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- 2375 Application of the Lifshitz-Slyozov theory to precipitation of phosphorus in silicon-germanium thermoelectric alloys
E.L. Burgess, R.D. Nasby
- 2382 Effect of neodymium on dislocation velocity in CaF_2
R. Nathan Katz, R.L. Coble
- 2389 Dielectric measurements using alternating currents and ionic thermocurrents
M.M. Perlman, S. Unger
- 2394 Change of electrical resistivity of metal films bombarded with helium ions of low energy
A. Nomura, M. Kanayama, S. Kiyono
- 2396 Ultrasonic attenuation in cesium halides
S.K. Kor, U.S. Tandon, P.K. Mishra
- 2398 Electric field dependence of the dielectric constant of PZT ferroelectric ceramics
N. Bar-Chaim, M. Brunstein, J. Grünberg, A. Seidman
- 2406 Critical misfit for generation of dislocations at second-phase particles
K. Jagannadham, E.G. Ramachandran
- 2409 Ultrasonic measurements of Young's modulus and extensional wave attenuation in refractory metal wires at elevated temperatures with application to ultrasonic thermometry
E.P. Papadakis, K.A. Fowler, L.C. Lynnworth, A. Robertson, E.D. Zysk
- 2421 Electrical contact activation: A phenomenological model
Rudolf Schubert
- 2428 Thermal conductivity of magnesium plumbide
J.J. Martin, H.R. Shanks
- 2432 Transient electric field and space-charge behavior for unipolar ion conduction
Markus Zahn, Cheung Fung Tsang, Shing-Chong Pao
- 2441 Effect of antiphase boundary energy on mutual cross slip of unlike dislocations
M.J. Marcinkowski, K. Sadananda
- 2447 Dielectric properties of amorphous Se-Bi and Se-Bi-As films
I.S. Goldstein
- 2452 Microwave properties of liquids and solids using a resonant microwave cavity as a probe
K.H. Hong, J.A. Roberts
- 2457 Ion and electron parameters measured with cooled plane electrostatic probes in a cesium plasma
C. Michel, M. Kornmann, J.C. Braun
- 2463 Plasma-induced field emission and the characteristics of high-current relativistic electron flow
R.K. Parker, Richard E. Anderson, Charles V. Duncan
- 2480 Electron impact cross sections and rates for one- and two-electron aluminum ions
E. Oran, J. Davis
- 2484 Comment on approximations involving the transport equation for ions in drift tubes
T.V. Vorburger, S.B. Woo, J.H. Whealton
- 2486 An asymptotic approximation for reaction rates in ion drift tubes
Maurice V. Barnhill III
- 2489 Beam-plasma waves from a finite-length plasma
C.W. Roberson, J. Fukai
- 2493 Radiation into anisotropic plasma from flanged parallel-plate waveguide
Yoshihisa Ogawa, Kohei Hongo
- 2497 Measuring rotationally symmetric potential profiles with an electron-beam probe
W.M. Black, James W. Robinson
- 2502 Theory of potential-well formation in an electrostatic confinement device
W.M. Black, E.H. Klevans

(Continued)

Boeing Ex. 1009, p. 4 of 15
Boeing v. Regents Univ. Cal.

- | | | |
|------|--|---|
| 2512 | Microwave propagation in magnetized ferrite-dielectric composite transmission lines | You-Sun Wu, F.J. Rosenbaum |
| 2521 | Oblique incidence of an electromagnetic wave at a moving ferrite medium | P.K. Mukherjee, S.P. Talwar |
| 2530 | Hypervelocity impact heating of porous aluminum | Donald E. Rehfuss |
| 2536 | Gas migration in vitreous B_2O_3 | J.E. Shelby, R.C. Wayne |
| 2540 | Interferometric measurement of shock-induced internal particle velocity and spatial variations of particle velocity | J.R. Asay, L.M. Barker |
| 2547 | Simple nonequilibrium model for shock-heated monatomic gas irradiated by an intense laser beam | Howard D. Cohen, F.Y. Su, A.A. Boni |
| 2551 | Energy dissipation in a thin polymer film by electron beam scattering | R.J. Hawryluk, Andrew M. Hawryluk, Henry I. Smith |
| 2567 | The rise of small bubbles in water | Carl Bachhuber, Cecil Sanford |
| 2570 | Semiconductor conductivity measurements using a high-sensitivity microwave technique | S. Dixon Jr., R.F. Giordano, H. Jacobs |
| 2579 | Zero-bias anomaly in GaAs/Mo tunnel junctions | B.R. Spod, Y.L. Pak |
| 2582 | Measurement of diffusion length in solar cells | J.H. Reynolds, A. Meulenbergh Jr |
| 2593 | Discussion of the surface-potential fluctuations caused by oxide-charge fluctuations | G. Declerck, R. Van Overstraeten, G. Broux |
| 2596 | Monolithic solid-state traveling-wave amplifier | Avraham Gover, Amnon Yariv |
| 2601 | Stress waves generated in thin metallic films by a Q-switched ruby laser | L.C. Yang |
| 2609 | Electron-beam spreading and its effect on sustainer current and field distribution in CO_2 lasers | J.H. Jacob, J.P. Reilly, E.R. Pugh |
| 2614 | Observation of compressed picosecond pulses of high repetition rate from a Nd-glass laser | K.H. Drexhage, K.B. Eisenthal |
| 2616 | Effect of surface regions on the electrical conductivity of extrinsic semiconductor films | D.W. Covington, D.C. Ray |
| 2621 | Gain-narrowing studies with an organic dye laser | Gerd Marowsky |
| 2624 | Two-dimensional current density distribution within three-terminal semiconductor devices | Shih-Pei Hu, Benjamin M. Rabinovici |
| 2631 | Elastoresistance of <i>n</i> -type silicon on sapphire | Jaroslav Hyncek |
| 2636 | On the efficiency of coupling light from stripe-geometry GaAs lasers into multimode optical fibers | C.A. Brackett |
| 2638 | Double injection in insulators: Theoretical description and analysis of some double-injection experiments in anthracene crystals | H.P. Schwob, D.F. Williams |
| 2650 | Concentration dependence of the refractive index for <i>n</i> - and <i>p</i> -type GaAs between 1.2 and 1.8 eV | D.D. Sell, H.C. Casey Jr., K.W. Wecht |
| 2658 | Optical properties of gold acceptor and donor levels in silicon | S. Braun, H.G. Grimmeiss |
| 2666 | Electronic collision: Induced transition between the upper levels of a He-Cd laser | Takeki Sakurai |
| 2668 | Extrinsic absorption in 10.6- μ m-laser-window materials due to molecular-ion impurities | C.J. Duthler |

