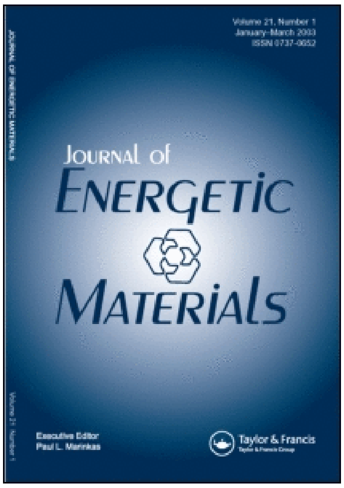

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Laser ignition of explosives: Effects of laser wavelength on the threshold ignition energy

Henric Östmark[a]; Magnus Carlson[a]; Katrin Ekvall[a]
[a] National Defence Research Establishment, Sundbyberg, Sweden

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LASER IGNITION OF EXPLOSIVES: EFFECTS OF LASER WAVELENGTH ON THE THRESHOLD IGNITION ENERGY

Henric Östmark, Magnus Carlson and Katrin Ekvall
National Defence Research Establishment
S-17290 Sundbyberg, Sweden

ABSTRACT

A method, based on a tunable 180 W CW CO_2-laser (9-11 μm), for measuring the effect of laser wavelength, and hence optical absorbance coefficient, on the threshold ignition energy of explosives, has been developed. To determine how the laser beam interacts with the high explosives, a theory for the connection between the ignition energy and the optical absorption coefficient is formulated. The dependence of the threshold ignition energy of RDX on laser wavelength, and hence on the optical absorption coefficients for RDX, was studied at two pressures: 1.1 and 3.1 MPa. In this wavelength interval (9-11 μm), the ignition energy varied between 30 and 330 mJ for a beam radius of 0.9 mm and at a pulse width of ≈ 4ms. At wavelengths with large optical absorption depth (25-60 μm), the ignition energy was the same for both pressures, while at wavelengths with small optical absorption depth (< 15 μm), the ignition energy was much higher at 1.1 MPa than at 3.1 MPa. This behavior is discussed in detail, and a number of sub-processes taking part in the complete ignition process are identified: 1) Lambert-Beer absorption in the solid phase leading to a decomposition of the HE into gaseous products. 2) At low pressure, the gaseous decomposition products can absorb a portion of the laser energy leading to a lowering of the laser energy deposited in the high explosives, and hence a higher ignition energy. 3) At high pressure, the gaseous reaction products are collapsed onto the sample surface. 4) At large optical absorption depths (>25μm), a large volume of explosive is heated, "confining" the decomposition, and thus lowering the ignition energy.

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INTRODUCTION

The use of a laser as ignition source has, since its introduction by Brish *et al.*[1, 2] and Minichelli and Yang[3] in the late sixties and early seventies, been extensively applied to studies of the ignitability of explosives. The first discussion of laser ignition mechanisms is also found in one of the papers by Brish *et al.*[2]. Since then, numerous papers on laser ignition of explosives have been published dealing with the ignition of detonations in high explosives[4-7], the ignition of deflagrations[8-12] and models for laser ignition[13-16]. Different laser ignition methods applied to pyrotechnics[17], propellants[9], and high explosives[18], have shown that the energy necessary for ignition depends strongly on the surrounding gas pressure. The interpretation of the pressure dependence has been that the ignition process is not solely a solid phase reaction but is in fact a complex process where gas, liquid and/or solid phase reactions are involved.

All these studies have shown the importance of gas phase processes for the ignition, but the influence of a finite absorption depth in the explosive, and the fact that the gas phase decomposition products can absorb energy, has not been taken into consideration. In some papers it has been pointed out that by changing the absorption by adding different additives such as carbon, alumina and organic dyes, the ignition energy can be lowered[19, 20]. The suggestion that a Lambert-Beer absorption might be one of the main interaction processes between the HE and the laser beam has been put forward in many papers[2, 6, 14, 21-23]. The IR absorption spectrum for RDX is shown in Figure 1. As can be seen in this figure, RDX (and all other common high explosives[24]) have a very large variation of the optical absorption coefficient with the wavelength in the IR region. The interaction processes between laser beam and explosive need to be better understood for a number of reasons: 1) to be able to make mathematical models of the ignition processes; 2) to be able to calculate the temperature when combining laser ignition with spectroscopic methods for studying chemical reactions occurring in an ignition zone; and 3) for more practical purposes such as the construction of laser ignition systems where it is very important to optimise the interaction between the laser beam and the explosive in order to be able to use the smallest (and cheapest) possible laser.

