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SHOCK WAVE INTERACTIONS ARISING FROM NEAR
SURFACE DISPLACEMENT CASCADES

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SHOCK WAVE INTERACTIONS ARISING FROM NEAR
SURFACE DISPLACEMENT CASCADES*

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Abstract

Kaminsky has observed that during 14-MeV neutron irradiation of a variety of materials substantial micrometre-sized particles are ejected from the surfaces of the bombarded specimens. Similarities between Kaminsky's observations and the macroscopic phenomena of spall suggest the possibility that shock waves and transient thermal stresses, generated by energetic primary knock-ons, may be responsible. Spall phenomena are discussed briefly as background to calculations of the shock wave interactions. The results indicate that cascade generated stresses may be responsible for the emission of such particles by triggering the release of stored internal energy.

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1. Introduction

Kaminsky and Das [1] have observed that particles containing as many as 10^{12} atoms are ejected from the surfaces of materials undergoing 14-MeV neutron irradiation. J. Beeler and M. Kaminsky, and separately, J. Robinson [2], suggested that shock waves might be involved. This raised the possibility that the ejections may be related to the macroscopic phenomenon of shock spalling. Here, we consider the extent to which shock waves and transient thermal stresses generated by an energetic neutron collision can account for the phenomenon of chunk emission.

Since quantitative data on chunk emission is available only for niobium, we restrict our discussion to this material. However, the results are readily adapted to other materials. The necessary physical properties of niobium are given in the text.

2. Spall

Shock spalling can be simply described as the separation of material under dynamic tensile loading. Experimentally, this is usually accomplished by the reflection of a plane compressive shock pulse from a parallel free surface or by rapidly heating a thin sheet so that thermal expansion at the surfaces generates the required tensile pulse.

Initial quantitative studies [3,4] were concerned primarily with the tensile stress required for complete or nearly complete spall.

For engineering materials, these stresses are typically tens of kilobars on a microsecond time scale. As techniques improved, it became apparent [5,6] that spall involved the time-dependent nucleation, growth, and eventual linking of defects (voids in ductile materials and microcracks in brittle materials). Observable micrometre-sized damage in the form of microcracks or voids is produced in many materials at threshold stresses of less than 10 kbars and in times of the order of a few hundred nanoseconds. Damage is cumulative: repeated stress pulses increase both the number density and size of observable defects.

3. Collision Cascades

To evaluate the effects arising from 14-MeV neutron collision cascades, we need to know the energy transferred to a lattice atom (primary knock-on atom) in a scattering event, the number of such events per unit volume, and the volume over which the PKA energy is dispersed in the collision cascade.

For 14-MeV neutrons on niobium, the maximum energy transfer is 600 keV, while the mean PKA energy is 180 keV. When electronic losses are accounted for through the Lindhard formalism [7,8], the average damage energy is 65 keV per neutron event [9,10]; however, the median damage energy is still above 100 keV. This situation arises from the fact that in niobium, half of the events result from elastic collisions with an average damage energy of 24 keV. The other half, resulting from nonelastic collisions, have an average damage energy of 106 keV [10].

The total neutron scattering cross section for 14-MeV neutrons in niobium is 4 b. The total fluence to which the niobium samples employed by Kaminsky and Das were subjected was 4.5×10^{15} n/cm². Since the atomic density of niobium is 5.5×10^{22} atoms/cm³, the density of events in these samples was 10^{15} cascades/cm³.

For stress generation, the energy density in a collision cascade is of major importance. Following the primary event, displacements cease in a relatively short time, about 10^{-14} s. Shock waves [11,12] and thermal stresses [12] develop after a few collision times ($\sim 10^{-13}$ s for niobium). It is the energy density at this somewhat later time which will determine stresses in the surrounding media.

While cascade dimensions can be estimated by random cascade theory [13,14] from the projected PKA range, R_p , crystalline effects require some caution in applying these results [8,14] to a description of an individual cascade. To provide a frame of reference, we have calculated R_p from the theory of Lindhard, Scharf, and Schiott (LSS) [15]. Table 1 gives the results as a function of PKA energy, E , as well as damage energies, E_d [7,8], which are the only portion of the PKA energy relevant to our considerations.