(Continued)

- 2672 Dynamic properties of high-mobility garnet films in the presence of in-plane magnetic fields
 2678 Josephson frequency conversion in the range 145–1600 GHz with Nb-Nb superconducting point contacts used as oscillator-mixer
 2689 Crystal growth and recovery of ferromagnetism during thermal cycle in MnBi films
 2693 Losses in laminar composite Nb₃Sn at 60 Hz
 2700 Optical absorption in ion-bombarded magnetic garnet films
 2705 Theory of Bloch-line and Bloch-wall motion
 2716 Study of magnetic structures induced by bending in (100) Fe-Si single crystals
 2724 Effects of low-temperature fast-neutron irradiation on ac losses in NbTi
 2728 Saturation magnetization of gallium-substituted yttrium iron garnet
 2733 Influence of the crystalline electric field on the heat capacity and resistivity of PrAl₃
 2739 A comparison of calculated images generated by six modes of transmission electron microscopy
 2748 Wave conversion in stratified media with hyperbolic-tangent nonuniformities
 2759 Undoped high-resistivity cadmium telluride for nuclear radiation detectors
- COMMUNICATIONS
- 2766 Refractive index of GaAs_{0.62}P_{0.38} between 1.2 and 2.0 eV
 2768 An equation for fracture energy determination by double-cantilever cleavage
 2770 Magnesium fluoride metal atom oxidation laser from explosively generated metal vapor
 2773 Superconductive tunneling in junctions containing lead-gold layered films
 2775 Temperature dependence of minority-carrier lifetime in low-resistivity GaAs
 2777 Change of surface composition of SiO₂ layers during sputtering
 2780 Resistivity of Permalloy thin films
 2782 Electron distribution in a gridded Penning discharge
 2785 Transverse-junction-stripe-geometry double-heterostructure lasers with very low threshold current
 2787 Diffusion effects in one-carrier space-charge-limited currents with trapping
 2789 Thermomagnetic writing on thin-film MnBi using an electron beam
 2792 Determination of hafnium-p-type silicon Schottky barrier
- Roger W. Shaw, J.W. Moody,
 R.M. Sandfort
 G. Vernet, R. Adde
 Shigeo Honda, Tetsuzo Kusuda
 D.P. Snowden, C.H. Meyer Jr.,
 S.A. Sterling
 J.A. Seman, S.H. Wemple,
 J.C. North
 J.C. Slonczewski
 M. Labruñe, M. Kleman
 B.S. Brown, H.C. Freyhardt,
 T.H. Blewitt
 P. Hansen, P. Röschmann,
 W. Tolksdorf
 J.V. Mahoney, W.E. Wallace,
 R.S. Craig
 A. Engel, J.W. Wiggins,
 D.C. Woodruff
 J.L. Doane
 R. Triboulet, Y. Marfaing,
 A. Cornet, P. Siffert
 H.C. Casey Jr.
 D.L. Hindman, J.T. Jones,
 W.R. Determan
 Jon G. De Koker,
 Walter W. Rice
 S. Basavaiah, S.K. Lahiri
 D.I. Kladis, P.C. Euthymiou
 A. Turos, W.F. van der Weg,
 D. Sigurd, J.W. Mayer
 A.F. Mayadas, J.F. Janak,
 A. Gangulee
 Dominick Conte, Gordon Kent
 H. Namizaki, H. Kan, M. Ishii,
 A. Ito
 A. Rosental, A. Sapor
 R.C.F. Heyworth,
 G.R. Hoffman
 M. Beguwala, C.R. Crowell

(Continued)

- | | | |
|------|---|---|
| 2795 | Uniqueness of time-independent electromagnetic fields | Per W. Karlsson |
| 2796 | Grüneisen constants of some group V and VI elements | M.D. Aggarwal,
J.K.D. Verma |
| 2797 | Point-cathode electron sources—Electron optics of the initial diode region: Errata and addendum | J.C. Wiesner, T.E. Everhart |
| 2799 | Doppler interpretation of the frequency shifts of light diffracted by sound waves | Virendra N. Mahajan,
Jack D. Gaskill |
| 2801 | Lateral spread of ion-implanted impurities in silicon | E. Pan, F.F. Fang |
| 2804 | The effects of ionizing radiation on the hardening of $\text{KBr}_x\text{Cl}_{1-x}$ | J.R. Hopkins, J.J. Martin,
J. Larkin |
| 2806 | Hall effect in silicon on sapphire | J. Hyneczek |
| 2808 | Test of a semiempirical equation describing laser beam divergence | Harry E. Bates, Robert W.
Phillips, Thurman Sasser |

ERRATA

- | | | |
|------|---|------------------------------|
| 2811 | Erratum: Complex rigidity of dispersions of spherical shell structures with material compressibility [J. Appl. Phys. 44, 4023 (1973)] | Yo Takano,
Akio Sakanishi |
| 2811 | Erratum: Magnetic resonance spectrometer measurements of atomic hydrogen surface recombination [J. Appl. Phys. 44, 4776 (1973)] | A. Mandl, A. Salop |
| 2811 | Erratum: Theory of optical waveguide distributed laser with nonuniform gain and coupling [J. Appl. Phys. 45, 209 (1974)] | R. Shubert |

ANNOUNCEMENTS

- | | |
|------|---|
| 2812 | Third national conference on crystal growth |
| 2813 | CUMULATIVE AUTHOR INDEX |

(Continued)

Stress waves generated in thin metallic films by a Q-switched ruby laser*

L. C. Yang

Jet Propulsion Laboratory, Pasadena, California 91103
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Vacuum-deposited thin films of B, C, Mg, Al, Si, Ti, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ge, Zr, Mo, Ag, In, Sn, Sb, Pt, Au, Pb, and Bi in several thicknesses, confined between glass substrates, were irradiated by a uniform Q-switched ruby laser beam. The laser pulse was 15 ns in duration, with a maximum energy of 5 J in an area 6.35 mm in diameter. The stress waves were studied using the piezoelectric response of X-cut quartz-crystal disks of the same diameter. The peak stresses are presented as a function of the energy fluence, with possible interpretations of the phenomenon.