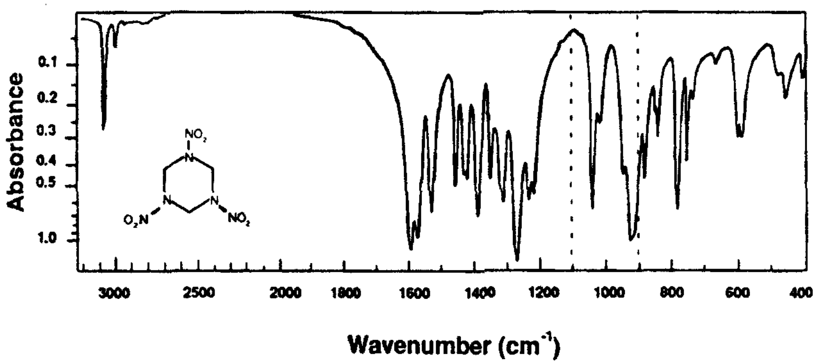

FIGURE 1

IR absorbance spectrum for RDX. The hatched lines enclose the wavelength domain accessible by the CO_2*-laser.*

The laser ignition method developed at the Swedish Defence Research Establishment (FOA) has proved useful in parametric studies of the ignition processes of pyrotechnic compositions[23,25] and high explosives[18], and, in combination with spectroscopical methods, e.g. Raman spectroscopy[26] and mass spectroscopy[27], for more basic studies of the ignition processes at the molecular level. By using a tunable CO_2-laser as ignition source, the method can be extended to study the influence of laser wavelength on the ignitability of a high explosive.

The object of this study has been to determine the influence of laser wavelength on the threshold ignition energy, which, with the absorption coefficients known, can be interpreted as the influence of optical absorption depth on the ignition energy. The study interpreted as the influence of optical absorption depth on the ignition energy. The study was undertaken in order to investigate the interaction between a laser beam and a high explosive and to increase our understanding of the radiative ignition processes. The parameters studied were ignition energy as a function of the laser wavelength (9-11μm) for RDX at two different pressures, 1.1 and 3.1 MPa.

THEORY

As stated above, the radiative (e.g. by laser) ignition of a high explosive is a multi-phase process, but as Atwood and Price[9] pointed out, the ignition process must at least consist of the following consecutive parts: inert heating, pre-ignition reactions, and self-sustained ignition. These parts can be modelled separately, at least as a starting approximation. The model outlined here will only treat the solid phase and we have chosen to ignore the energy contribution from the chemical reaction.

The preferred model is based on the following assumptions: Homogeneous solid sample, Arrhenius kinetics, and thermal conduction as the sole energy transport mechanism. The consumption of the sample due to reaction / decomposition will be neglected since ignition will have occurred long before any substantial mass loss has taken place. The temperature distribution in an explosive, irradiated by a laser, can with these assumptions be described by the heat flow equation with two energy source terms:

$$\frac{1}{\kappa}\frac{\partial T}{\partial t}-\nabla^2 T=\frac{1}{\lambda}(\Lambda_{chem}+\Lambda_{laser}) \qquad (1)$$

