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THE INITIATION OF EXPLOSIVE CRYSTALS BY SHOCK OR IMPACT

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Résumé La formation des points chauds est une étape primordiale pour l'initiation par choc ou impact des cristaux explosifs. Par conséquent, toute tentative pour comprendre l'initiation des cristaux explosifs passe par l'étude préalable de la localisation de l'énergie du choc ou de l'impact pour former des points chauds. Plus généralement, il apparait que la localisation de l'énergie a lieu dans tous les solides cristallins, qu'ils soient inertes ou énergétiques, dès qu'ils sont soumis à une déformation plastique induite par choc ou par impact. Cet article est en partie une revue de nos recherches en cours qui visent à comprendre le processus de localisation de l'énergie à un niveau microscopique en examinant la dissipation de l'énergie dans les mouvements des dislocations. La production des dislocations et leur mouvement constituent le mécanisme fondamental de la déformation plastique. L'énergie dissipée dans le champ de phonons engendré par le mouvement des dislocations est déterminée et l'on montre qu'elle est à la fois localisée et intense. Les vitesses de dissipation de l'énergie sont calculées pour des chocs et impacts modérés. Pour des chocs intenses, on montre que le pompage des niveaux d'énergie interne des molécules près du centre de dislocation est possible et qu'il peut produire une dissociation moléculaire rapide. La formation de bandes de cisaillemnt est modélisée. Ces bandes de cisaillement proviennent de l'accumulation de nombreuses dislocations dans de petites régions de cisaillement intense dans le cristal et elles sont considérées comme sites macroscopiques des points chauds.

Abstract The formation of hot spots is an essential step in the process of initiating explosive crystals by shock or impact. Consequently, any attempt to understand the initiation of explosive crystals must first establish how the energy of the shock or impact is localized to form hot spots. More generally, it appears that energy localization occurs in all crystalline solids, both inert and energetic, when they are subjected to shock or impact induced plastic deformation. This paper is in part a review of our current research to understand the process of energy localization on a microscopic level by examining the energy dissipated by moving dislocations. Dislocation generation and motion are the fundamental mechanism of plastic deformation. The energy dissipated in the phonon field generated by moving dislocations is determined and shown to be both localized and intense. Energy dissipation rates are calculated for mild shocks and impacts. For high level shocks it is shown that direct pumping of the internal energy levels of the molecules near the dislocation core is possible which can produce rapid molecular dissociation. The generation of shear bands is modelled. These shear bands are formed by the accumulation of many dislocations in small regions of intense shear in the crystal and are taken to be the macroscopic sites of the hot spots.

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Introduction

There is substantial experimental evidence that the initiation of an explosive crystal by shock or impact is due to plastic deformation of the crystal. Furthermore, this plastic deformation is not a bulk or continuum process but rather occurs in small local regions often referred to as hot spots. The energy localization or concentration that occurs in these local hot spot regions provides the means by which the explosive molecules are excited to dissociation which in turn provides the additional energy to cause reaction to begin to spread throughout the crystal.

Prompted by these observations, that plastic deformation is essential to the initiation of explosive crystals during shock or impact, a theoretical investigation was undertaken to understand the fundamentals of the deformation process. The analysis focused on the microscopic root cause of plastic deformation, the generation and movement of dislocations, and examined the rate at which energy is dissipated by rapidly moving dislocations, how fast these dislocations move under an applied shear stress, and how they might be generated and congregate in shear bands during an applied shock or impact. This present paper is an effort to draw together the above mentioned research and to extend it to predict the response of a nominal explosive crystal to either mild impact or to a more violent shock loading.