INTRODUCTION

The generation of stress waves in thin metallic films by a Q-switched laser is of interest here mainly in terms of its applicability to the detonation of insensitive high explosives. The detonation of such explosives can be initiated directly by a shock wave generated by Q-switched laser irradiation of a thin metal film confined between a glass window and the explosive.^{1,2} The relevant experimental observable of such a confined configuration is the magnitude of the stress. X-cut quartz crystal is a valuable sensor for high-amplitude short-duration stress waves.³ Limited experimental work^{1,4-7} using this technique has shown that stress of the order of tens of kilobars can be generated in transparent substrate-confined films using a Q-switched laser at moderate energy levels. It is the purpose of this paper to report the results of detailed experimental studies of the peak stress as a function of the film thickness, film material, and laser fluence ϕ .

DESIGN OF THE EXPERIMENT

Film thickness

The choice of film thickness was based on two considerations: heat conduction and light absorption. Previous studies^{8,9} on laser interaction with bare metal targets have shown that the depth of the damage or the interaction corresponds closely to the depth of thermal penetration $l \approx (\kappa t_L)^{1/2}$, where κ is the thermal diffusivity of the metal and t_L is the laser pulse duration. Therefore, using nominal values of κ and t_L for a Q-switched laser, $l \approx 10^{-4}$ to 10^{-3} cm.

Limited data on the optical properties of metals and semiconductors as well as the formulas for the physical optics of a substrate/thin film/substrate system are available in the literature.¹⁰ At the ruby laser wavelength (6943 Å), the absorption and transmission behaviors of the system are similar to those of the bulk film materials, except in the case of semiconductors such as carbon, germanium, and silicon, where interference effects are present.

It was decided to examine both thin and thick films. The thinner films selected were about 10 times the optical skin depth at room temperature to absorb the laser beam efficiently, since there is evidence that the absorption coefficient decreases at high temperatures [see Fig. 5(a)]. The thicker films were about three to four times thicker than the thinner ones in order to show the influence of heat conduction.

The films from reagent-grade materials were vacuum deposited on borosilicate glass disks (windows) 7.62 mm in diameter and 5.2 mm in thickness by means of tungsten filament, boat, and electron-beam vaporization techniques. The film thicknesses were monitored and controlled by a 5-MHz AT-cut quartz-crystal microbalance with an accuracy of $\pm 3\%$. For some materials, excessive film stress prevented the growth of the thicker films, so that only thinner films of these materials were prepared for the study.

X-cut quartz crystal

Synthetic X-cut quartz crystals, 1.27 ± 0.075 mm thick and 6.35 ± 0.025 mm in diameter, were used for the experiment. With this rather large diameter-to-thickness ratio, a guard-ring electrode was not necessary,³ which considerably simplified the design of the crystal holder. The crystals had a flatness of a 0.75 wave (5500 Å), less than a 0.25-min deviation from parallelism, a 0.3- μ m surface finish, and 8- μ m-thick chrome plate-based gold electrodes. The relationship between the piezoelectric current I and the stress σ is linear³ for $\sigma < 25$ kbar:

$$I = (\sigma - \sigma_0) F \left(\frac{1}{4} \pi D^2 \right) V_q / h, \quad (1)$$

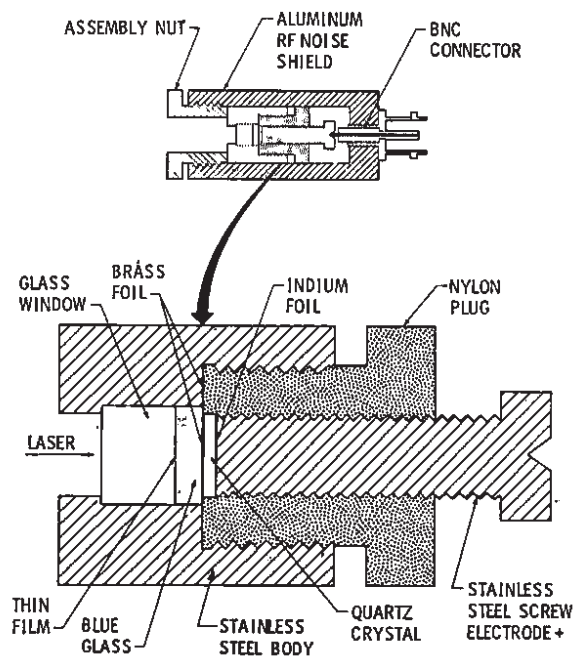


FIG. 1. Scale drawing of experimental fixture.

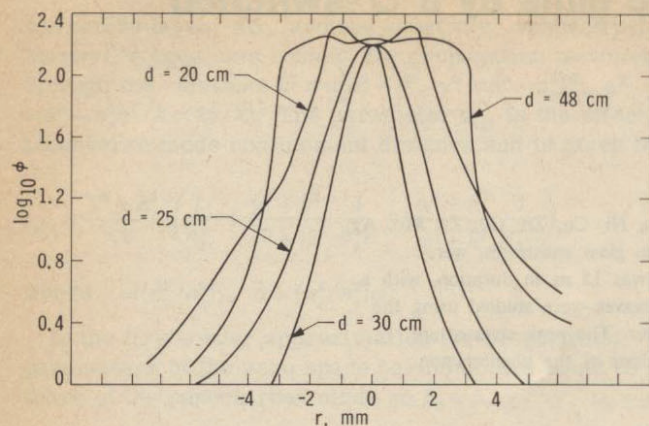


FIG. 2. Relative laser fluence ϕ as a function of radial position r and distance d from 30-cm focal-length convergent lens.

where D and h are the diameter and the thickness of the crystal, respectively; $V_q = 5730$ m/s, the speed of sound along the x axis of the quartz; F is the piezoelectric coefficient³ $= 2.05 \times 10^{-8}$ C cm⁻² kbar⁻¹ for $D/h = 5$; and σ_0 is the stress at the electrode opposing that of σ .