The first term on the right hand side, Λ_{chem}, is the heat generation due to the exothermic reaction in the explosive, and the second term, Λ_{laser}, is the volumetric heating caused by the laser. We will use first order Arrhenius kinetics to describe the energy released by the explosives. The laser beam has a Gaussian intensity distribution $I(r)$, which, in the plane of the sample, can be described with the following equation:

$$I(r) = I_0 e^{-r^2/\omega^2} \tag{2}$$

If the absorption of the laser energy follows Lambert-Beer's law, $I/I_0 = e^{-\alpha x}$, the absorbed power in the explosive is given by the following expression:

$$\dot{E}(r,z,t) = I(r,t)\alpha e^{-\alpha z} \tag{3}$$

which, combined with an assumption of first order Arrhenius kinetics for the energy release in the high explosive, gives the following equation:

$$\frac{1}{\kappa}\frac{\partial T}{\partial t} - \nabla^2 T = \frac{\rho Q K_0}{\lambda} e^{-E_a/RT} + I_0(t)\frac{\alpha}{\lambda} e^{r^2/\omega^2} e^{-\alpha z} \tag{4}$$

We are not directly interested in the temperature distribution itself but rather to determine if a given stimulus ($I_0(t)$) leads to an ignition (e.g. is a critical condition to the equation). To do this a mathematical condition for ignition needs to be defined. Equation 5 cannot be solved analytically, but with some simplifications it can be solved for some special cases. To be able to extract some information from the laser ignition experiment we are mainly interested in solutions which in a simple way correlate the ignition energy with the absorption coefficient, and in order to do this some simplifications have to be made. Thus we assume inert heating (no internal chemical energy release), short pulses (no heat conduction), large beam radius, and that the ignition occurs at a specific temperature (T_{ign}), which gives the following expression for the threshold ignition energy[23, 25]:

$$\varepsilon^0_{ign} = I_0 \tau_p = \frac{\rho C_p}{\alpha}(T_{ign} - T_0) \tag{5}$$

The reflectivity will probably also vary with the wavelength, this is not taken into account in these calculations. The relation betwen the measured power, P_{tot}, and I_0 can, if the laser beam has a Gaussian intensity distribution, be written as:

$$I_0 = \frac{P_{abs}}{\pi\omega^2} = \frac{(1-R)P_{tot}}{\pi\omega^2} \tag{6}$$

which combined with Equation 5 gives the following equation:

$$E = P_{tot}\tau_p = \frac{\pi\omega^2\rho C_p}{(1-R)\alpha}(T_{ign} - T_0) \tag{7}$$

Here we have our desired connection between the threshold ignition energy and the absorption coefficient. This equation, although very simplified, is probably useful for a comparison between experiments and model, especially if any major deviation from the predicted outfall could be detected.

When the laser ignition technique is combined with different spectropscopic techniques for basic ignition studies, the interesting parameters are temperature ($T(\tau_p)$), and maximum heating rate (H_{max}), which can be deduced from Equation 7 and are:

$$\begin{aligned} T(\tau_p) &= T_0 + \frac{\alpha I_0 \tau_p}{\rho C_p} = T_0 + \frac{\alpha(1-R)P_{tot}\tau_p}{\rho C_p \pi\omega^2} \\ H_{max} &= \left(\frac{\partial T}{\partial t}\right)_{max} = \frac{\alpha(1-R)P_{tot}}{\rho C_p \pi\omega^2} \end{aligned} \tag{8}$$

Experimental

The main characteristics of the basic laser ignition instrumentation and techniques have been described elsewhere[18, 23, 25], so only a brief summary complemented with the special equipment and technique needed for this study is given here. The experimental set-up for a laser ignition experiment is shown in Figure 2. It consists of the following parts: a 180 W step tunable (9-11μm) continuous wave CO_2-laser (Edinburgh Instruments PL6), a delay/pulse generator (Stanford Research Systems inc. model DG 535) for producing laser pulses with the desired pulse width (1ms-cw), a laser beam analyser (ALL GmbH LBA1/A) for measuring the beam parameters, a power meter (Laser Precision Corp. Rk-5720/RkT-1000-HD), a photo detector (wavelength range 300-700 nm) for measuring the light emitted from the reaction, a digital oscilloscope, an explosives test chamber, and a spectrometer for measuring the wavelength of the laser.