Analysis

In crystals, plastic deformation occurs due to the creation and motion of dislocations. At the microscopic level these dislocations represent a substantial distortion of the crystal lattice in the region near the core of the dislocation. Here, a review is given describing the energy localization and dissipation due to the motion of edge dislocations in a crystalline lattice. This summarizes the analyses of references (1, 2, 3) to which the reader is referred for details. These results are then extended to attempt to unify the shock and impact initiation of explosives by establishing that the hot spot - energy localization process is the same for both events. The difference in the response of an explosive to either of these stimuli is just due to the way the different loading rates and stress amplitudes drive the dislocations, and the resultant intensity of energy localization.

The interaction Hamiltonian that describes how a moving dislocation interacts with the lattice can be written as

$$H_I = \frac{1}{2} \vec{\sigma}_0 \cdot \vec{\varepsilon}' \qquad (1)$$

where $\vec{\sigma}_0$ is the local stress field of the dislocation and $\vec{\varepsilon}'$ is the lattice perturbation strain due to the presence of the moving dislocation. For a moving edge dislocation the local stress field can be approximated by its stationary value which in polar coordinates is[4]

$$\sigma_0 = \frac{Gb}{2\pi\,(1 - \upsilon)\,r}\begin{Bmatrix}\sin\theta \\ \cos\theta\end{Bmatrix} \tag{2}$$

where G is the shear modulus, b is the Burgers length, ν is the Poisson ratio, r is the radial distance for the dislocation core, and θ is the polar angle. The local strain field can be expressed in terms of its compressive and shear components as

$$\varepsilon_r' = \frac{x - x_\theta}{d_1}\cos\theta \tag{3a}$$

$$\varepsilon_\theta' = \frac{x - x_0}{d_1}\sin\theta \tag{3b}$$

and

$$\varepsilon_{r\theta}' = \frac{x - x_0}{d_2}\cos\theta \tag{3c}$$

$$\varepsilon_{\theta r}' = \frac{x - x_0}{d_2}\sin\theta \tag{3d}$$

Where d_1 and d_2 are the intermolecular or interatomic spacings along and perpendicular to the slip plane respectively of the moving dislocation. The interaction Hamiltonian can now be written as

$$H_I = \frac{x - x_0}{r}\,\frac{Gb}{4\pi\,(1 - \upsilon)}\left[\sin\theta + \cos\theta\right]\left(\frac{\cos\theta}{d_2} - \frac{\sin\theta}{d_1}\right) \tag{4}$$

It has been observed that dislocations can move relatively long distances of 10μ or more under an applied shear stress pulse.[5] In this spirit it is possible to construct a wave function for a moving dislocation in terms of a sum of plane waves.

$$\psi = \frac{1}{V^{1/2}}\sum_{\vec{k}} a_{\vec{k}}\, e^{i(\vec{k}\,.\,\vec{k} - E_k t/h)} \tag{5}$$

In much the same way the molecular or atomic displacement of the lattice due

to the presence of the dislocation can be written as[6]

$$x - x_o = \frac{1}{(Nm)^{1/2}} \sum_{\vec{q}} \left[\frac{\hbar}{2Cq}\right]^{1/2} \left(b_{\vec{q}} + b^{+}_{-\vec{q}}\right) e^{i\{\vec{q} \cdot (\vec{R} + \vec{r}) - \omega_{\vec{q}} t\}} \qquad (6)$$

where $b^{+}_{-\vec{q}}$ and $b_{\vec{q}}$ create and annihilate lattice phonons of wave vector $\vec{q}$, Nm is the crystal density, C is the local sound speed, $\hbar$ is Plank's constant divided by 2π, and ω_q is the phonon frequency. The coordinates $\vec{R}$ and $\vec{r}$ locate the positions of the dislocation and of the observer.