Experimental fixture

Figure 1 shows the design of the easy-to-assemble experimental fixture. The blue glass disk, 1.9 mm thick and 7.62 mm in diameter, was made of polished cobalt-blue glass (density $= 2.5$ g/cm³, speed of sound $V \approx 5500$ m/s) commonly used for welding view shields. It served both as a heat insulator between the film and the brass foil and as a light absorber to prevent possible laser residue from interaction with the foil. Because of the small absorption coefficient (~ 20 cm⁻¹) of the blue glass at laser wavelength, the stresses generated in it were insignificant (< 0.7 kbar) even under direct laser irradiation ($\phi \approx 14$ J/cm²).

The function of the 250- μ m-thick 5-mm-diam indium

disk was to allow uniform pressure to be applied during assembly so that hermetic contacts were achieved at the window/film/blue glass and the blue glass/brass foil/quartz crystal interfaces. Application of hand torque was sufficient to assure a zero-order interference fit across the entire area of the film interface.

The 25- μ m-thick 1.26-cm-diam brass foil disk was held in contact with the steel body by the nylon plug to complete the ground lead of the crystal. The disk is rigid enough to withstand the assembly procedure yet sufficiently deformable to form hermetic contact with the blue glass disk and the crystal. The acoustic transmission at this interface has been calculated¹¹ to be greater than 98%. The aluminum shielding fixture was necessary to eliminate the interference from the electrical noises generated by the Pockel cell. The piezoelectric signal was transmitted via a 2.5-m-long RG/58-C cable to a 50- Ω load at the input of an oscilloscope.

Laser system

A Korad K-1Q Pockel cell laser system consisting of a 5-kV 500- μ F energy bank, an eight-turn helical xenon flashlamp (7 $\times$ 9-mm quartz envelope), a temperature-controlled cooling system, and a 1.43 $\times$ 7.62-cm ruby crystal was used for the experiment. The system was capable of producing a maximum prelasing-freed Q-switched output of 5 J.

The energy output was monitored by a system including a transparent diffuser, attenuators, a spike filter, and an S-1 vacuum photodiode, located behind the 100% cavity reflector. It was calibrated with independent measurements made by a liquid-cell-type laser calorimeter. The laser beam was imaged by a double-convex lens (diameter $\times$ focal length $= 5 \times 30$ cm) into a 6.35-mm-diam spot onto the thin film 48.0 cm away from the lens, while the experimental fixture was rigidly mounted on a fully adjustable alignment fixture. The target feedback effect was found to be negligible. Tests both in vacuum and in air demonstrated that the expo-

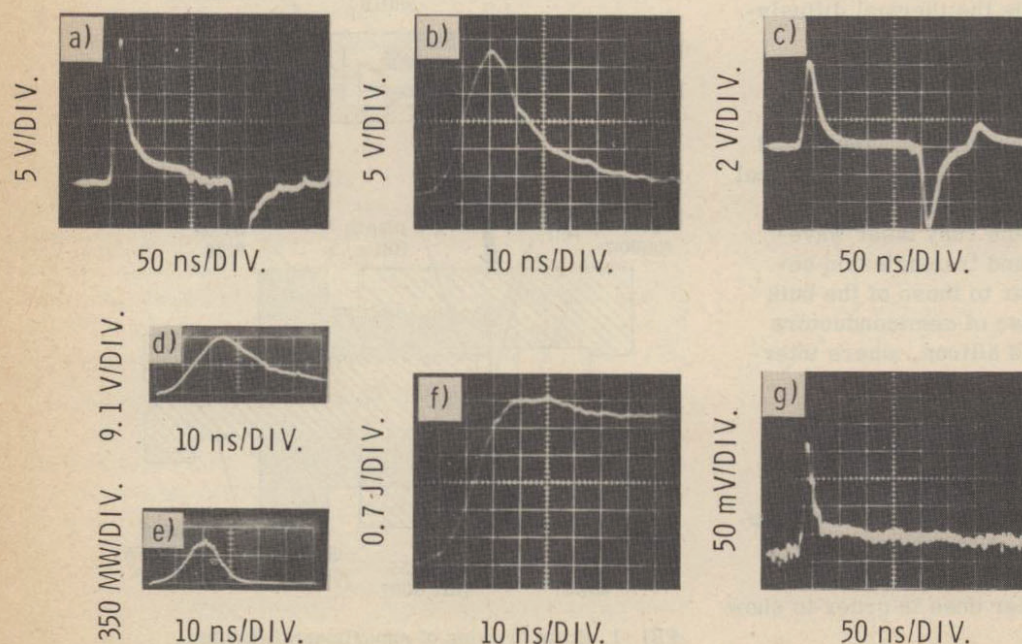


FIG. 3. (a) Typical high-amplitude stress signal, as seen by a Tektronix 7704 oscilloscope. (b) Same signal with better time resolution. (c) Typical low-amplitude stress signal. (d) Typical high-amplitude stress signal as seen by a Tektronix 519 oscilloscope after attenuation. (e) Laser pulse power trace. (f) Laser pulse energy trace. (g) Intensity of visible radiation emitted by an irradiated film.

sure of the focal point in air did not result in any noticeable laser attenuation; therefore, the experiments were performed in air.

The beam profile was studied at a few locations along the beam axis by Eastman-Kodak high-speed infrared film.¹² The results are shown in Fig. 2. It is evident that the local laser fluence ϕ is uniform within $\pm 7\%$ at the thin-film position. Both this profile and the pulse duration were found to be insensitive to the laser output as controlled by the initial energy-bank voltage, except near the threshold of lasing. Low laser fluences ($\phi < 3 \text{ J/cm}^2$) were obtained by attenuation of the beam ($\phi \approx 12 \text{ J/cm}^2$) via a cupric sulfate solution cell.

Oscilloscopes

Only two oscilloscopes were suitable for measuring the short-duration stress signal: Tektronix models 519 and 7704. Model 519 has a rise time of 0.3 ns and a fixed voltage sensitivity of about 10 V/cm. Model 7704 has a rise time of 2.4 ns and a variable voltage sensitivity ranging from 5 V/cm to 5 mV/cm. The latter has the higher voltage resolution and was selected for the experiment.