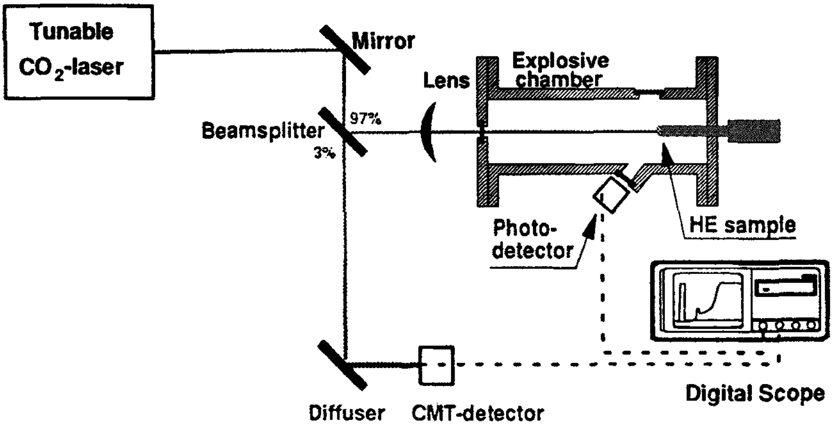

FIGURE 2.

Laser ignition apparatus.

The explosive sample was prepared by pressing about 300 mg of RDX to a pellet 10 mm in diameter, 2.5 mm thick, and its density 90 % of the crystal density. The processing gives a very uniform sample and sample surface with very small sample-to-sample variation in the sense of its absorbance/reflectance/density properties. Of importance is also the fact that the diameter of the particles is much less than the diameter of the laser beam. The pellet was glued on to a sample holder in the explosive chamber. The laser beam was chopped to the desired pulse width by electrically pulsing the laser with the delay/pulse generator, and focused by a zinc selenide lens through a window onto the RDX sample. If ignition did not occur, the sample holder was rotated between each laser pulse so that unreacted explosive was exposed to the laser beam.

The beam intensity distribution was measured with a laser beam analyser and fitted to a Gaussian intensity distribution using the ω and I_0 as parameters. By using Equation 6 and measuring the laser power, P, the laser beam intensity distribution can be obtained in absolute units. The laser power, P, was measured with a calorimetric power meter. The laser pulse width, τ_p, was measured with a CMT detector. The laser ignition, E, was calculated as $\int P(t)dt$.

The measurements were conducted using the Bruceton up-and-down method[29] (30-120 tests) by varying the laser fluence at a fixed pulse width (≈ 4ms). This was done at different wavelengths and at two different pressures, one in the low pressure regime and the other in the high pressure regime. The gas used for prepressurization was air. The results from the up-and-down measurements were analysed with the ML-14 computer code[30] in order to determine the mean, the standard deviation and the 80 % confidence interval of the ignition fluence.

The optical absorption coefficient of RDX was measured with a dual beam infrared (IR) spectrometer (Perkin-Elmer 599B) by varying the amount of explosive under test

(1.5 - 7.0 mg) in a 300 mg pressed pellet of KBr. The absorption coefficient (α) was calculated from the IR-spectra by fitting the acquired data to Lambert-Beer's law, $I/I_0 = e^{-\alpha z}$. The coefficients for the wavelengths which were to be examined by laser ignition were calculated at some different concentrations and then interpolated to 100 % HE after which an average value was calculated.

Results

The optical absorption coefficient was measured for a number of wavelengths in the IR region accessible with the CO_2-laser. The threshold ignition energy was then measured at the same wavelengths at two different pressures, 1.1 and 3.1 MPa (one in each of the two pressure regimes[18]). The values for threshold ignition energy and the corresponding optical absorption coefficents are shown in TABLE 1. The parameters for obtaining this data were as follows: Pulse width 3-5 ms (some variation from the chosen value of 4 ms); the pulse width of each laser shot was measured with the CMT-detector and the energy was calculated as $E = P_{tot} \cdot \tau_p$. The laser beam radius was 0.9 mm, and an up-and-down series of 30-120 shots were conducted at each wavelength. A different number of shots were conducted at each wavelength and pressure, as we chose to shoot a larger number of shots where the standard deviation was large, in order to decrease the confidence interval.

