smaller GNRs being able to absorb more laser energy. The results suggested the use of smaller GNRs for laser ignition applications. Furthermore, Fang et al. [65] have carried out experimental studies on pulsed laser initiation of RDX crystals which were embedded with gold nanospheres (GNSs) having a SPR wavelength of around 532 nm. A pulsed YAG:Nd laser at 532 nm wavelength and 6 ns pulse duration with a laser spot diameter of 1.2 mm was used as the initiating laser source. In this novel research, a high-speed camera was used to capture the modes of interaction between the GNS-doped RDX crystals and pulsed laser irradiation. A new classification method with defined mode of initiation was introduced to classify the observed initiation reaction of RDX crystals. It was observed that both pure RDX crystals and CB-coated RDX crystals showed no signs of initiation, whereas GNS-doped RDX crystals initiated at a low energy density of 0.04 J cm^{-2} . The results from this research showed that the GNSs-sensitised RDX crystals were three orders of magnitude more sensitive than pure RDX crystals to pulsed laser irradiation at 532 nm wavelength. The research also concluded that GNSs are effective optical sensitisers for direct initiation of RDX crystals by low-energy pulsed laser.

In addition to the noble metals like GNPs as optical sensitisers, other materials, like aluminium nanoparticles (AlNPs), have surface plasmon resonance properties and have been used in laser ignition of energetic systems. Uhlenhake et al. [66] in their experimental study successfully ignited films of nano-aluminium (Al) particles combined with a polyvinylidene fluoride binder/oxidizer using a low energy broadband xenon flash source and a Nd:YAG laser in CW mode at wavelengths of 1064 and 532 nm. Laser ignition showed that the plasmonic resonant enhancement of heating was more significant at shorter wavelengths near 250 nm than at longer wavelengths of 1064 and 532 nm. Xenon flash ignition also showed that plasmonic thermal effects were more effective at wavelengths from 200 to 400 nm. Ohkura et al. [67] used a camera xenon flash to successfully ignite AlNPs through photothermal effects. Their results showed that for successful optical ignition, AlNPs require a submicron diameter range and a high packing density in order to achieve

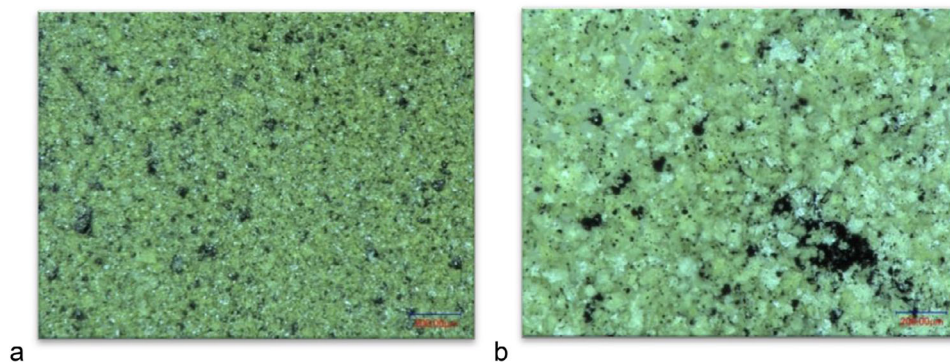


FIGURE 6 | Microscopic images (200 $\times$ zoom) of the pressed samples of mixture FOX-7/CB using the methods: (a) ground mix and (b) tumble mix [43].

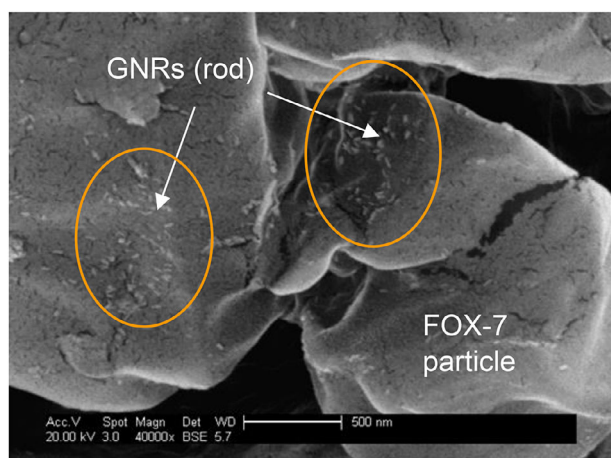


FIGURE 7 | SEM images of sensitised FOX-7 with citrate-capped GNRs [62]. GNRs, gold nanorods.