It is now possible to calculate an interaction matrix element, $\langle \psi_{\vec{k}} | H_I | \psi_{\vec{k}'} \rangle$, describing the processes by which an edge dislocation with momentum $\vec{k}'$ interacts with the lattice to generate or absorb a phonon of wave vector $\vec{q}$ and transition to a new momentum $\vec{k} = \vec{k}' \pm \vec{q}$. Determination of this transition probability is straightforward.[1] Of particular interest here is the probability, $R_{\vec{k}', \vec{k}-\vec{q}}$, that a dislocation with momentum $\vec{k}'$ will interact with the lattice to generate a phonon of momentum $\vec{q}$ and then moves with a reduced momentum $\vec{k} = \vec{k}' - \vec{q}$. This can be written as

$$R_{\vec{k}', \vec{k}' - \vec{q}} = \frac{1}{\hbar} \left(\frac{Gb}{4\pi (1 - \upsilon)}\right)^2 \frac{1}{(2\pi)^3 \, 2 \, Nm} (n_{\vec{q}} + 1) \; \frac{a^{+}_{\vec{k}'} \, a_{\vec{k}'} \, a_{\vec{k}} \, a^{+}_{\vec{k}}}{q}$$

$$\left\{ \frac{1 - 2 \cos\theta \sin\theta}{C_T^2 \, d_2^2} + \frac{1}{C_L^2 \, d_1^2} \left| \left[\sin\theta + \cos\theta - \ln \left| \tan\left(\frac{\pi}{4} + \frac{\theta}{2}\right) \right| \right] \right|^2 \right\}. \qquad (7)$$

The logarithmic singularity that occurs at $\theta = \pi/2$ in the longitudinal or compressive wave term arises from the simplified expression for the stress of an ideal edge dislocation. Recall that an ideal edge dislocation is visualized as an extra half plane of atoms or molecules inserted into the crystal lattice.[7] When an edge dislocation moves through the lattice, the portion of this extra half plane away from the dislocation core also moves and generates a plane compressive wave of coherent longitudinal phonons as it approaches the column of atoms or molecules immediately in front of it. The case when $\theta = \pi/2$ describes this extra half plane component of the edge dislocation. The transition probability is limited to a value of unity so that the prediction of an infinite transition probability just reflects the inadequacy of the idealized rigid model for an edge dislocation when it is applied to the dynamic problem of a moving edge dislocation. The motion of this extra half plane is essentially a bulk process so that we can ignore this portion of the idealized edge dislocation stress, and treat just the region near the dislocation core. This only involves disregarding the logarithmic term and permits an evaluation of the energy

dissipation due to just the core of the moving dislocation.

The energy dissipation rate by the phonons generated in the core region of a moving edge dislocation can be written as

$$\frac{dE}{dt} = \int \sum_{i,\vec{k}} \hbar \vec{q} \, C_i \, R_{\vec{k},\, \vec{k}-\vec{q}} \quad d\vec{q} \tag{8}$$

where the sum over i refers to the three possible phonon polarizations. An examination of the angular dependence of equation (7) combined with the conservation of momentum reveal that most of the emitted phonons originate in the region near the dislocation core and propagate in the direction of the dislocation motion along the active slip plane. This in itself provides a first amount of energy localization. It also allows the vector notation to be dropped on $\vec{q}$ and $\vec{k}$ since these now become scaler quantities. Integrating over the angle θ and extending equation (7) to include the higher order perturbation terms, the energy dissipation rate due to phonons emitted by the moving edge dislocations simplifies to [2]

$$dE/dt = \Gamma R^2 \int \sum_k (n_q + 1) \, N_k \, (N_{k-q} + 1) \, dq + \int \hbar \, \omega \sum_{f,u} \left| \sum_{\ell} \frac{H'_{f\ell} \, H'_{\ell u}}{\omega_\ell - \omega_u} \right|^2 dq \, dV$$

where

$$\Gamma = \frac{2}{(4\pi)^3 \, Nm} \left[\frac{Gb}{1 - \nu} \right]^2 \frac{1}{C^2 d} \tag{9}$$

The phonon frequency distribution generated by a dislocation moving with velocity v can be estimated by observing that if d is the intermolecular spacing distance, then the time for the dislocation to travel that distance is $t = d/v$ and the radial frequency of encounter with the intermolecular or interatomic potential is $\omega = 2\pi v/d$. . For most intermolecular or interatomic potentials this will correspond to the center frequency of the generated phonon spectrum. For the simple Peierls-Nabarro sinusoidal potential this will be the exact frequency of the dislocation generated phonons.