In Figure 3 the threshold ignition energy is plotted vs. laser wavelength. The ignition values are given as 50% probability for ignition and the error bars are the 80% confidence interval. As can be seen in this figure, no simple correlation between the wavelength and the ignition energy exists. An interesting observation can be made, however: at some wavelengths the ignition energies at both pressures are the same, and at other wavelengths they differ with more than a factor ten.

TABLE 1

Threshold ignition energy of RDX vs. laser wavelength, with the corresponding optical absorption coefficient for RDX for these wavelengths. The number in brackets is the standard deviation.

Laser Line	Wavelength (µm)	Wavenumber (cm^{-1})	Absorption (m^{-1})	E_{ign} 10atm (mJ)	E_{ign} 30atm (mJ)
9R16	9.29	1076	18000	149.60 (8)	162.90 (30)
9P14	9.50	1052	36800	98.10 (12)	130.30 (21)
9P20	9.64	1037	84500	161.40 (66)	72.20 (19)
10R36	10.15	986	18900	78.30 (14)	61.80 (25)
10R16	10.27	973	31400	118.40 (44)	120.80 (24)
10P20	10.59	944	87500	246.40 (111)	56.30 (15)
10P36	10.76	929	146000	329.40 (75)	30.70 (6)

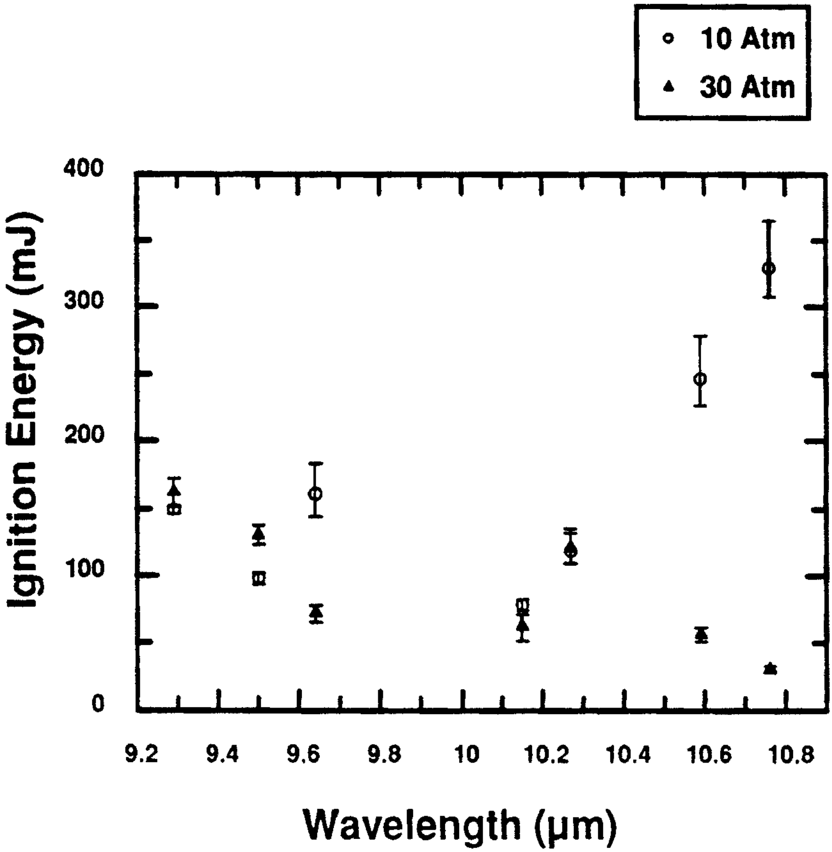

FIGURE 3.

