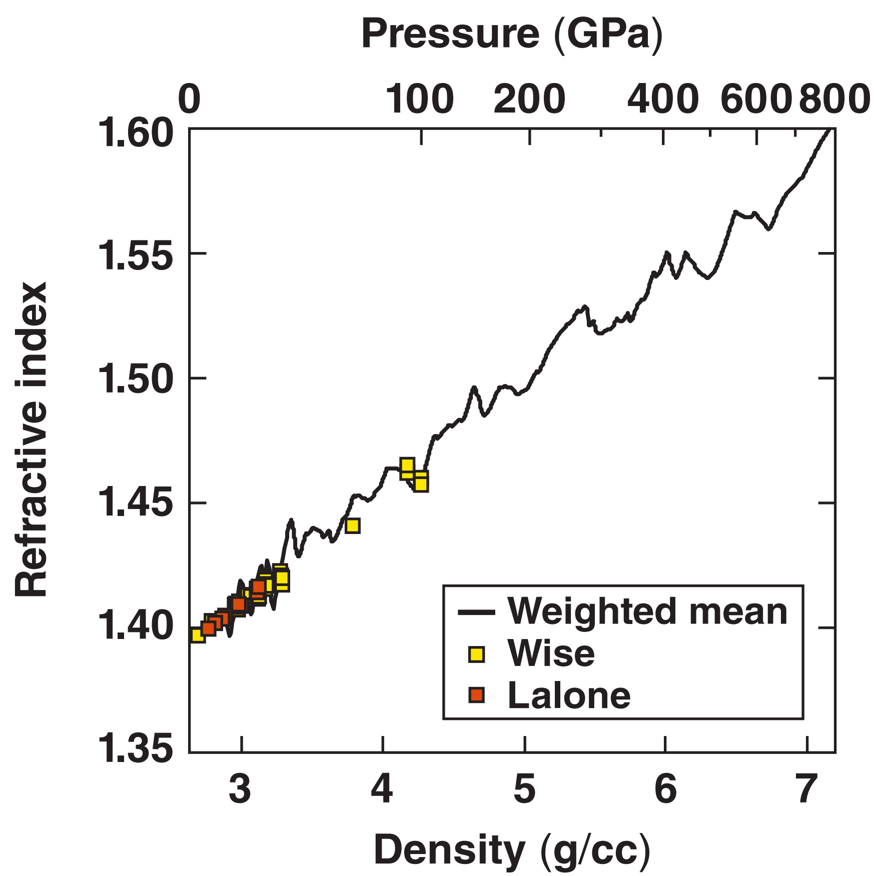
Refractive-Index Measurements of LiF Ramp Compressed to 800 GPa



D. E. Fratanduono
Lawrence Livermore
National Laboratory

53rd Annual Meeting of the
American Physical Society
Division of Plasma Physics
Salt Lake City, UT
14–18 November 2011

Summary

The refractive index of ramp-compressed LiF depends linearly on density up to 800 GPa

- Knowledge of LiF's compressed index of refraction is important for high-pressure equation-of-state (EOS) measurements
- The refractive index of shock-compressed LiF has previously been measured to 115 GPa*
- Ramp-compressed LiF is measured up to 800 GPa
  - LiF is observed to remain transparent over this range
- A single-oscillator model suggests that the band gap will close at pressures above 4200 GPa
  - these are the highest ever refractive-index measurements of an insulator

E19145b

*J. L. Wise and L. C. Chhabildas, Sandia National Laboratories, Report SAND-85-0310C, NTIS (1985).