At energies below 50 keV in niobium ($\epsilon < 0.1$ in LSS notation), nuclear stopping dominates and R_p is proportional to $E^{2/3}$ [16]. In this range, the results of Sigmund et al. [13,14] predict a cascade volume equivalent to that of a sphere with a diameter equal to R_p . Above

150 keV, R_p is proportional to E and the predicted volume is 60% of a sphere with diameter R_p . On this basis, energy densities E_D/V in electron volts per atom are included in Table 1.

Crystalline effects modify the random cascade theory picture at both high and low energies. Computer simulations of cascades [17] at low energies indicate that rapid energy transport along close-packed directions limits the maximum energy density to a few electron volts per atom. Simulations at energies above 100 keV [18,19] indicate a marked tendency for the formation of isolated subcascades. These have higher individual energy densities than the values shown in Table 1. For our purposes, energy densities between a few electron volts and a few tenths electron volts per atom seem appropriate for the end of the displacement spike. For stress determination, energy densities should be reduced by up to a factor of two for small cascades. This is necessary because atoms at the boundary of the cascade volume share energy with their nearest neighbors in times of the order of 10^{-13} s.

4. Transient Stress Analysis

When energy is instantaneously and locally deposited in a solid, the resulting increases in temperature and pressure give rise to an outgoing shock wave. Displacements resulting from the passage of this shock leave the material outside the heated portion in a state of mechanical equilibrium with the reduced pressure in the heated portion.

The resulting mechanical equilibrium stresses are called transient thermal stresses, since they vanish once thermal equilibrium is reestablished. For cascade dimensions of the order of 100 Å, cooling takes place in about 10^{-10} s.

4.1 Code Calculations

We performed a series of calculations with spherical geometry using a one-dimensional version of the Lagrangian finite-difference code, HEMP [20]. A modified Grüneisen equation of state [21], valid through melt, was employed. The constitutive relations take into account the pressure and temperature dependence of the effective isotropic elastic constants and the yield stress [22]. Since relaxation of the pressure in the cascade region occurs in 10^{-12} s, initially both melting and yield were intentionally suppressed. The calculations indicate that yielding at the theoretical strength and melting in the cascade should begin to influence the results at approximately 1 eV/atom.

To more than cover the range of interest, calculations were made at energy densities of 5.12, 0.64, and 0.08 eV/atom. The energy density determines the magnitude of the stresses. Radial distance, r , is measured in units of cascade radius, a , while time is given in units of a/c , where c is the longitudinal sound speed, 5.05×10^5 cm/s in niobium. Fig. 1 displays the radial and tangential stresses, σ_r and σ_t , as a function of radius at time, $t = 10 a/c$, for an initial energy density of 0.64 eV/atom.

The transient thermal stresses decrease rapidly as they are proportional to r^{-3} , while the amplitude of the outgoing shock decreases with distance as r^{-1} .

Figure 2 gives σ_r as a function of time for all three energy densities at the indicated distances from the cascades. Table 2 summarizes the results and gives initial and final pressures, P_i and P_f , and the stress amplitudes shown in Fig. 2 for each calculation. For comparison, the results of the simple linear elastic analysis (LEA), which follows, are also given.

4.2 Linear Elastic Analysis

The initial pressure is given simply by

$$P_i = \gamma E_D / V, \quad (1)$$

where $\gamma = 1.75$ is Grüneisen's constant. The final pressure, after the shock wave passes, is given by the solution to the quasi-static, thermal stress problem originally solved by Lamé [23]:

$$P_f = \frac{4G/3}{B + 4G/3} P_i, \quad (r < a). \quad (2)$$

Outside the cascade radius, we have

$$\sigma_r = P_f a^3 / r^3 \quad (3)$$

and

$$\sigma_t = -\frac{1}{2} \sigma_r, \quad (r > a), \quad (4)$$

where $G = 377$ kbar is the shear modulus and $B = 1692$ kbar is the bulk modulus. Writing $B + 4G/3 \equiv C_{11} \equiv \rho c^2$, where $\rho = 8.57$ g/cm³ is the density of niobium, we find the change in internal energy of the cascade;

$$\Delta E = \frac{1}{2} (P_i^2 - P_f^2) V / B = \frac{1}{2} P_i^2 V (C_{11} + 4G/3) / C_{11}^2. \quad (5)$$

The remaining energy, $E_D - \Delta E$, still resides in the cascade region, primarily in the form of thermal energy.