a sufficient temperature rise for ignition. In order to manipulate photo-ignition using AINPs, Mutlu et al. [68] fabricated AINPs with well-defined nano-sizes and shapes. They observed well-controlled heating and ignition of the AINPs using a laser of wavelength 532 nm and power up to 200 mW. Ji et al. [69], using a short laser pulse of 14 ns duration and an NIR wavelength of 1064 nm, achieved direct initiation to detonation of CL-20 doped with AINPs. The laser energy for detonation was reduced from >1 J for undoped CL-20 to 0.02 J for the AINP-doped CL-20. This is a typical example of using AINPs to achieve SPR at an IR spectral region which results in rapid hotspot formation and deflagration to detonation in an explosive where a laser pulse provides significantly high power. Diwu et al. [70] investigated laser-absorption enhancement of HMX doped with AINPs and GNPs, respectively. The study showed that for a laser wavelength 808 nm and metallic nanospheres up to 100 nm, AINPs with a thickness of 1–2 nm and GNPs thickness of 3–4 nm provided the best absorbing performance. GNPs were observed to be better than AINPs as the doping metal particles. Their experimental results showed that laser absorption of energetic materials was significantly enhanced by doping with GNPs which had SPR at wavelength of 820 nm. Compared to gold or silver, Al is a relatively economical and manipulable material and therefore more attractive for use in laser initiation of energetic materials. AINPs plasmonics has also been widely studied [71–76]. The

authors have concluded that AINPs have stronger SPR at the UV to visible wavelength region, whereas GNPs typically have stronger SPR at the visible to NIR wavelength region. Therefore, it is suggested that AINPs would be a better optical sensitizer in laser ignition applications when used for a UV laser, whereas gold or silver nanoparticles are better suited for a visible to IR laser.

4 | Effects of Laser Power and Energy on Ignition

Laser power and energy are both essential parameters, in addition to other parameters like confinement, when determining the ignition performance of energetic materials. Li et al. [77] used a CW Nd:YVO₄ NIR laser at wavelength 1064 nm. They successfully ignited pellets of polyhedral boron clusters ($[N(C_2H_5)_4]_2B_{10}H_{10}$). The results of their study showed that at a laser intensity of 1.14×10^3 W cm⁻² and a laser focus of 0.053 cm in diameter, the ignition delay time of 350 ms was achieved. The threshold ignition energy was observed to be 0.7 J at a laser beam diameter of 0.053 cm (i.e., laser fluence of 317 J cm⁻²). Using a CW NIR laser of 1064 nm wavelength, Itouyama et al. [78] observed the ignition of ammonium dinitramide-based energetic ionic liquid propellants (ADN-EILPs) with added carbon fibres. In their study, the laser ignition delay time was investigated using a range of laser powers and a fixed laser focus size of 1.9 mm in diameter. The results showed an increase in igniting laser power (from above 2.84 to 5.1 W) which resulted in a decrease in the ignition delay time (from 0.8 to 0.4 s) for the propellant sample. The threshold of laser ignition fluence was 80 J cm⁻². A laser power of ~ 5.13 W was used for the CW laser ignition in this study. Medvedev et al. [79, 80] carried out experimental investigations on laser ignition of ammonium perchlorate (AP) and Al mixtures which used a laser of 1.06 μ m wavelength with 3.5 and 0.8 ms durations. They studied the conditions of ignition, AP/Al thermostability and the effect of composition density on laser ignition. Their study showed that the optimum ignition threshold of laser fluence and its ignition delay time were observed to be 0.71 J cm⁻² and 4 ms, respectively, for powder mixtures of AP and nano-Al. Their later experimental investigations into the effect of AP/nano-Al composition density on laser ignition showed that the laser fluence threshold increased from 0.78 to 5.15 J cm⁻² as the sample packing density increased from 324 to 1893 kg m⁻³. This was explained by the decrease of sample porosity and the increase of thermal dissipation into the sample.

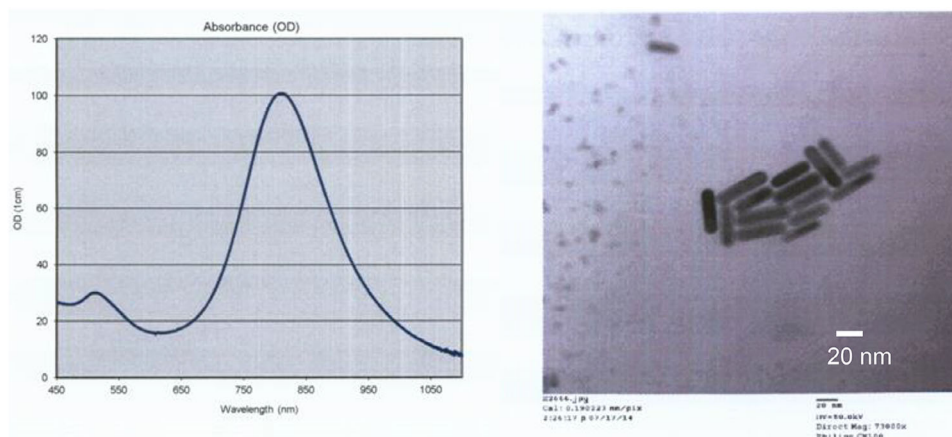


FIGURE 8 | Absorption spectrum and SEM image of GNRs used in this study [62].