Outline

I. Motivation

II. Theory/technique

III. Experiments
   A. Method of characteristics
   B. VISAR measurements
   C. Analysis

IV. Results
   A. Refractive index
   B. Effective oscillator mode
   C. Metallization

Collaborators

T. R. Boehly and D. D. Meyerhofer

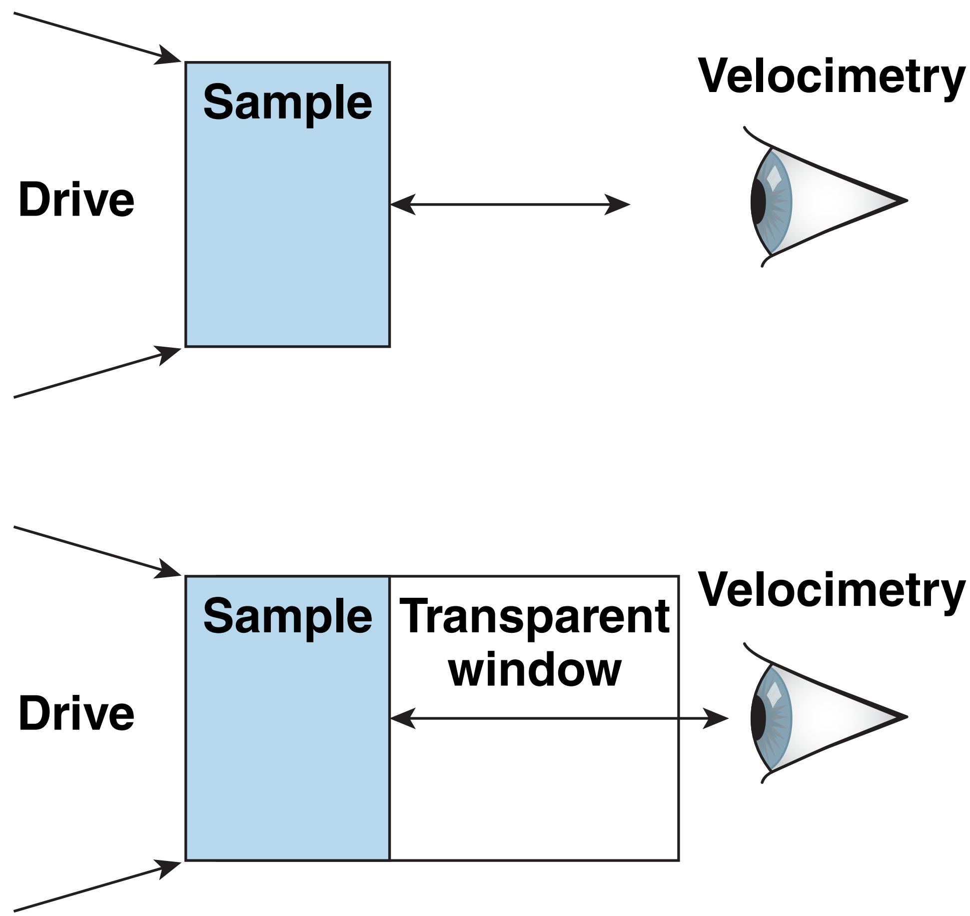
University of Rochester
Laboratory for Laser Energetics

**M. A. Barrios, J. H. Eggert, D. G. Hicks, R. Smith,
D. Braun, P. M. Celliers, and G. W. Collins**

Lawrence Livermore National Laboratory

There is significant technical utility in ramp-compressed optical windows for material studies

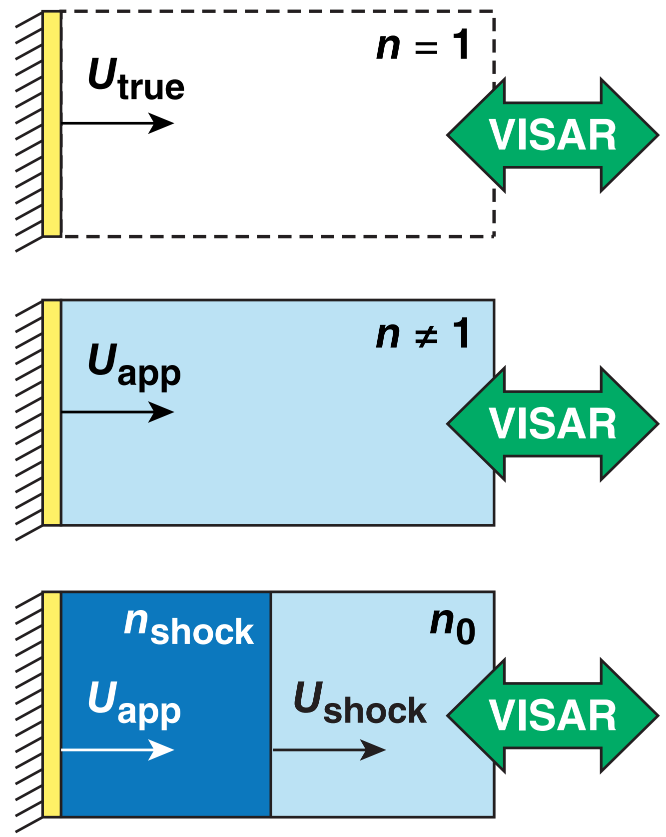
- Optical velocimetry is a powerful tool that probes the response of materials under dynamic compression
- Transparent optical windows maintain compression within a sample enabling *in-situ* particle-velocity measurements for EOS studies
- Accurate optical velocimetry measurements require that the $n(\rho)$ of the window is known