EXPERIMENTAL RESULTS

Figure 3 shows some typical characteristics of the stress signal and the laser pulse. Comparison of Fig. 3(b) to 3(d) verifies that the response time of Tektronix 7704 is adequate. In Figs. 3(a) and 3(c), the signal in the first wave transit time ($h/V_s = 221.6 \text{ ns}$) records the true stress history. The negative peaks are due mainly to the change of the sign of the factor $(\sigma - \sigma_0)$ in Eq. (1) rather than being a rarefaction or reflection. In comparing Figs. 3(b) and 3(f), it can be seen that the stress appears to correspond to the laser energy, but its duration up to the peak is slightly shorter than the latter (by about 5 ns). The laser energy trace in Fig. 3(f) exhibits a slow tailoff due to the signal decay characteristic of the photodiode. Figure 3(g) shows a record of the visible radiation emitted by a confined 1000-Å-thick aluminum film under laser irradiation of $\phi \approx 12 \text{ J/cm}^2$. It was obtained by the same S-1 photodiode via a 4-mm $\times$ 15-cm plastic fiber optics bundle inserted in place of the steel screw, and after the removal of the crystal and the brass foil in the crystal holder. It is evident that the light pulse had approximately the same shape as the stress pulse.

Figure 4 presents the main experimental results. The ordinate of each graph is the peak stress $\sigma_{\max}$, converted from the peak voltage of the stress signal according to Eq. (1). The abscissa is the laser fluence ϕ , calculated from the laser energy after deduction of the Fresnel reflections at the lens (8%) and at the front surface of the glass window (4%), then divided by the disk area (6.35 mm in diameter). The highest value of $\sigma_{\max}$ of about 21 kbar was well within the linear regime of the crystal response.³ The peak stress decreases very rapidly when $\phi < 1 \text{ J/cm}^2$, characterizing the transition from a pressure stress to a thermal stress¹³; however, this latter regime was not studied in detail.

Postexperiment examination showed that if $\sigma_{\max}$ exceeded 3 kbar, the window/blue glass/crystal assembly

was damaged. At $3 < \sigma_{\max} < 7 \text{ kbar}$, these components were crack damaged, and the film interface showed fusion and bonding effects. When $\sigma_{\max} > 7 \text{ kbar}$, they were completely crazed into fine particles, preventing further examination. The damage was apparently produced when the stress waves reached the external boundary of these confining substrates, where there were large mechanical discontinuities. Therefore, the damage in the glass window always appeared more severe than in the rest of the components. Symmetrical damage was observed under identical mechanical conditions, i.e., when the film was sandwiched between two identical glass windows and clamped in the same way.

The negative polarity of the crystal was always chosen as the ground electrode, so that a positive stress current (voltage) was observed. However, the reversal of this polarity did not result in abnormality of the pulse shape as reported in Ref. 14. Applying bonding compound (Eastman 910) to the film/blue glass interface did not appear to change the experimental results. Gaps at this interface created by deliberately loose assembling caused a large decrease in the observed $\sigma_{\max}$. At thicknesses below 3 mm, the attenuation of the blue glass disk had no noticeable effect on the measured results.

The condition of a high explosive in contact with the thin film was simulated by replacing either the glass window or the blue glass with a Lucite disk. The $\sigma_{\max}$ at $\phi \approx 12 \text{ J/cm}^2$ with a 1000-Å-thick aluminum film was about 14 kbar in both cases. (Acoustical impedance mismatch and the resulting reflection must be considered in the latter case.)

Irradiation by a defocused beam (Fig. 2, $d = 20 \text{ cm}$), in which $\phi (\approx 14 \text{ J/cm}^2)$ varies by approximately a factor of 4 over the film surface, yielded an "average peak stress" $\langle \sigma_{\max} \rangle$ equal to that of a homogenous beam with the same laser energy. This seems to imply that the linear relationship between $\sigma_{\max}$ and ϕ holds for ϕ up to four times larger than 14 J/cm^2 . The decrease in $\langle \sigma_{\max} \rangle$ upon further sharpening of the beam profile (Fig. 2, $d = 25$ and 30 cm) was probably due to the loss of one-dimensionality of the stress wave propagation.

DISCUSSION

In general, Fig. 4 shows that (i) in most cases, $\sigma_{\max}$ varies linearly with ϕ ; (ii) different film materials do not yield greatly different results; and (iii) the dependence on the film thickness is much smaller than would be predicted by heat transfer calculation. Aluminum was studied for eight different film thicknesses. It is apparent that a thickness of 250–500 Å or so is sufficient to generate stress efficiently. This feature should also be qualitatively true for other film materials.

The total acoustical energy per unit area, E_s , associated with the stress waves in both confining substrates (glasses) up to $\sigma_{\max}$ can be approximately estimated as follows. Since

$$\sigma \approx \sigma_{\max} t / T_s,$$

where t is the time and $T_s \approx 2.0 \times 10^{-8} \text{ s}$ is the duration of σ up to $\sigma_{\max}$,

$$E_s \approx 2 \int_0^t \frac{\sigma^2}{Z} dt$$

$$E_s \approx \frac{2\sigma_{\max}^2 t^3}{3Z T_s^2}, \quad (2)$$

where Z is the acoustical impedance of the confining substrates. For $t = T_s$, $\phi \approx 12 \text{ J/cm}^2$ and $Z \approx 1.3 \times 10^6 \text{ g/cm}^2 \text{ s}$; $\sigma_{\max} \approx 20 \text{ kbar}$. Therefore, $E_s \approx 0.4 \text{ J/cm}^2$.

The internal energy per unit area E_i of the plasma produced by the film can be estimated by assuming an ideal gas law:

$$E_i = 2R \frac{3}{2} \sigma, \quad (3)$$

where $2R$ is the thickness of the plasma. Since

$$R = \int_0^t \frac{\sigma}{Z} dt$$

$$R \approx \frac{\sigma_{\max} t^2}{2Z T_s}, \quad (4)$$

therefore, for $t = T_s$, $\phi \approx 12 \text{ J/cm}^2$ and $\sigma_{\max} \approx 20 \text{ kbar}$; $2R \approx 3.1 \times 10^{-4} \text{ cm}$ and $E_i \approx 0.93 \text{ J/cm}^2$. The sum $E_i + E_s \approx 1.3 \text{ J/cm}^2$ is about 10% of the laser fluence of $\phi \approx 12 \text{ J/cm}^2$. This discrepancy is, of course, even more severe at lower $\sigma_{\max}$ and corresponding ϕ , because $E_i + E_s$ is proportional to $\sigma_{\max}^2$.