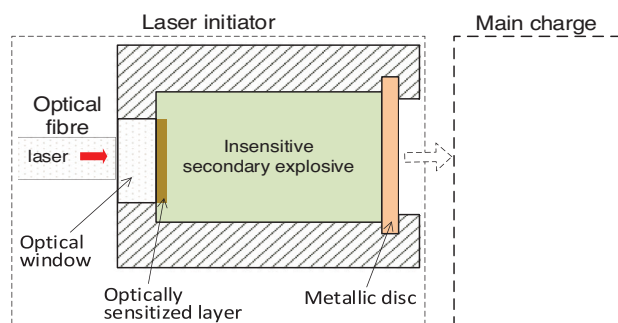


FIGURE 9 | A basic schematic of laser initiator [62].

From the information of the individual experimental results for laser ignition thresholds of fluence, it is shown that for various propellants, their required laser fluence to ignite can be lowered from 317 [1] to 80 [2] and 0.71 J cm^{-2} [4] when carbon fibres and nano-Al are added. Therefore, laser fluence may control ignition parameters. Other work carried out by Herreros et al. [81] for laser ignition of an elastomer-modified cast double-base (EMCDB) propellant using a diode laser focused on the effects of laser parameters, laser power and the duration of ignition such as ignition delay. Their research showed that ignition delay time of the propellant samples decreased significantly when the laser power increased. This can be seen in Figure 10; with ignition delay time as $\tau = 12.5/P_0^2$ where P_0 is the laser power. The ignition power threshold decreased to almost a constant level as the laser irradiation duration increased, as shown in Figure 11. These results show that the ignition performance is controlled by the laser power and energy.

Laser ignition was also used to initiate pyrotechnics, where laser power and energy play an important role in the ignition performance of pyrotechnics. In their early research, Lee et al. [82] had developed a reliable, simple pyrotechnic match head with a 1 W diode laser (980 nm wavelength) as its igniting source. A pulse energy of 2 mJ and a beam spot of 0.1 mm in diameter were sufficient for ignition of a pyrotechnic slurry which was coated on the tip of an optical fibre through which the laser was delivered. The results showed that when laser fluence of 6 J cm^{-2} was used, 80% ignition was achieved, whereas at 25 J cm^{-2} ignition rates of 100% were achieved. Opdebeck et al. [83] studied

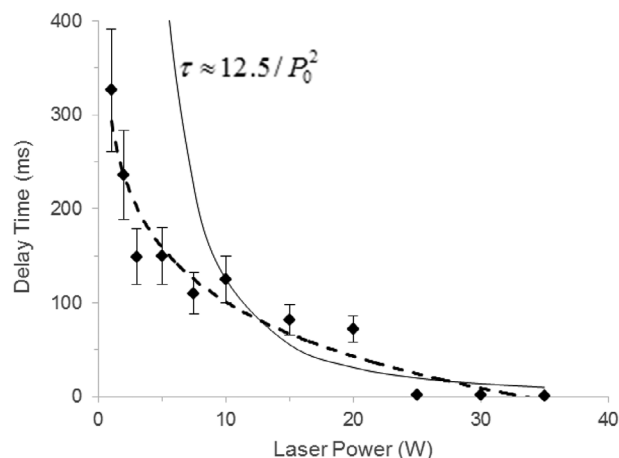


FIGURE 10 | Ignition delay time versus laser power for 0.7 mm beam width [81].

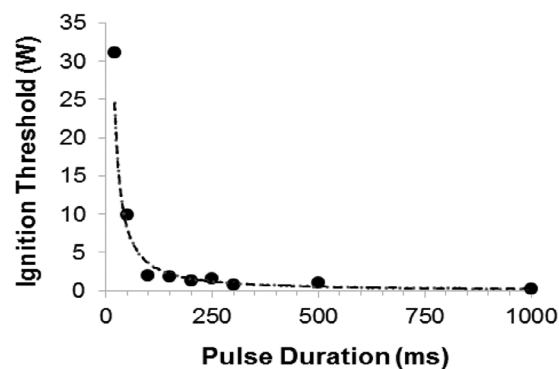


FIGURE 11 | The effect of pulse duration on ignition threshold at a laser spot size of 0.7 mm [81].

laser ignition of pyrotechnic mixtures by numerical modelling and experimental testing for validation. They used a 7 W diode laser of 820 nm wavelength and a pellet of a pyrotechnic mixture containing 62% iron and 38% KClO_4 by mass and a porosity of 25.8%. The ignition threshold of laser fluence obtained from both their ignition modelling and experiment tests showed an exponential decrease from ~ 6 to 3.3 and 9 to 3.8 J cm^{-2} , with an increase in the applied laser intensity from 0.75 to 9 kW cm^{-2} .