Theory

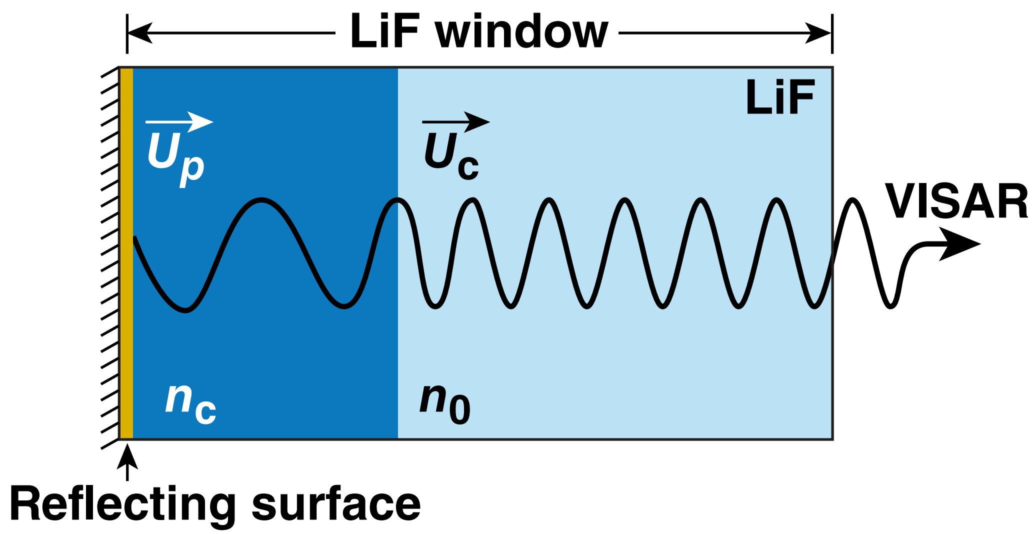
The refractive index of optical windows affects VISAR velocity measurements

- VISAR detects Doppler shifts from moving surfaces
- Optical windows influence the Doppler shift
- Changes to the window alter the optical path length of the probe beam
- The refractive index is determined if the apparent velocity (U_{app}) and the true velocity (U_{true}) are known



E19483a

The transparency of shocks in LiF windows makes it possible for VISAR to probe the material interface



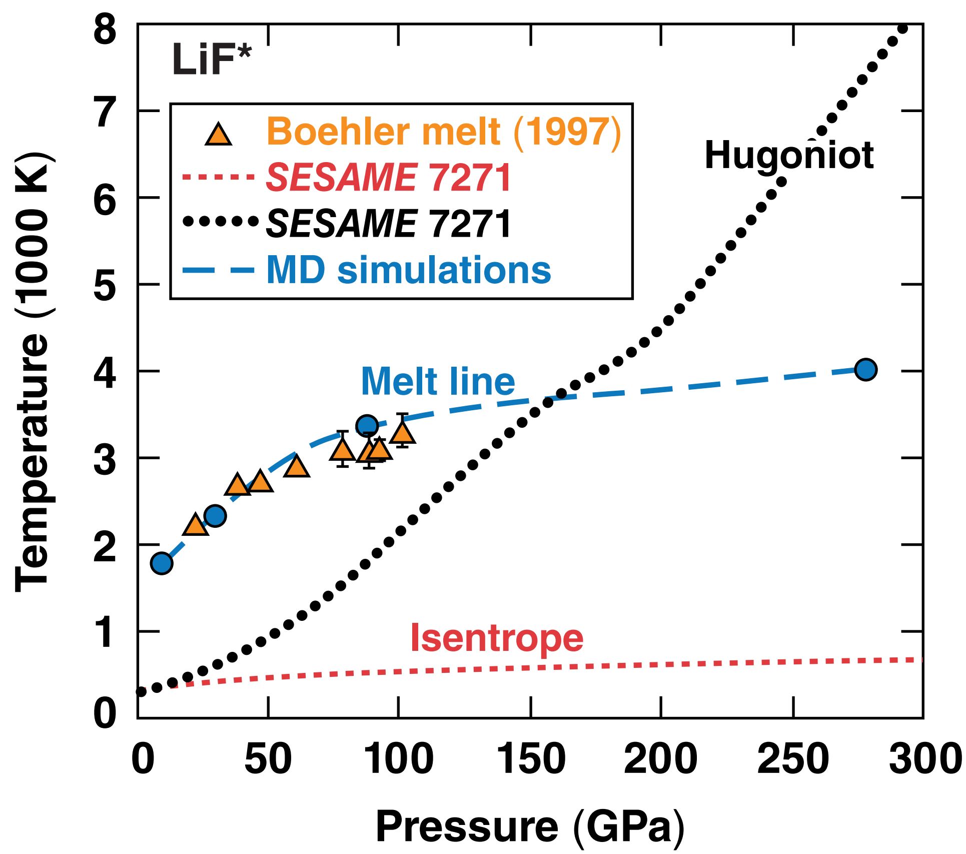
- Single shocks up to 160 GPa are transparent in LiF*
  - multishocks up to 500 GPa are transparent
- VISAR probes through the compressed material; this alters its sensitivity
- VISAR measures the rate of change of the optical path length
  - corrections must be made for the compressed refractive index (n_c)