Unfortunately, the dynamical properties of dislocations are not well known. Of particular concern here are the dislocation number densities N_k and N_{k-q}. However, there are two limiting situations of considerable interest which lend themselves to easy evaluation. One of these occurs when $N_{k-q} \ll 1$ and the second occurs at very large dislocation number densities where $N_{k-q} \gg 1$. If it is assumed that after emitting a phonon the moving dislocation essentially stops so that $k-q \simeq 0$, then $N_{k-q \simeq 0}$ can be approximated by its value in a static dislocation pile up behind some barrier[8]

$$N_{k-q\approx 0} = \frac{2(1-\nu)\sigma}{Gb} \frac{x}{[(\ell/2)^2 - x^2]^{1/2}} \tag{10}$$

where ℓ is the length of the dislocation pile up and x is the position of interest in that pile up. For both of these limiting cases the number of dislocations $N_{k-q\approx 0}$ is independent of k and q or is insignificant compared to unity so that in both cases $N_{k-q\approx 0}$ can be taken out from under the summation and integration operations. The summation reduces to

$$\sum_k N_k (N_{k-q\approx 0} + 1) \approx (N_{k-q\approx 0} + 1)\, \overline{N} ,$$

where

$$\overline{N} = \sum_k N_k .$$

The energy dissipation rate per moving dislocation can now be expressed as

$$\frac{E}{\overline{N}} \approx \frac{2\pi\Gamma}{Cd} vR^2 (N_{k-q\approx 0} + 1) + \frac{1}{\overline{N}} \int \hbar\omega \sum_{f,u} \left| \sum_{\ell} \frac{H'_{f\ell} H'_{\ell u}}{\omega_\ell - \omega} \right| dq\, dV. \tag{11}$$

Proceeding further along these lines, it is possible to derive an expression relating the applied shear stress and the dislocation velocity[3]

$$v = v_0 e^{-\frac{\sigma_0}{\sigma}} \tag{12}$$

that resembles an earlier result obtained from experimental data for LiF by Gilman.[9]

While equation (11) contains the energy dissipation rate per moving edge dislocation, considerably more than one moving dislocation is needed to make a hot spot. Bowden and Yoffe [10] estimated that the critical hot spot size required to sustain ignition is approximately 10 to 50 microns. The maximum number of dislocations in a typical organic crystal is about 10^{10} to 10^{11} dislocations/cm^2. This suggests that a minimum of 10^4 to 10^5 dislocations are required to sustain a hot spot ignition.

During shock or impact the generation of dislocations is generally localized in thin narrow shear band regions for most crystalline solids.[5,11,12] These shear bands are regions of intense shear and contain most of the plastic deformation and plastic work preformed on the deforming solid. Consequently they should get very hot. [13,14] Currently there is no adequate theory to account

for the generation and concentration of dislocations in these shear bands although recently one has been attempted.[15] Here it will be assumed that the shear bands are the natural sites for energy localization/hot spot formation in rapidly deforming crystalline solids. It will also be assumed that the shear bands are formed by the action of a few dislocation sources that under high shear stress conditions rapidly generate dislocations which move away from the source and along the shear band with some average velocity v. To a first approximation the energy dissipated in the shear band during deformation is just the product of the energy dissipated per moving dislocation, equation (11), and the number of dislocations in the shear band. In this picture it is worthwhile to examine the energy dissipation rate predicted by equation (11) in three limiting regions. The first region is that of a very small dislocation density, $N_{k-q\approx 0}<<1$, the second is the domain of very high dislocation number density, $N_{k-q\approx 0}>>1$. The third region of interest is that of very high dislocation velocities.