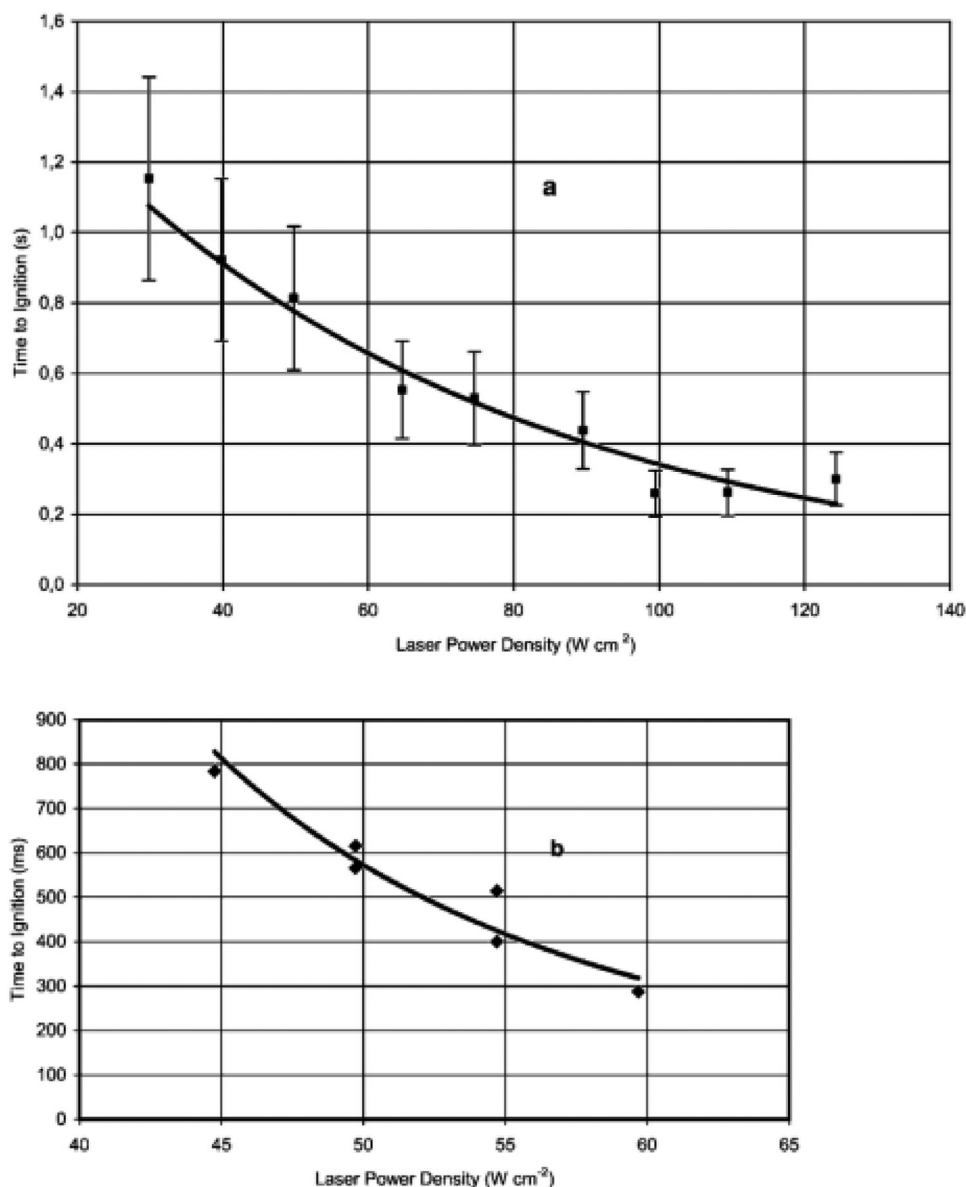


FIGURE 12 | Plot of time to ignition (ignition delay time) versus laser power density (i.e., laser intensity) for gunpowders: (a) G20 composition and (b) commercial gunpowder (CGP) [24].