E18046d

*J. L. Wise and L. C. Chhabildas, Shock Waves in Condensed Matter 1985, DEAC04-DP00789

Shock experiments are limited by Hugoniot melt and shock entropy

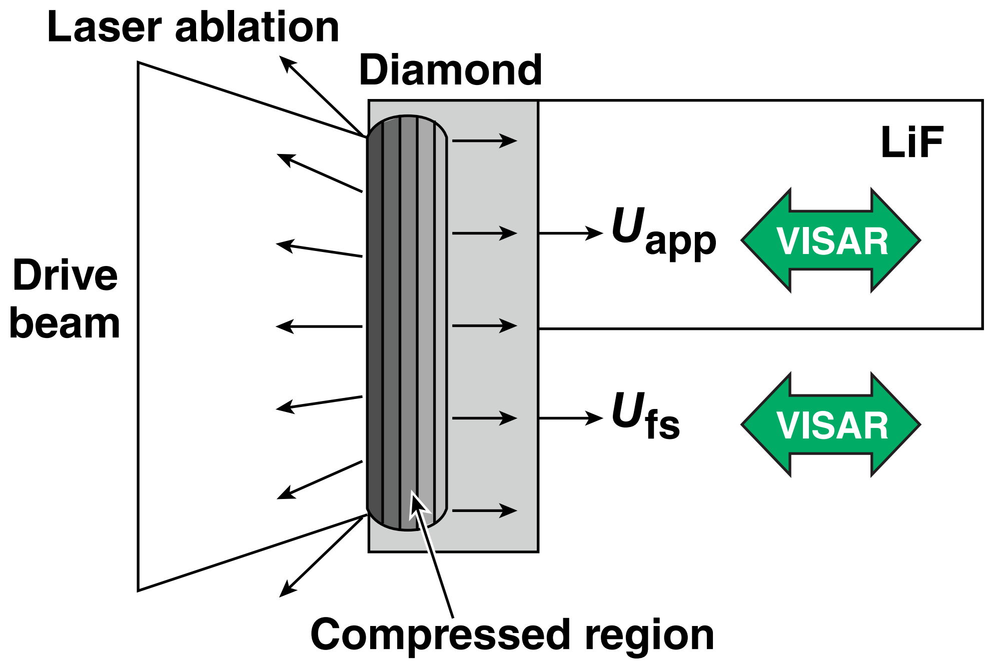
- Transparent insulators transform into conducting fluids
  - pressure-induced reduction of the band gap and thermal promotion of electrons across that gap
- Entropy produced by high-pressure shock waves transforms materials into conductive matter
  - highly reflective at the shock front



E19149

*R. Boehler, M. Ross, and D. B Boercker, Phys. Rev. Lett. 78, 4589 (1997).