The first limiting region, that of very low dislocation number density, implies that the sample crystal was initially nearly perfect with very few defects or dislocations and perhaps even very few potential dislocation sources. It also implies that to operate in this region the incident shock or impact must be very rapidly rising and shorter in duration than the response time of the dislocation sources. Otherwise long duration shocks or impacts will invariably create dislocations during the plastic deformation of the crystal. Equation (11) indicates that for $N_{k-q\approx 0}<<1$ the energy dissipation rate within the shear band will likely be small.[2] Few significant hot spots will develop, so that the explosive crystals are unlikely to ignite even if the applied shock strength is sufficient to shatter and fracture the crystal.

The second case of interest in which the dislocation number density is large, $N_{k-q\approx 0}>>1$, is the more common situation with crystals that have not been prepared with extreme care. Calculations of the energy dissipation rate for dislocations moving in the inert material LiF compare well with experimental data and give energy dissipation rate per moving dislocation of the order of 1 Joule/s for a .5 Gpa shock.[2,5] Uncertainties in G, ν, and ℓ of RDX allow only an estimate of the energy dissipation for this material. This estimate is only slightly less than that for LiF. What is important is that the energy dissipation occurs immediately with the onset of deformation, is localized to the shear band regions, and can attain high energy density values when 10^5 or more dislocations are involved in the deformation. In these calculations thermal diffusion and thermal softening have been ignored because the typical time duration of the shocks and impacts of interest are less than 50μs which is the approximate time for thermal diffusion to become important.

The third response region of interest in equation (11) is that of very high dislocation velocities, $v \to C$. This corresponds to the situation in which the

dislocations are driven by a strong shock, possibly even a detonation wave. The phonons generated by the rapidly moving dislocations have a radial frequency of $\omega = 2\pi v/d$. It is known that the maximum dislocation velocity can at least approach the local shear wave speed. For $v = 1.8 \times 10^3$ m/s and $d = 10^{-9}$m, as for RDX, then $\omega \approx 10^{13}$ rad/s which is near to the internal mode molecular vibrational frequencies of the RDX molecule. It is possible that even higher frequency optical mode phonons may be generated by dislocations moving with the large amplitude plastic component of the detonation wave. The detonation wave speed in RDX is greater than 8×10^3 m/s and has stress amplitudes in excess of 30 Gpa. At these levels the RDX crystal must be deforming plastically so that it may be possible that the plastic wave dislocation velocity can approach the detonation wave speed. The dislocations within the detonation wave would still move at speeds less than the sound speed in the wave so that causality requirements would be met. However, to the molecules at rest ahead of the wave it would appear that the dislocations in the wave were moving at velocities far in excess in the infinitesmal amplitude sound speed. In any event it can be seen that during a strong shock or detonation it is possible with these high energy phonons, $\omega \geq 10^{13}$ to directly pump and excite the RDX molecule by multiphonon absorption to produce the very rapid molecular dissociation associated with detonation.[16,17] This initial molecular dissociation would be confined to the shear bands - hot spot sites of the shocked crystal.

Summary

Equation (11) describes the energy dissipation rate per moving dislocation over a wide range of plastic deformation conditions. The localized energy concentrations resulting from a large collection of dislocations in the shear bands of a rapidly deforming explosive crystal may provide the means of initiating chemical reaction for a broad range of impact or shock driven deformation. It has long been known for instance from dead pressing experiments and from early impact experiments[18] that deformation is an essential element to shock or impact initiation of explosives. It is also known that explosive crystals with few defects or dislocations are difficult if not impossible to shock initiate even at extremely high shock levels.[19] It has also been observed that the size of the explosive crystals influences their ignition sensitivity.[20] It is well known that the shear stress for deformation and failure of any crystal depends on its particle size. While this relation has not been explicitly derived here, it has been derived elsewhere,[2] and gives the classical Hall-Petch relation.[21, 22]