The transmission and reflection factors ($T\%$ and $R\%$) of confined thin aluminum films were measured and are shown in Figs. 5(a) and 5(b). Since the transmitted and reflected beams were found to be collimated, these measurements were performed by common MgO block-photodiode techniques, with calibrations and deduction

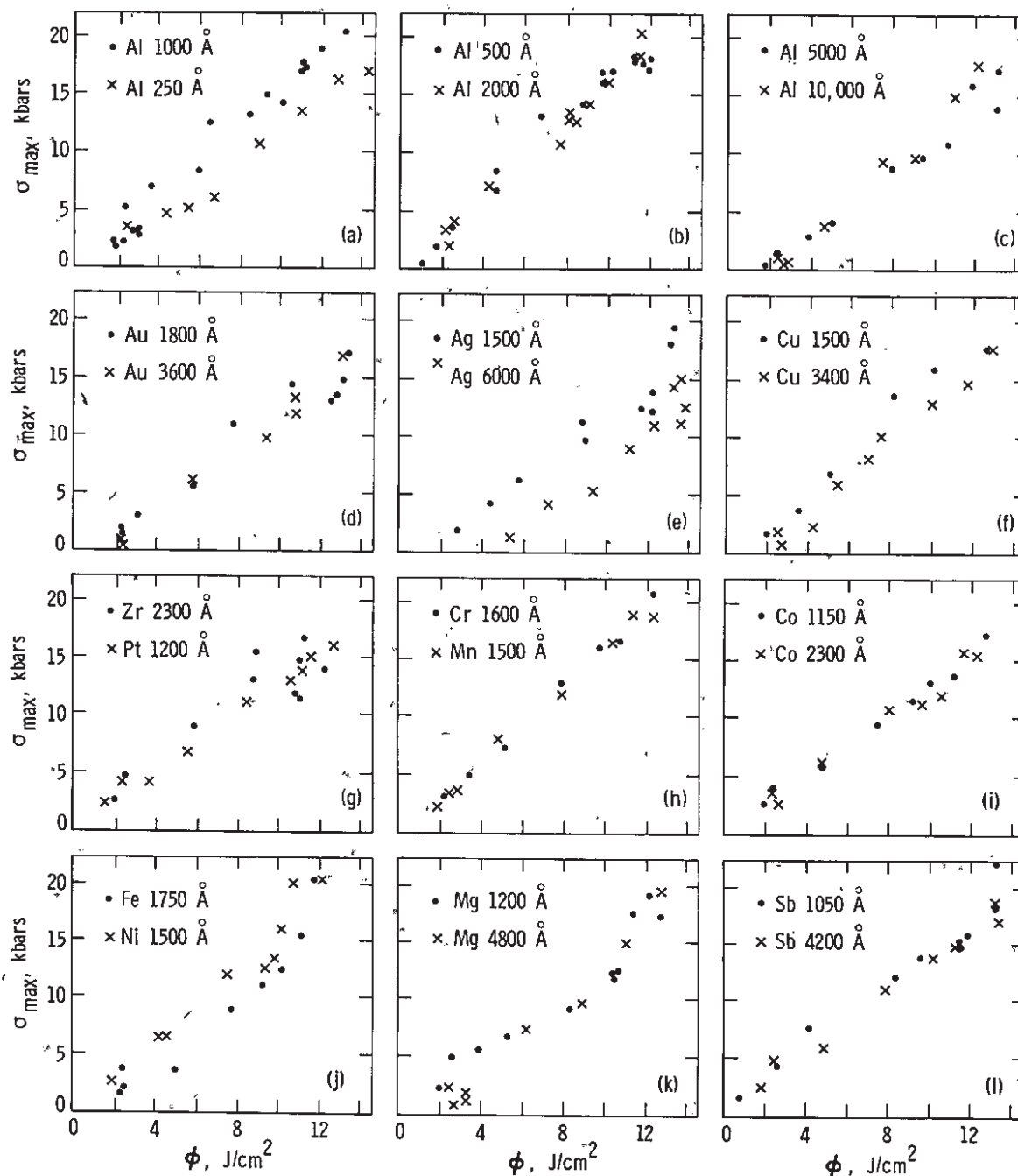


FIG. 4. Peak stress $\sigma_{\max}$ as a function of film material, film thickness, and laser fluence ϕ .

of Fresnel reflections at the free surfaces of the confining glass substrates. It is evident that for a 250-Å-thick aluminum film, the laser absorption is more than 90% in the high- ϕ regime. The typical shape of a transmitted pulse through a 250-Å-thick aluminum film at $\phi = 6$ and 12 J/cm² is shown in Figs. 5(c) and 5(d), respectively. For purposes of comparison, some data on unconfined films (marked U) are also shown in the graph. The relationship $T\% \propto \phi^{-1}$ appears to be a general feature in all cases.

In the early stage (i.e., from 0 to 5 ns), the phenomenon occurring at the film window interface, including the laser absorption, heat diffusion, and propagation of melting, is quite easy to understand in terms of the

usual heat transfer estimations. After the melting, the electrical conductivity, and thus the light absorption coefficient, is much smaller in molten glass than it is in a liquid metal. Therefore, the laser-energy absorption remains in the metallic part of the interface and heats the system above the normal boiling point (BP). According to Grosse's assessments,^{15,16} the critical temperatures T_c of metals are about three to four times higher than the BP, and the critical densities ρ_c are about four times less than those at the BP. It is felt that the same situation should be true for ionic solids, such as glass.

Because of the high viscosity and low kinetic velocity of the liquid, this anomalous volume expansion should