A two-section target enables simultaneous measurements of the apparent and true particle velocity to be made



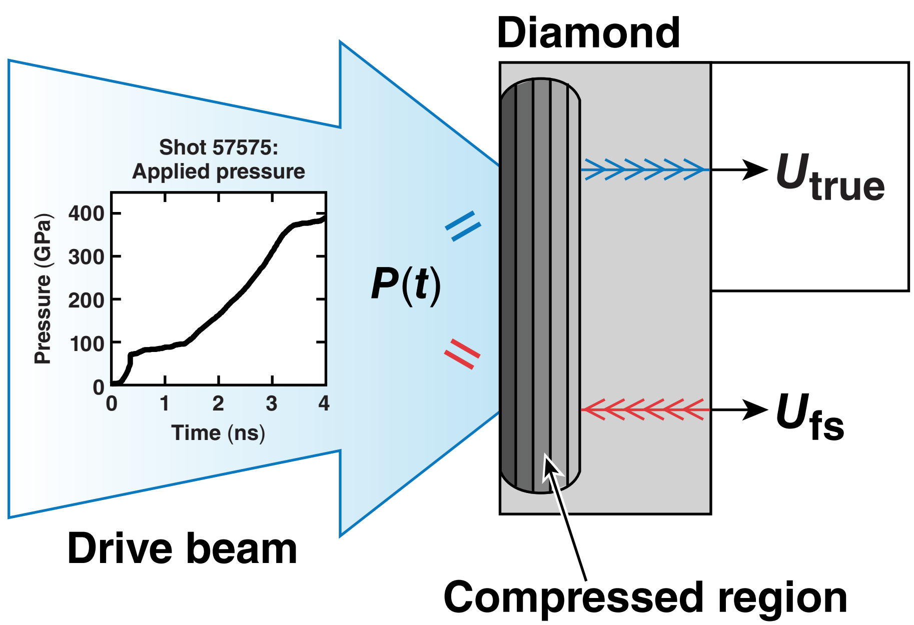
- Hayes* shows that for ramp compression

$$\frac{dU_{app}}{dU_{true}} = n - \rho\frac{dn}{d\rho}$$

- Two measurable parameters are required to determine $n(\rho)$
- U_{true} is determined using specially designed targets and the method of characteristics

*D. Hayes, J. Appl. Phys. 89, 6484 (2001).

Method of characteristics and impedance matching determines the true particle velocity



Measure U_{fs} (t)

↓

Backward characteristic algorithm to drive surface

↓

Calculate $P(t)$

↓

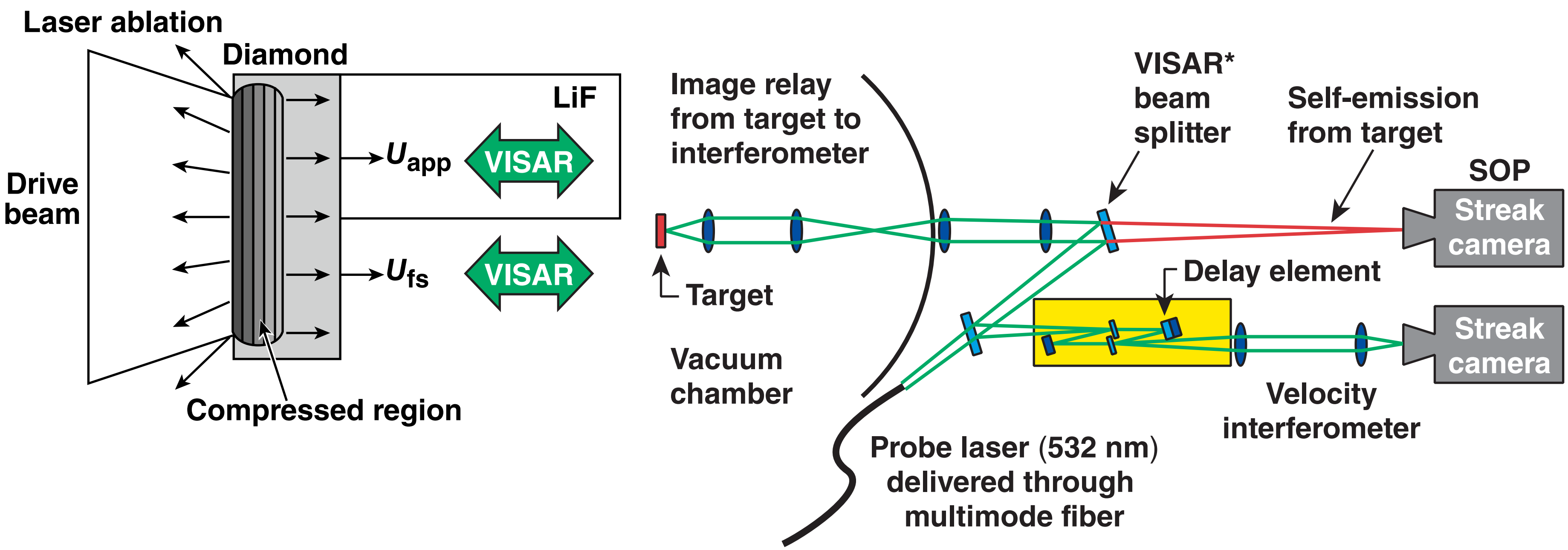
Forward characteristic algorithm to diamond/LiF interface