Threshold ignition energy vs. laser wavelength. The error bars are for a 80% confidence interval

Discussion

Earlier, a mechanistic model for laser ignition, assuming a multi-phase ignition, has been proposed[18]. The condensed phase is heated by the laser beam and starts to vaporize or decompose to gaseous products (for RDX e.g. NO_2, NO, H_2CO, N_2O) which diffuse into the gas phase in front of the sample and begin to react. This reaction in turn gives energy feedback to the condensed phase and enhances decomposition. This line of thought is also supported by the Price and Boggs theory[16] and references cited therein. The presence of decomposition products in the gas phase has been shown by combining the laser ignition measurements with mass spectroscopy[27] studies and laser induced fluorescence studies[31] of the pre-ignition / ignition reaction zone. The strongly reduced pressure dependence at high pressures (above 2.5 MPa) is due to the fact that the reaction zone is collapsed onto the sample surface, possibly because the pressure is high enough to condense the decomposition products, and depends only weakly on the pressure[18].

In order to better interprete the data in Table 1 and to test the hypothesis that the main interaction process between RDX and a laser beam is a Lambert-Beer absorption, the data in Table 1 was plotted as ignition energy vs. absorption depth ($1/\alpha$), Figure 4. The question mark in Figure 4 indicates that some interaction process coexists with the Lambert-Beer absorption, which is unknown.

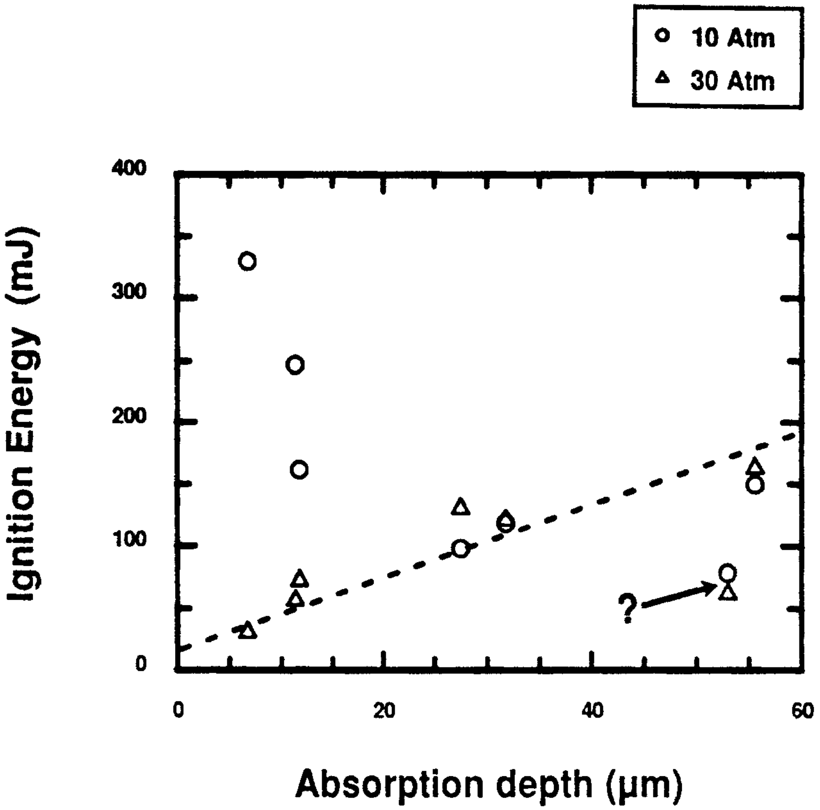

FIGURE 4.

Threshold ignition energy vs. absorption depth of RDX.

This figure, Figure 3 and the data in Table 1 show some distinct properties of the ignition processes which are worth recognizing:

- At small absorption depths (below 15 μm) the ignition energy is much higher at 10 atm than at 30 atm.
- At larger absorption depths (25-60 μm) the ignition energy is the same (within the statistical uncertainty of the measurements) for both pressure regimes.
- Most of the points in Figure 4 (excluding the low pressure points with a small absorption depth and the point marked with a question mark) fall on a straight line.
- The standard deviation for the ignition energy is much larger for the point with small absorption depth and low pressure than for the rest of the points.
- No "simple" correlation between laser wavelength and threshold ignition energy exists.