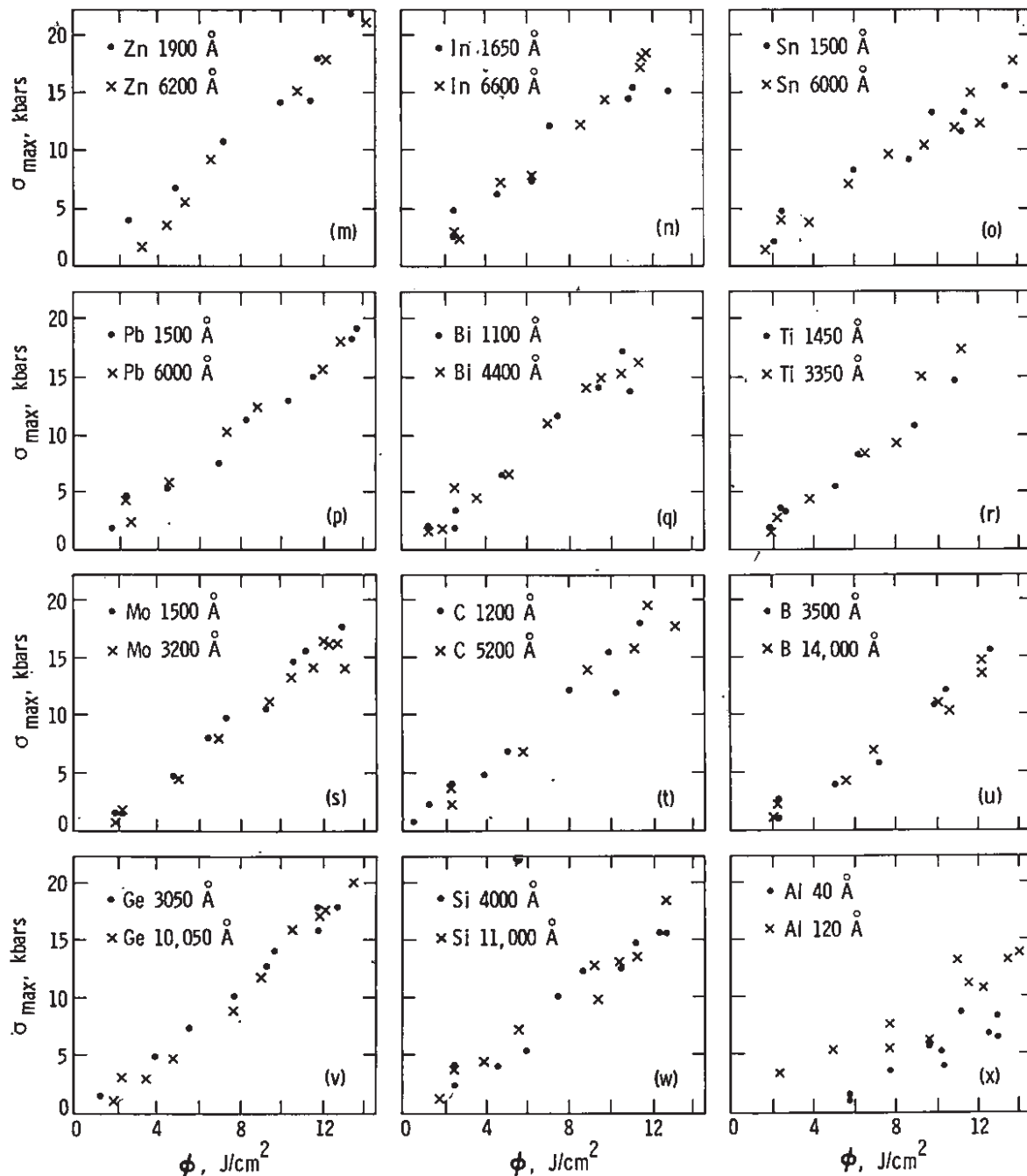


FIG. 4. Continued

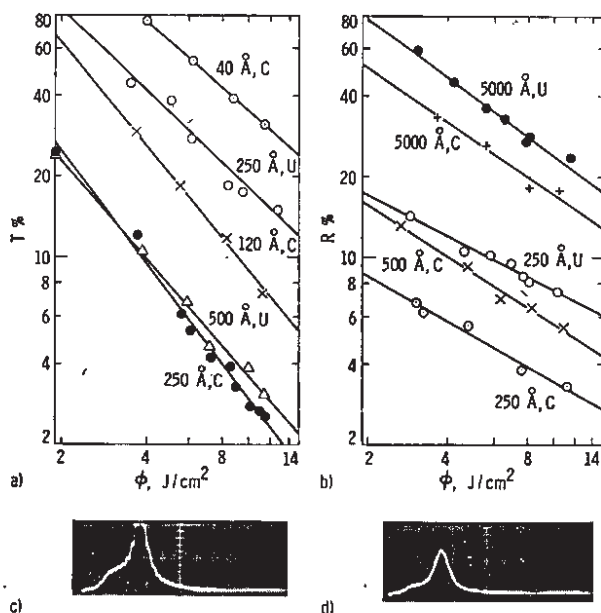


FIG. 5. Time-averaged laser transmission factor (a) and time-averaged laser reflection factor (b) of aluminum films of different thicknesses (U = unconfined condition with films facing laser on glass slides, C = films under confinement by two glass substrates), with records of laser pulses after transmitting through a confined 250-Å-thick aluminum film at (c) $\phi = 6 \text{ J/cm}^2$ and (d) $\phi = 12 \text{ J/cm}^2$ (relative vertical units and 10 ns/div).

be one dimensional along the direction of film thickness. The magnitude of a "thermal stress" so generated can be roughly estimated by the relation $\sigma = Z dR/dt$ to be much larger than the corresponding vapor pressures.¹⁷ Therefore, the system will be superheated to the critical temperature(s) in a liquid state. The apparent shift of the onset of stress waves in both high-thermal-conductance thick films and very thin aluminum films can be attributed to inefficient heating due to the large conduction loss or poor laser absorption.

In the critical regime, rapid decreases in the electrical and thermal conductivities and the laser absorption coefficient are expected. The subsequent process is presumably similar to the gas breakdown under intense Q-switched laser irradiation.¹⁸ Two facts encourage such an interpretation: (i) in this type of breakdown, the threshold decreases with the gas density and (ii) the initial breakdown should be much easier in the critical regime, since the degree of ionization, $\bar{m}$, is already of the order of 10^{-3} . The narrowing of the transmitted laser pulses in Figs. 5(c) and 5(d), as compared to the original pulse in Fig. 3(e), is quite similar to that observed in laser-induced gas breakdown.¹⁹ The knees at about 5 ns from the start of these pulses could be an indication of such a breakdown.

The breakdown under laser irradiation will rapidly heat up the gaseous phase. The model of the phenomenon is such that a high-temperature highly light absorptive plasma is confined by the solid substrates buffered by a thin layer of superheated liquid. Further growth is governed by the ablation—a combination of heat conduction and electronic bombardment. Very limited data are available on ablation of solid targets by a high-energy-

discharge plasma gun.²⁰ However, the drastic differences in experimental conditions make them inapplicable to the present problem. The breakdown of the gas at the rear plasma boundary could be a self-regulated mechanism.²¹ As can be seen in Fig. 3, after the termination of the laser pulse, the internal energy of the plasma is sufficient to radiate more stress energy than during the irradiation.