↓

Calculate U_{true} (t)

Diamond ramp isentrope from
D. K. Bradley *et al*., Phys. Rev. Lett. 102, 075503 (2009).

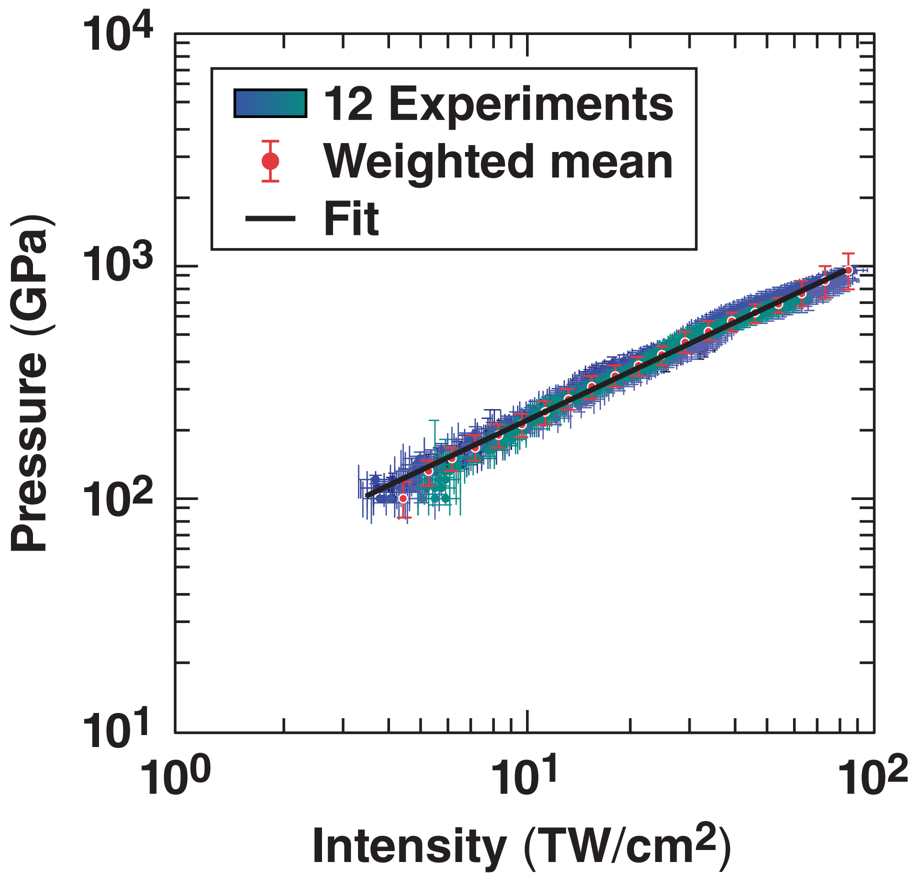
LiF refractive-index experiments were performed using laser-drive ramp compression on OMEGA



- Experiments used laser energies between 270 to 770 J delivered in 3.7- or 7.0-ns ramp profiles
- VISAR* has a time resolution of <30 ps

*Velocity interferometer system for any reflector

The applied diamond-pressure scaling law is determined from VISAR and laser-power measurements*