Of ΔE , an amount

$$W_T = \frac{1}{2} P_i^2 V (4G/3) / C_{11}^2 \quad (6)$$

appears as stored elastic energy in the transient thermal stresses. The remainder,

$$W_s = \frac{1}{2} P_i^2 V / C_{11}, \quad (7)$$

is carried away by the outgoing shock wave.

Combining eqs. (1 and 5), we find that only a small fraction of the deposited energy, E_D , is available to us, viz.,

$$\frac{\Delta E}{E_D} = \frac{\gamma^2 E_D (C_{11} + 4G/3)}{2C_{11}^2 V} . \quad (8)$$

For $E_D/V = 0.64$ eV/atom, $\Delta E/E_D = 4.9\%$; of this only about 1% is stored in the transient thermal stresses.

The outgoing shock wave cannot be of a particularly simple form, since it produces the displacements that generate the transient thermal stress in its wake. The solution for the shock stress as a function of position and time can be obtained from the integral solution for the displacement given by Love [23] with appropriate initial conditions. In this case, an initial displacement proportional to r , for $r \leq a$, is equivalent to the initial pressure. Some care must be exercised in properly handling the discontinuity at $r = a$. Velocities and stresses can be obtained by taking the appropriate derivatives.

Since the solution is somewhat cumbersome, we only give the magnitude of the peak stresses. The radial momentum carried in the outgoing shock is found by integrating $4\pi r^2 v_p$ over the wave, where v_p is particle velocity.

We have

$$\sigma_r^+ = P_i a/2r,$$

and

$$\sigma_r^- = P_i Ba/2C_{11}r \quad (r \gg a), \quad (9)$$

for the compressive and tensile peaks, respectively. The radial momentum, p_r , is given by

$$p_r = \gamma E_D r / 3ca \quad (r = ct > 3a). \quad (10)$$

A comparison is included in Table 2.

While the above is admittedly simplified, a more detailed linear elastic treatment including thermal diffusion, e.g., by the methods employed by Nowacki [24], does not seem justified. The transient thermal stresses are accurately predicted, and in the range of primary interest, the amplitude of the outgoing shock is predicted to better than 50%.

5. Discussion

5.1 Surface Area

A large chunk, containing 2×10^{12} atoms, in the form of a cylinder with a length-to-diameter ratio of 3:1, has a surface area of $6.4 \times 10^{-7} \text{ cm}^2$ and a volume of $3.5 \times 10^{-11} \text{ cm}^3$. Niobium has a specific surface energy: $\gamma_s = 2.1 \text{ J/m}^2$ [25]. The total energy required is 1700 MeV; on the other hand, 35,000 cascades have occurred in this volume during the experiment. The large number of events involved originally made the prospect of a cumulative damage model attractive since we have a total damage energy deposition of $3.5 \times 10^4 \times (0.065) = 2400 \text{ MeV}$. Of this, only a small

fraction, about 100 MeV, is available for the creation of free surface, primarily in the form of shocks. Thus, at best, energy is available to produce $3 \times 10^{-8} \text{ cm}^2$ of free surface. While this roughly equals the cross-sectional area of a cylindrical chunk, localization would require efficient use of all outgoing shocks.