Ahmad et al. [24, 84], in their earlier study, conducted experimental investigations into laser ignition of pyrotechnic materials, including a sulphur/charcoal/potassium nitrate-based composition (gunpowder), shellac binder-based boron/potassium nitrate composition (SR 44) and acaroid resin binder-based magnesium/potassium nitrate composition (SR 371C). The lasers used in their study were a visible Ar-ion laser and an NIR diode laser at wavelengths 500 and 784 nm, respectively. The experiment generated an ignition curve (Figure 12) [24] which showed that an increase in laser intensity exponentially shortens the ignition delay times for all the samples tested at both laser wavelengths. The minimum laser intensity required for successful ignition of gunpowder, SR44 and SR371C compositions was found to be 30 W cm^{-2} when using a visible laser and 120 W cm^{-2} when using an NIR laser for gunpowder, $\sim 55 \text{ W cm}^{-2}$ for SR44 and $\sim 63 \text{ W cm}^{-2}$ for SR371C, respectively. It was also observed that at higher laser intensity, the threshold laser fluence could be

drastically lowered due to faster laser heating leading to less heat loss to the surroundings.

Sivan et al. [85] experimentally investigated the ignition of pyrotechnic pellets, including TiH/KClO_4 (TiHPP), B/BaCrO_4 (BBaCrO_4), B/KNO_3 (BPN) and B/KClO_4 (BPP). A CW diode laser of 808 nm wavelength and 0.8 mm beam diameter was used as the igniting source. The research showed that ignition delay of the pyrotechnic samples depends on laser intensity and such dependence can be empirically fitted into the expression, $t_{\text{ign}} = aI^{-n}$, where t is ignition time, I is laser intensity on the sample surface and $n = 1.4\text{--}2.1$ depending on the sample materials. Therefore, an increase in the laser intensity generates a faster ignition. When the laser intensities were increased to $\sim 1.1 \text{ kW cm}^{-2}$, the ignition delay time decreased to $\sim 10\text{--}40 \text{ ms}$. Cao et al. [86] studied pyrotechnic laser ignition of airborne ejection escape systems, which used an optical fibre to transmit

the igniting laser power onto the pyrotechnics which activated the ejection of the seats. The research concluded that the laser ignition system would be an ideal pyrotechnic device for future airborne ejection escape systems, due to its novel structure, small volume, light weight, high safety and reliability.

An interesting and demanding aspect of laser initiation of energetic materials is the direct initiation of secondary explosives. Setchell et al. [87] developed a two-stage laser initiation which resulted in detonation of secondary explosives HMX and PETN. The first stage involved laser ignition of confined HMX which led to the build-up of high pressure gas which, in-turn, caused the pressure disc to rupture. The disc impacted the lower density HMX at such a high velocity that it caused the HMX to detonate on impact. Their experimental study used Nd/glass, Nd/YAG and Ti/sapphire pulsed lasers as the igniting energy sources and graphite as an explosive additive to increase optical absorption of the explosive mixtures. In this research, an overall function time of less than 50 μs was achieved, and the result also showed that decreasing laser ignition power/energy density increased the ignition time. Gottfried et al. [88–91] were able to directly initiate detonation of a secondary explosive by laser-induced plasma shockwave. They studied the explosive detonation performance using a developed technique called laser-induced air shock from energetic materials (LASEM). In their study, a focused pulsed laser of 6 ns duration at a wavelength of 1064 nm was used to initiate exothermic reactions in the laser-induced plasma on the surface of powdered secondary explosives (TNT, RDX, dinitrofurazanfuroxan (DNF) and 3,4-bis(4-nitro-1,2,5-oxadiazol-3-yl)-1,2,5-oxadiazole (LLM-172)) with metal additives, for example, Al and B. A typical laser energy of 900 mJ was used with a focused laser beam diameter of 0.8 mm. The estimated laser pulse intensity to detonate secondary explosives was $\sim 30 \text{ GW cm}^{-2}$. A detonation velocity of $\sim 8.8 \text{ km s}^{-1}$ was observed from an RDX nano-powder (NP) sample using a laser-induced shockwave of $\sim 800 \text{ m s}^{-1}$. Their research observed a linear correlation between the velocities of laser-induced shock and the detonation of a selection of explosives, including dinitroanisole (DNAN), TNT, HNS, TATB, nitrotriazolone (NTO), PETN, RDX, HMX and CL-20. They were able to estimate the detonation velocities based on the measured laser-induced air-shock velocities for the explosives which typically exceeded 750 m s^{-1} . Ramaswamy et al. [92] carried out an experimental investigation into the interaction of pulsed laser (a non-Q-switched Nd/glass and Q-switched Nd/YAG) at 1060 nm wavelength and a focused laser spot of 1 mm in diameter, on a single crystal of RDX using high-speed photography. Their research showed that a laser with a long-pulse duration of 300 μs with laser intensity of $\sim 4 \text{ MW cm}^{-2}$ could initiate violent burning of the RDX single crystal. A short laser pulse duration of 8 ns with an intensity of $\sim 12.7 \text{ GW cm}^{-2}$ only induced some damage to the single crystal, including ejection of the material and crack formation without ignition. It was observed that although the reaction started at local hot-spot sites, it never transferred to detonation, even though the hot-spot size ($\sim 20\text{--}30 \mu\text{m}$) was large enough to be ‘critical’ for detonation if there was confinement. When compared to the results reported by Gottfried et al. [88–91], where detonation was observed for powdered RDX by LASEM, a single crystal of RDX requires a higher laser pulse intensity at the NIR laser wavelength of 1060 nm for detonation. Yan et al. [30, 31] also investigated laser ignition of RDX single crystals using a UV 355 nm pulsed laser (6.4 ns pulse duration)