Based on these observations some conclusions about the radiative ignition processes can be drawn. The higher ignition energies at some laser wavelengths in the low pressure regime are probably caused by some of the gaseous decomposition products absorbing laser energy, thus lowering the laser energy deposited in the high explosives. This has not been considered in earlier work[18] nor in any of the theories for radiative ignition (e.g. Price and Boggs theory[16]). The observation that the ignition energy is the same for both pressures at some of the wavelengths is interesting in that it tells us that we do not have any gas phase interaction here!! (cf. Reference 18). This may depend on either of two reasons: 1) The possible gas phase decomposition products do not have any IR absorption at these specific wavelengths and the flame feedback is neglectable, or at least not enough to compensate for the loss of energy due to absorption. This contradicts, to some extent, the Price and Boggs theory. 2) At larger absorption depths, a larger volume of explosive is heated, and this leads to a "confinement" of decomposition products / reactions starting at depth inside the explosive. Hence we have an equivalent of the collapse reactions in the high pressure regime [see above] and thus we should have the same ignition energy in this case as in the high pressure regime. The second explanation is further supported by differences in ignition delays between the low and high pressure regime observed in earlier work[18], where in the low pressure regime the ignition is preceded by a light pulse due to a vapour phase reaction as a luminous cloud is ejected from the sample before the deflagration starts. In the high pressure regime no signs of a pre-ignition light emission were found and the reaction delay is notably shorter[18].

It is the authors' opinion that the second of these explanations is the most probable, but to reliably distinguish between these two (and other possible) alternatives some more experiments are to be conducted. In particular, it would be interesting to make the same series of experiments with an explosive with totally different IR absorption spectra. It would also be very interesting to conduct these experiments at some more pressures (e.g. 0, 1, 5 and 50 atm).

The possibility of fitting most of the points to a straight line, see Figure 4, leads to the conclusion that one of the main interaction mechanisms between the high explosive and the laser beam might be a Lambert-Beer absorption (cf. Equation 7), at least at these power levels and interaction times. The ignition temperature can be calculated from this line by using Equation 7. (T_{ign}=290°C).

CONCLUSIONS

The results and discussion presented in this paper allow the following conclusions about the ignition process to be drawn:

- Lambert-Beer absorption in the solid phase is probably the main energy interaction mechanism between a laser beam and a solid high explosive. This heating leads to a decomposition of the HE into gaseous decomposition products.

- At low pressure, an absorption of the laser energy in the gaseous decomposition products can take place, leading to a lowering of the laser energy deposited in the high explosives (and hence a higher ignition energy).

- At high pressure, the gaseous reaction products are collapsed onto the sample surface, possibly because the pressure is high enough to condense the decomposition products.

- At large absorption depth (>25μm), a large volume of explosive is heated, and this leads to a "confinement" of decomposition products from reactions starting at depth inside the explosive. We have here an equivalent of the collapse reactions in the high pressure regime [see above] and thus we should have the same ignition energy in this case as in the high pressure regime.

Another interesting conclusion can also be made: it might be possible for RDX (and hence probably for other explosives) to select a laser wavelength where the influence of any gas phase processes of the initial stages of ignition might be negligible. This is of course of great importance for the construction and optimisation of laser ignition systems.

LIST OF SYMBOLS

Unit	Dimension	Description
α	m^{-1}	Optical absorption coefficient
ε_{ign}	J/cm^2	Threshold ignition energy
κ	m^2/s	Thermal diffusivity, $\kappa=\lambda/\rho\cdot C_p$
λ	$W/m\cdot K$	Thermal conductivity
ρ	kg/m^3	Density
τ_p	s	Laser pulse width
ω	m	Laser beam radius
Λ	W/m^3	Heat generation
C_p	$J/mol\cdot K$	Heat capacity
E	J	Energy
E_a	J/mol	Activation energy
I	W/m^2	Power density
I_0	W/m^2	Power density at center of laser beam
K_0	s^{-1}	Frequency factor
P_{tot}	W	Laser power
Q	J/kg	Heat of reaction
R	--	Reflection coefficient
T_0	K	Room temperature
T_{ign}	K	Ignition temperature
r,z	m	Cylinder coordinates

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