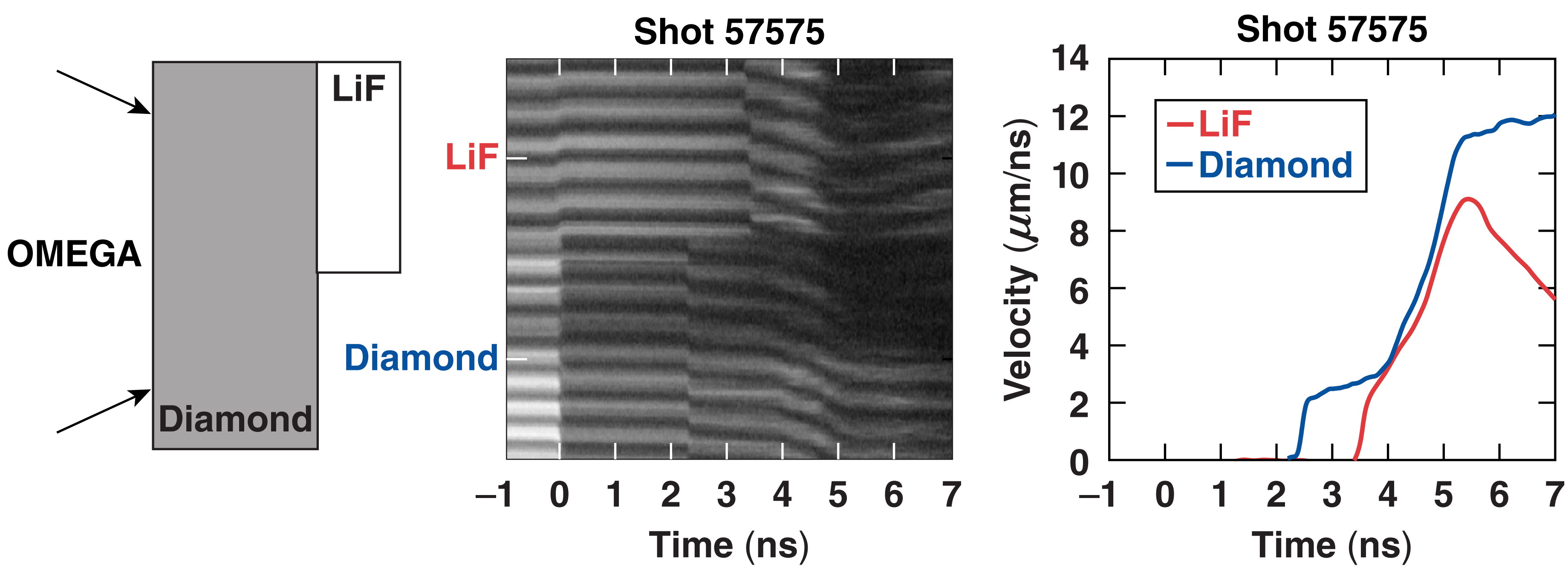


$$P(\text{GPa}) = 42(\pm 3)\left[I\left(\text{TW}/\text{cm}^2\right)\right]^{0.71(\pm 0.01)}$$