instead of an NIR laser. Their results showed that the probability of ignition of an RDX crystals increased as the laser fluence increased with a threshold of 1.7 J cm^{-2} (i.e., 0.27 GW cm^{-2} of laser intensity) for a 50% success rate of ignition. The laser-induced ablation only caused material to eject on the facets of the crystals, generating pits and cavities. Laser fluence of up to 120 J cm^{-2} (i.e., $\sim 19 \text{ GW cm}^{-2}$ of laser intensity) was applied to the RDX crystals in order to measure the laser-induced damage on the facet surface of the crystal. An increase in laser fluence was observed to increase both area and depth of damage in the crystals. However, from this research, neither detonation nor deflagration of the RDX crystals was reported, although RDX has a high optical absorption at the UV wavelength used in the study, and the applied laser influx was also high. This could suggest that the short laser pulse did not generate adequate hot spots from the well-absorbed optical energy within the RDX crystals to induce deflagration or a strong shockwave to induce shock to detonation. For such energetic crystals of RDX, research conducted by Fang et al. [65] demonstrated successful initiation of GNS-embedded RDX crystals by using a short-pulsed Nd-YAG laser at a laser wavelength of 532 nm and a pulse duration of 6 ns. The modes of ignition and initiation for the observed reaction of the RDX crystals on applied laser pulse, as defined by Fang et al., are shown in Table 1. Here, the minimum required laser flux was reduced to 0.04 J cm^{-2} (i.e., 6.7 MW cm^{-2} of laser intensity) for initiation and 0.01 J cm^{-2} (i.e., 1.7 MW cm^{-2} laser intensity) for ignition. It was observed that as laser flux increases from the ignition threshold to beyond the initiation threshold, the GNS-embedded RDX crystals underwent violent ignition through to full initiation mode with visible flames and explosion, with the crystal samples being fully consumed. Such research using a short-pulsed laser as the initiation optical energy clearly shows that an increase in laser flux or laser intensity increases the success rate of ignition and initiation of the energetic crystals.

As shown from the above research, pulsed laser intensity (i.e., power density) plays an important role in inducing the initiation and ignition of explosives. Compared to short-pulsed lasers for laser ignition of secondary explosives, CW or long-pulsed lasers, particularly diode lasers, have the advantage of being smaller, compact and cheaper, with power density determining ignition performance. As estimated and shown in Harrach’s early research [93], using one-dimensional theory, the laser intensity of around kW cm^{-2} or lower was required for thermal ignition of secondary explosives with a long laser pulse duration, but at a higher laser intensity, the thermal ignition would not occur due to rapid laser ablation and removal of the material. However, six orders of magnitude higher of laser power density (tens of GW cm^{-2}) was required for the detonation initiation of secondary explosives such as PETN and TNT, when using a short, pulsed laser (pulse durations in the hundreds of ns region). This difference in requirements for laser power density between a CW or long-pulse duration laser and short-pulsed laser was attributed to the two identified initiation processes: a thermal initiation process and an impulse or shock initiation process. Using a CW CO_2 laser (10.6 μm wavelength) as the igniting source, Li et al. [94] conducted research in laser ignition of secondary explosive FOX-7 mixed with carbon. Successful ignitions were achieved with laser power densities of $130\text{--}560 \text{ W cm}^{-2}$. An increase in laser power density was observed to decrease the ignition delay time from ~ 30 to $\sim 7 \text{ ms}$. Yu et al. [95] studied the ignition properties of RDX and

TABLE 1 | Modes of interaction and classification of their severity [65].

Modes			
Ignition		Initiation	
Violent	Plasma explosion or violent movement or fast event (<6 ms)	Full	Sample consumed, visible flame, explosion, change in shape
Moderate	Small plasma explosion or slow but significant movement	≥50%	As above, but ≤50% of sample recovered
Weak	Some movement or damage on inspection by microscope	<50%	As above, but >50% of sample recovered
Nil	No interaction observed	Nil	Sample fully recovered

FOX-7 composites with a CO₂ laser at a wavelength of 10.6 μm. The ignition delay time of the composite was found to be less than either RDX or FOX-7. Increasing the laser power density from ~30 to 179 W cm⁻² reduced the laser ignition time of the composite from ~115 to 10 ms. CW CO₂ lasers have been a good choice for many laser ignition applications [96–99] due to hot-spot generation induced by their high power and long-wavelength IR radiation in explosive materials [100]. These applications have obviously demonstrated the dominant role of laser power density to achieve reliable and fast ignition.