The foregoing discussion implies that a highly light absorptive thin film of the order of several hundred angstroms thick is sufficient to serve as a "seed" for stress generation. After the creation of a plasma, the subsequent event is only slightly material dependent, since properties such as thermal diffusivity, ionization potential, and electron penetration depth are not very different for materials at high temperatures. The amount of the gaseous phase (total mass or number of atoms per unit area N) is time dependent and is not accurately known. A reasonable guess in terms of an equivalent solid thickness L is $L \approx 2lp_c/\rho \approx (1-2) \times 10^3 \text{ Å}$, where ρ is the solid density, l is estimated by the diffusivity at T_c , $\kappa \approx 0.05-0.5 \text{ cm}^2/\text{s}$.²² The number of atoms per unit volume n is then equal to $N/2R$. However, using this information and standard formulas²³⁻²⁵ for free-free absorption yielded too large a laser absorption coefficient as compared to that shown in Fig. 5. This presumably reflects the inapplicability of these formulas to such a high-density plasma.

The following three energy-loss mechanisms were considered to be insignificant. (i) The presence of a stress wave in the glass window should not affect the laser transmission, since in the solid, ionization will not occur and the change of refraction index under stress is small for $\sigma < 30 \text{ kbar}$.²⁶ (ii) The radiation is essentially blackbody in character, because with such a high-plasma density, the calculated Rosseland radiation mean free path²³ is much shorter than the plasma thickness. During the laser pulse, the total radiation is not significant as compared to the absorbed laser energy until $T > 4 \times 10^4 \text{ K}$ or mean radiation wavelength $< 1000 \text{ Å}$. The ultraviolet radiation will be readily absorbed by the liquid boundaries in a skin depth²⁷ of the order of 10^3 Å and will contribute to the ablation; i.e., energy is trapped in the plasma rather than appearing to be lost. (iii) Since the plasma expansion rate $dR/dt \approx 10^4 \text{ cm/s}$ is small, the consumption of energy by kinetic expansion is small in contrast to a freely expanding laser-induced plasma.^{25,28}

Two significant processes occur. (i) To illustrate the effect of ionization, let us assume that $L \approx 1600 \text{ Å}$ of aluminum, $\sigma_{\text{max}} \approx 20 \text{ kbar}$, $\phi \approx 12 \text{ J/cm}^2$, and $2R \approx 3.1 \times 10^{-4} \text{ cm}$. Then $N \approx 10^{18} \text{ cm}^{-2}$, $n \approx 3.3 \times 10^{21} \text{ cm}^{-3}$, and the energy per atom $\epsilon \approx 70 \text{ eV}$. Using Raizer's approximation for the Saha equation,²³ one finds $T \approx 10^5 \text{ K}$, $\bar{m} \approx 2.65$, the total ionization energy per atom $Q(\bar{m}) \approx 23 \text{ eV}$, and the total kinetic energy per atom $\epsilon_k = \frac{3}{2} k_B T (1 + \bar{m}) \approx 47 \text{ eV}$. Therefore, the ionization energy is important but cannot totally account for the energy imbalance. This is, of course, even more true in the low- σ regime. (ii) Even though there is a large acoustical impedance mismatch at the interface between the liquid phase and the confining solid, the reflective attenuation

should be small because of the small thickness of the former. On the other hand, in an ablation process, the transfer of kinetic energy and momentum between the gaseous and liquid phases is inelastic; therefore, large stress wave attenuation could occur. In other words, Eq. (3) is invalid in this case. In any event, the system is probably too dense to be considered an ideal gas.

Further work that should be done to gain a better understanding of the phenomenon might include the following:

- (i) Measurement of the electrical conductivity of the confined plasma.
- (ii) Experiments on very thin films for all materials similar to aluminum.
- (iii) Measurement of the stress generated by a confined film, including precisely controlled air gaps (e.g., vacuum deposition of a ring-shaped dielectric film around the interaction interface).
- (iv) Extension of the experiment to higher laser fluence.
- (v) Studies of the Q-switched laser-induced breakdown of solids in the critical regime, which can be created by exploding confined films either electrically or by the irradiation of a pulsed free-running laser.
- (vi) Studies of the effect of laser pulse duration. In every respect, a picosecond laser pulse should be more efficient in stress generation than a normal Q-switched laser pulse, as has been indicated in Ref. 5. However, oscilloscopes with picosecond rise times are currently unavailable.
- (vii) Experiments using a pulsed CO₂ laser and an ultraviolet laser, even though the phenomenon is not expected to be as strongly dependent on the laser wavelength as in an unconfined irradiation study.

Finally, it should be noted that the phenomenon offers a unique condition, simultaneously high in temperature, pressure, and shock velocity, favorable to the detonation initiation of insensitive explosives compared to other techniques such as exploding wire and flyer foil. Therefore, a small initiation time transient and excellent simultaneity for multiple initiations can be achieved.¹ Within the experimental range of ϕ , $\sigma_{\max}$ has already reached the shock sensitivity of PETN, RDX, and Tetryl.¹ Using a focused beam, direct initiation can now be achieved with energy of the order of 0.5 J, and use of a compact Q-switched laser with such an output has recently become feasible²⁹ in laser-initiated explosive systems.³⁰

CONCLUSIONS

Stress waves generated by Q-switched ruby laser irradiated thin films under confinement have been studied for a wide range of film materials and several film thicknesses. It was found that the dependence on these parameters was much weaker than would be predicted by heat transfer estimations commonly used to describe the interaction of laser irradiation with uncon-

fined bulk-solid surfaces. For example, an aluminum film 250–500 Å thick was highly efficient in generating stress. In general, the peak stress is linearly dependent on the laser fluence. The energy of the stress wave was considerably smaller than the energy absorbed by the film. Quantitative interpretations were hindered by the rather high plasma density and the lack of an accurate knowledge of the total plasma mass. A reasonable qualitative picture of the phenomenon based on the critical properties of solids, laser-induced breakdown of gases, thermal ionization, and ablation is suggested. The mechanism is favorable for practical applications such as the direct initiation of insensitive explosives or as a tool for generating high-amplitude short-duration stress waves.

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