Unfortunately, the duration of the tensile stress in the outgoing shock is too short ($\sim 10^{-12}$ s) to be effective in either nucleating cracks or lengthening existing flaws. Extrapolation of experience [5,6] with macroscopic spall suggests negligible damage. The best we can hope for is spontaneous nucleation of a penny crack in the near cascade region. The calculations indicate that the combination of developing thermal stress and outgoing shock could produce a crack with a diameter slightly larger than the cascade for $E_D/V > 0.5 \text{ eV/atom}$. While this might occur with some reasonable probability, the resulting damage would be randomly distributed over the volume. The necessary linking could not possibly occur at the present fluence.

It appears that residual internal stresses will be required to provide the bulk of the free-surface energy. Macroscopic residual tensile stresses are a common feature [26,27] in cold-rolled foils. They are generally biaxial, extending through one quarter of the foil, and are an appreciable fraction of the yield strength, i.e., kilobars. While these stresses could provide the energy to extend flaws, they will only become effective once the initial flaw is of the order of micrometres.

Since chunks are also observed, albeit at a lower density, in annealed foils, we are eventually led to microscopic residual stresses as a possible internal energy source. Attractive possibilities are so-called unstressed dislocation pileups [28,29,30]. These can result from the trapped remnant of a previously stressed (double-ended) pileup or the equivalent climb configuration presented by a discontinuous subboundary. Both have been proposed as possible crack nuclei in metals, not requiring plastic flow prior to fracture.

The important feature for our purposes is that the stress fields are nearly proportional to or that they fall off slower than $r^{-1/2}$ at distances somewhat larger than the dislocation spacing. This situation extends to distances of the order of the size of the group, typically micrometres. This feature can be combined with the familiar Griffith criterion for brittle crack growth, which can be written,

$$\sigma^2 L = \text{a constant}, \quad (11)$$

where σ is the tensile stress and L is the crack length.

Thus, if a crack somewhat longer than the dislocation spacing, can be nucleated at the head of the group, it will continue to develop to a length comparable to the group size. This development will occur if the Griffith criterion is initially satisfied. For an unstressed pileup in niobium, 1 μm in length, a dislocation spacing of 250 $\text{\AA}$ (i.e. 40 dislocations)

is required. The initial 200-Å-diameter crack would have to be centered within 100 Å of either end of the pileup. For a discontinuous subboundary, 1 μm in length, a dislocation separation of 50 Å (corresponding to a 3.5 deg tilt) is required. In this case, the initial 200-Å crack needs to be produced within 200 Å of the boundary termination.

The possibility of nucleating such a crack with the aid of the transient stresses surrounding a collision cascade appears likely, in fact, the stress elevations near pileups reduce the required energy density to approximately 0.25 eV/atom. This results in unzipping the entire length of the dislocation, spreading the crack transversely a distance of the order of micrometres, and thus, providing the required surface as the result of a single cascade event. In cold-rolled samples, the superposed macroscopic tensile stresses could play a role in extending such an event a considerable distance toward the surface. This could account for the observation of cylindrical chunks only in cold-rolled samples.

From the discussion above, the volume surrounding the group or subboundary in which a cascade will be effective amounts to approximately $10^{-15} \text{ cm}^3/\mu\text{m}$ of dislocation length. Thus, each such configuration would experience several suitable cascade events. To account for the observed chunk emission in cold-rolled samples, a minimum of 1000 groups or boundaries per square centimetre would be required within approximately 10 μm of the surface. A density of $10^6/\text{cm}^3$ containing a total dislocation density of $10^4 \text{ cm}/\text{cm}^3$ would be adequate. These densities seem reasonable

since the dislocations account for only 1 part in 10 million of the total cold-rolled density, 10^{11} cm/cm³ [31].

In a well-annealed sample with much better surface finish, a density of 50/cm² within 2 to 3 μ m of the surface requires 2×10^5 groups/cm³ with a corresponding dislocation density of 2×10^3 cm/cm³. This is 1 part in 10^5 of the total density, 10^8 cm/cm³ [31]. Again, the numbers are not unreasonable. The fact that such configurations are ever observed in transmission microscopy requires densities of at least this magnitude.