Solid-state lasers, including Nd-YAG lasers and diode lasers with CW power output at visible or NIR wavelengths, have been attractive to laser ignition studies and applications for their reliability and system miniaturisation. Research conducted by Hu et al. [101] used an Nd-YAG laser with a long-pulse duration of 10 ms and a wavelength of 1064 nm to investigate laser ignition of HMX. An optical sensitizer Al/CuO NPs was used to significantly increase the sensitivity of the composite of HMX/Al/CuO to laser radiation, and it was shown that the ignition delay time of the composite decreases with increasing laser power. With a laser power of 75 W and a laser beam focus of 1.5 mm in diameter (i.e., the power density of ~4.2 kW cm⁻²), an average pulse duration of 0.23 ms (i.e., the energy density of 0.97 J cm⁻²) was required to cause ignition, whereas at a laser power of 17 W (i.e., the power density of ~1 kW cm⁻²), a longer duration of 5.6 ms (i.e., the energy density of 5.6 J cm⁻²) was required for ignition to occur. Therefore, a higher igniting laser power will generate a faster ignition which requires a lower laser energy for ignition to occur. Diode lasers at NIR wavelength of 808 nm were commonly used for ignition of explosives. Using a CW diode laser of 808 nm wavelength and a laser beam focus of 1.25 mm in diameter, Ehrhardt et al. [102] experimentally studied the ignition of RDX/nitrocellulose (NC) propellants. They showed a threshold laser power for ignition at ~5 W (or 408 W cm⁻²) with an ignition delay of ~450 ms. A shorter ignition delay time of ~100 ms was achieved at a higher laser power of 10.2 W (or 832 W cm⁻²). Research conducted by Tan et al. [103] showed that 3,5-dinitro-4-chlorobenzoic acid and 3,5-dinitro-4-hydroxybenzoic acid could be ignited by a diode laser at 808 nm wavelength through photothermal effects which resulted in deflagration when the compounds contained ions. Zverev et al. [104] demonstrated the successful ignition using CW diode lasers at wavelengths of 450 and 808 nm of PETN mixed with zinc oxide NP doped with copper (PETN-ZnO:Cu) and iron (PETN-ZnO:Fe). In this study, a minimum laser ignition fluence at 450 of 26 nm and 95 J cm⁻²

with a laser radiation duration of 50–60 ms was required for the ignition of PETN-ZnO:Cu and PETN-ZnO:Fe, respectively. Their research showed the ignition of PETN-ZnO:Cu using 808 nm diode laser at low power (11 W). This suggests that photochemical initiation may play a role in the initiation of PETN-ZnO:Cu at 450 nm wavelength. Using a long-pulsed laser at 808 nm wavelength (pulse width of 50 ms), Bai et al. [105] achieved initiation to detonation of confined RDX doped with star-shaped gold-copper alloy nanocrystals with the focused laser beam of 0.2 mm in diameter and a laser power of 41 W and density of 1 W cm⁻². Their research showed a strong photothermal effect and low-energy laser initiation. More experimental studies on laser ignition of explosives using an NIR diode laser have been performed by Fang et al. [41–43, 62–64]. Here, they investigated individual factors which affected the laser power density for ignition in order to achieve low-power diode laser applications. One of these factors was found to be optical sensitizers which were added to explosives. These sensitizers significantly enhanced the optical absorption of explosives and reduced the required laser power and power density for the ignition of explosives. Another factor was found to be the size of the focused laser beam. Here, a smaller laser spot resulted in an increase in the igniting laser power density. In their experimental studies, the effect of the laser spot size was investigated [42]. They showed that decreasing the spot size reduced the ignition delay time and lowered the threshold laser ignition power for a fixed threshold power density. However, it was found that for the ignition of FOX-7, there was an optimum size of laser spot (~1 mm in diameter) to achieve the fastest ignition, and any further decrease in the laser spot size slowed down the ignition. This was attributed to radial heat dissipation from the exothermic reaction zone. It was concluded that the threshold ignition energy density depends inversely on the laser beam radius for laser spots which are smaller than the optimum size. From their research, a typical threshold laser power density for the insensitive explosive FOX-7 doped with CB was around 1.3 kW cm⁻².