The prediction that energy can be directly transferred from the shock into the internal molecular modes by multi-phonon excitation provides a very rapid way to achieve molecular excitation in inert crystals and possibly even molecular

dissociation of explosive molecules during detonation. Several recent experiments preformed on the inert materials quartz and lithium niobate show a similar rapid molecular excitation and energy localization, probably in the shear bands, immediately on application of high level shock.[23,24,25] Even during relatively mild impacts energy localization occurs immediately with the onset of deformation and apparent temperatures are reached that far exceed those possible due to bulk processes.[26]

Conclusion

Shock or impact generated hot spots in crystalline explosives involve the same physical process of energy localization. Both require plastic deformation of the sample which occurs by the fundamental processes of the generation and movement of dislocations and the localization of these dislocations in shear bands in the deforming solid. It is likely that the range of response of an explosive crystal from that of a relatively weak reaction due to a mild impact to the extreme reaction of detonation due to strong shock are but different manifestations of the same energy localization- hot spot formation process.

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Comment on Hot Spots - J. LEE

It is generally assumed that chemical reactions in a shock are initiated thermally. True enough, if you send a strong shock wave through the medium, the medium gets thermalized and if the temperature is high enough, you get thermal decomposition. On the other hand reactions can be initiated via non-thermal means, e.q. mechanical shear. This is the field of tribochemistry. In a detonation wave, the question is how nature does it ? Generally speaking, nature does things in the most efficient manner. Thermal initiation is not very efficient, the second law of thermodynamics poses a limitation on the interconversion of thermal to mechanical energy. To get an idea of the order of magnitude of the difference in energy requirement between a thermal and a non-thermal ignition, consider the photoinitiation of a gaseous explosive. If you send a strong pulse of light through a gaseous explosive, free radicals are generated in a photo-dissociation. If the radical concentration is high enough locally, ignition occurs. Deflagration and even detonation can be initiated by this way. If you compute the photo-energy absorbed to achieved ignition, the order of magnitude is in the nanojoule range. However using a thermal source like a spark, the ignition energy is of the order of a millijoule (and then to four orders of magnitude higher for detonation initiation). Thus the difference between thermal and non-thermal initiation is over three orders of magnitude in this example. In solid explosives shear bands can readily be generated. The high mechanical shear rate can lead to the decomposition of the molecules locally. Thus iniation is not thermal. Of course the dissipation of the mechanical energy at these shear bands also thermalizes the medium locally. One can still argue that the initiation is still thermal. However I don't believe so. The evidence of this is that Arrhenius kinetics fail to model these so called "hot spots". In a more

global view, we now know that in gaseous explosive where there is no shear bands, the detonation structure is always cellular due to instability. At cell boundaries (due to incident shock, reflected transverse wave and mach stems coincidence) there are strong shear layers which lead to turbulence via Kelvin-Helmoltz instability. Turbulence mixing then provides a means of autoignition. Cellular structure also persists in homogeneous liquid explosives (e.q. nitromethane). I think non-thermal means of initiation should be given more considerations in studying the mechanism of detonation. I feel that the term "hot spots" is a bit misleading for it and tends to suggest a thermal initiation mechanism instead of a possible mechanical initiation.

Commentaire - J. BOILEAU

Il serait intéressant d'étendre les considérations que vous avez présentées sur les cristaux et leurs dislocations, à des polymères explosifs, et notamment à des "alliages de polymères" par analogie aux "polymères-chocs" tels que le polystyrène dans lequel on a introduit des suspensions très fines d'un autre polymère non miscible.

Questions -J.P. RITCHIE

Is it an oversimplified restatement of your approach to say that impact sensitivity is essentially controlled by material properties, i.e. hard or soft crystals ? What factors lead to less sensitive explosives that can be controlled by the synthesis chemist ? Also, how then does one account for correlations of impact sensitivity with oxygen balance, which is presumably independent of materials properties ?