5.2 Momentum Transfer

The mass of a typical large chunk as described above is 3×10^{-10} g. The minimum velocity required to effect a transfer to the silicon collector, 0.28 cm away, was approximately 6 cm/s. This allows a gravity caused drop of 1 cm. Thus, the momentum required is 2×10^{-9} g·cm/s.

Up to one quarter of the radial momentum could be trapped during formation of a chunk. But, even with $E_D = 300$ keV, eq. (10) would only provide a momentum transfer of 1×10^{-9} g·cm/s. A typical event would only provide a few times 10^{-10} g·cm/s. While this is close to effecting a transfer, it allows little leeway. If we assume the chunk is already formed, although we have a large number of possible ejection events, there is no provision for even minimal interference or friction.

It is much more likely that the requisite momentum arises from the propagating crack itself. At terminal velocity, about half the shear velocity, there is not time for the crack to release all the energy

available. As much as one quarter of the energy stored in the configuration could be converted to kinetic energy directed normal to the crack. For a crack covering an area of 10^{-8} cm^2 this could amount to a momentum transfer of approximately $10^{-8} \text{ g}\cdot\text{cm/s}$, which is more than sufficient to effect a transfer.

6. Summary and Conclusions

While substantial transient thermal and shock stresses can be generated by collision cascades in the 10- to 100-keV range, the limited energy available in transient thermal stresses and the short duration of the radiating shock pulse lead to the conclusion that they play a minor role in directly creating chunks.

However, coupled with the internal stress at the end of discontinuous subboundaries or unstressed single-ended dislocation pileups, a plausible mechanism for chunk emission can be envisioned. The collision cascade provides a trigger for the release of internal energy stored in these configurations. Other internal stress sources providing very high local stresses may also be effective. For example, an internal twin lamella will provide a stress configuration very similar to the unstressed pileup, while accommodation bands which develop from twins intersecting the surface resemble a discontinuous subboundary.

If this is indeed the mechanism, chunk emission will not only be sensitive to the near surface microstructure, but also to surface topography

and macroscopic residual stresses; to be effective in creating a chunk, cascade initiated cracks must eventually intersect the surface. Thus, surface roughness and macroscopic tensile stresses will enhance chunk emission, while highly polished surfaces and compressive surface stresses will suppress emission as observed experimentally [1,31,32].

Key Words

Chunk emission

Collision cascades

Neutron sputtering

Shock waves

Spall

Transient thermal stress

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TABLE 1
Random collision cascades in niobium

$E(\text{keV})$	$E_D(\text{keV})$	$R_p(\text{\AA})$	$E_D/V \text{ (eV/atom)}$
10	8	44	3.16
50	35	129	0.55
100	68	228	0.33
150	100	316	0.18
200	128	406	0.11
250	155	499	0.07
500	285	977	0.02

TABLE 2
Comparison of calculations with
a linear elastic analysis

E_D/V (eV/atom)		5.12	0.64	0.08
P_i (kbar)	CODE	800	100	12.5
	LEA	800	100	12.5
P_f (kbar)	CODE	160	22.9	2.85
	LEA	185	23.1	2.88
σ_r^t (kbar)	CODE	18	7.1	2.2
	LEA	36	8.9	2.2
p_r (10^{-12} g·cm/s)	CODE	8.7	5.2	2.7
	LEA	11.2	5.6	2.8

[†]Peak to peak

Figure Captions

Figure 1. Radial (σ_r —) and tangential (σ_t) stresses as a function of radius for a cascade with $E_D/V = 0.64$ eV/atom and $t = 10$ a/c. The maximum stress is 23 kbar. Compressive stresses are taken as positive. The marked increments on the x axis are in units of a, increments on the y axis are 2 kbar/division. The outgoing shock is at 10 cascade radii.

Figure 2. Radial stress (σ_r) as a function of time for three energy densities. (a) 5.12 eV/atom, $r = 20$ a, (b) 0.64 eV/atom, $r = 10$ a, and (c) 0.08 eV/atom, $r = 5$ a. The peak-to-peak stress and radial momentum in each case are given in Table 2.

Guinan - Fig. 1

Guinan Fig. 2