5 | Conclusions

This review article has conducted a critical literature survey on research in laser ignition of energetic materials with the emphasis on the effects of essential laser parameters; laser wavelength, optical absorption, optical sensitizer and laser power density. A summary of the research in this review is summarised in Table 2. It is evident from this review that direct laser interaction

TABLE 2 | A summary of the main research reviewed on laser ignition of explosives with applied laser ignition parameters.

Energetic materials	Additives as optical sensitisers	Lasers (wavelength)	Ignition delay time (ms)	Laser power (W)	Laser energy density (J cm^{-2})	Laser power density (kW cm^{-2})	Cited references
RDX	GNSs	Pulsed Nd-YAG laser (532 nm)	1	—	0.01	1.7×10^3	[65]
	GNR	Diode laser (808 nm)	28	1	1.6	0.057	[63]
	Au-Cu alloy nanocrystals	Pulsed laser (808 nm)	28.7	41	3.5 k	130.6	[105]
	NC	Diode laser (808 nm)	450	5	2.04	0.408	[102]
HNS	CB	Diode laser (808 nm)	1.2	45	1.72	1.433	[41]
	Dye		2.1		3		
FOX-7	GNRs		150	8	67.9	0.453	[62]
			11	46	28.7	2.605	
	CB	Diode laser (974 nm)	8	10	10.2	1.27	[42]
			0.07	40	0.36	5.08	
	Graphite	CO ₂ laser (10.6 μm)	7	—	3.93	0.56	[94]
HMX	Graphite		40	—	0.95	0.19	[95]
		Pulsed XeCl laser (308 nm)	—	—	14	0.7×10^6	[25]
	Al/CuO NP	Nd-YAG (1064 nm)	0.23	75	0.97	4.2	[101]
			5.6	17	5.6	1.0	
	GNPs	Diode laser (808 nm)	5.5	3.32	—	—	[70]
PETN	ZnO/Cu NP	Diode laser (450 nm)	—	21	26	0.47	[104]
	ZnO/Fe NP				95	1.73	
	ZnO/Cu NP	Nd-YAG laser (532 nm)		—	0.7	50	
	ZnO/Fe NP						
	Graphite	Pulsed XeCl laser (308 nm)	0.1— 0.3×10^{-3}	—	10	5×10^5	[25]
		Pulsed Nd-YAG laser: (1064 nm)	1.4×10^{-3}	—	9	9×10^5	
C-20	AlNPs				3.6	3.6×10^5	
					4	4×10^5	
		Pulsed Nd-YAG (1064 nm)	—	1.4×10^6	0.063	4.5×10^3	[69]

Abbreviations: CL-20, hexanitrohexaazaisowurtzitane; CB, carbon black; GNRs, gold nanorods; GNSs, gold nanospheres; HNS, hexanitrostilbene; NC, nitrocellulose; PETN, pentaerythritol tetranitrate; RDX, cyclotrimethylenetrinitramine.

with materials has been an attractive topic for laser ignition of energetic materials because this technology is safer and more environmentally friendly compared with conventional ignition methods. The optical absorption capability of energetic materials plays a vital role in their laser ignitability. Most of energetic materials have strong optical absorption in the UV to visible spectral region but weak absorption in NIR region. It is also worth noting that the absorption spectra were mostly measured in dilute solutions of explosives, whereas in solid form other absorption mechanisms take place (e.g., at phonon levels). Optical absorp-

tion additives are necessary and are added to energetic materials in order to increase their optical absorption and achieve reliable ignitions using low-power lasers. In addition to commonly used additives like carbon-based materials, metallic nanoparticles, like gold, silver or AlNPs, are popular optical sensitizers for laser ignition due to their surface plasmon resonance at specific and selected laser wavelengths in NIR region. Laser intensity required for ignition can be significantly reduced and ignition delay time can be shortened when additives are included in energetic materials. It can also be confirmed that laser intensity

or fluence largely affects the ignition performance in terms of ignition time. An increase in laser intensity or fluence significantly reduces the ignition delay time up to a certain level. However, the doping method of additives needs further research, for example, embedding metallic nanoparticles in secondary explosives crystals may cause violent deflagration or detonation of the explosives. Another area of concern is the effect of a pulsed laser with high fluence which may directly initiate prompt detonation of explosive charges [25, 65]. However, a CW laser will only initiate detonation when there is adequate confinement of the explosive [62, 87]. This review has also shown that metallic nanoparticles could be the most promising optical sensitizers for embedding inside energetic crystals. These sensitizers allow ignition to occur at a selected laser wavelength from within the crystal as a confining matrix [63, 65]. Therefore, this review may provide a guideline in consideration of laser parameters for laser ignition and detonation of energetic materials and help to direct further research in this exciting area of laser initiation for secondary explosives.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

No new data were created or analysed in this study. Data sharing is not applicable to this article.

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