E19616

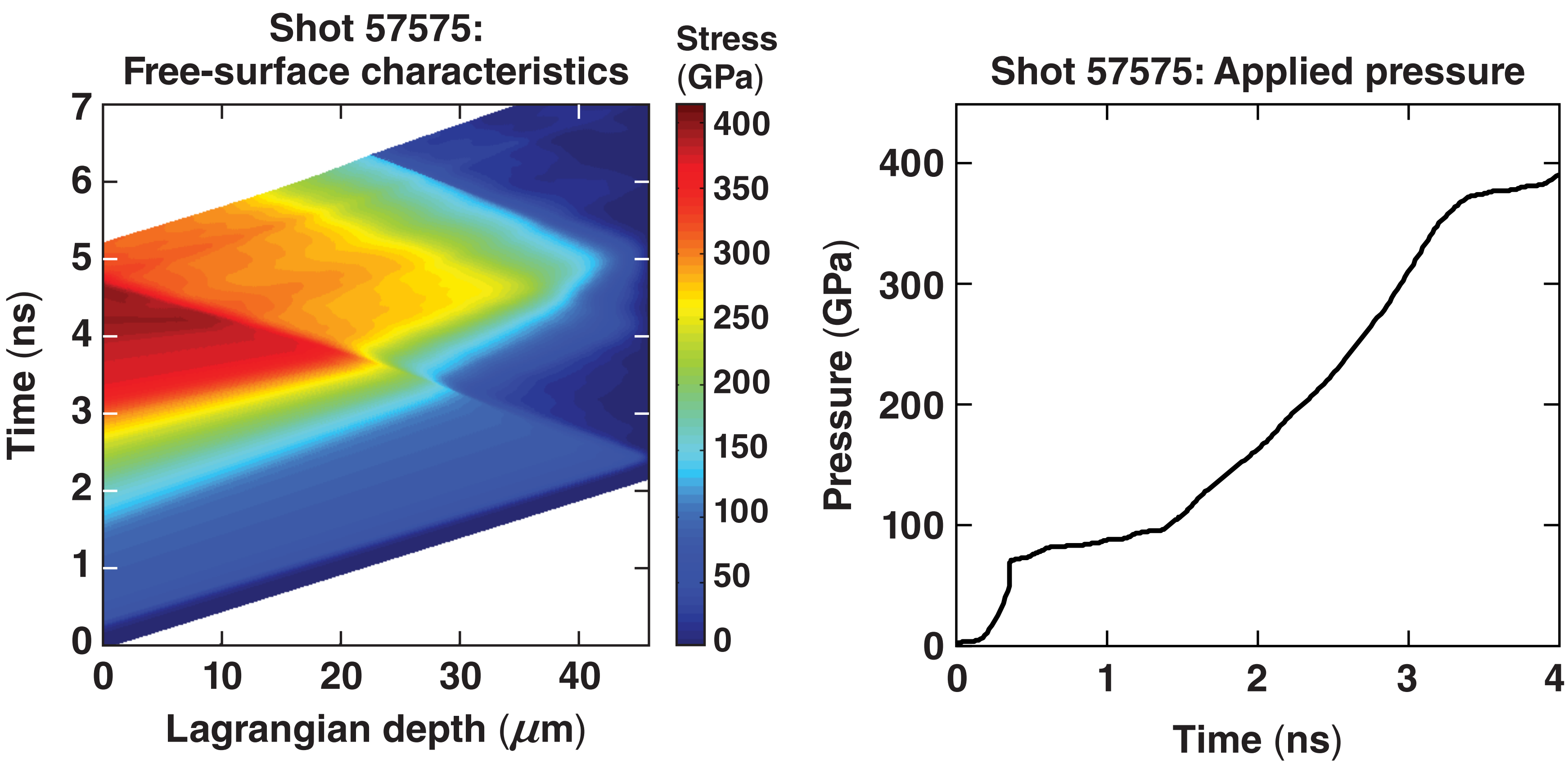
*D. E. Fratanduono *et al.*, J. Appl. Phys. 109, 123521 (2011).

Diamond free-surface and apparent interface velocity are measured simultaneously with VISAR on OMEGA



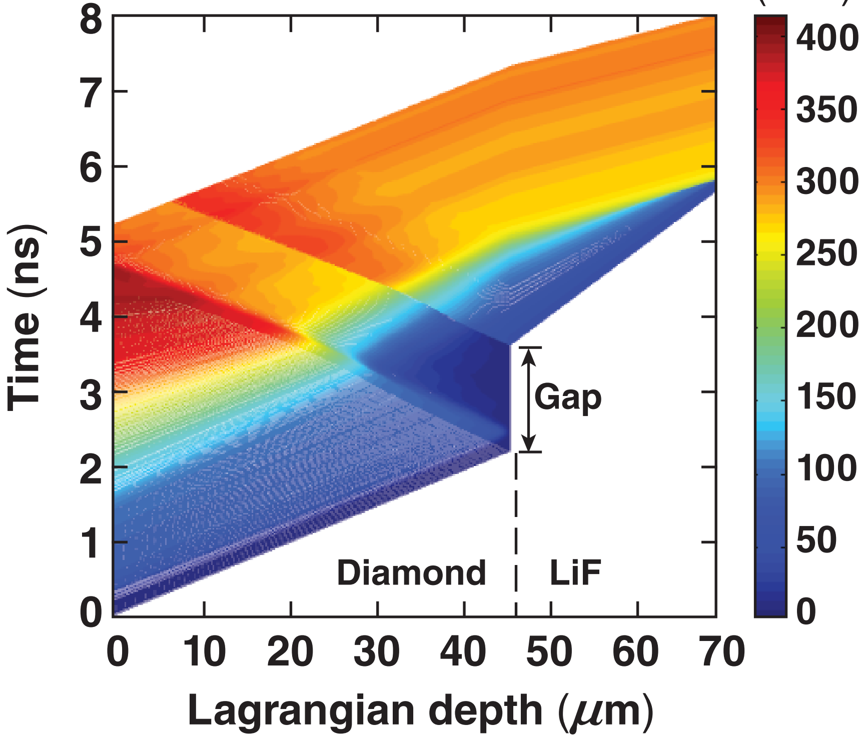
E19056c

Applied pressure is determined from diamond free-surface velocity using the backward characteristics

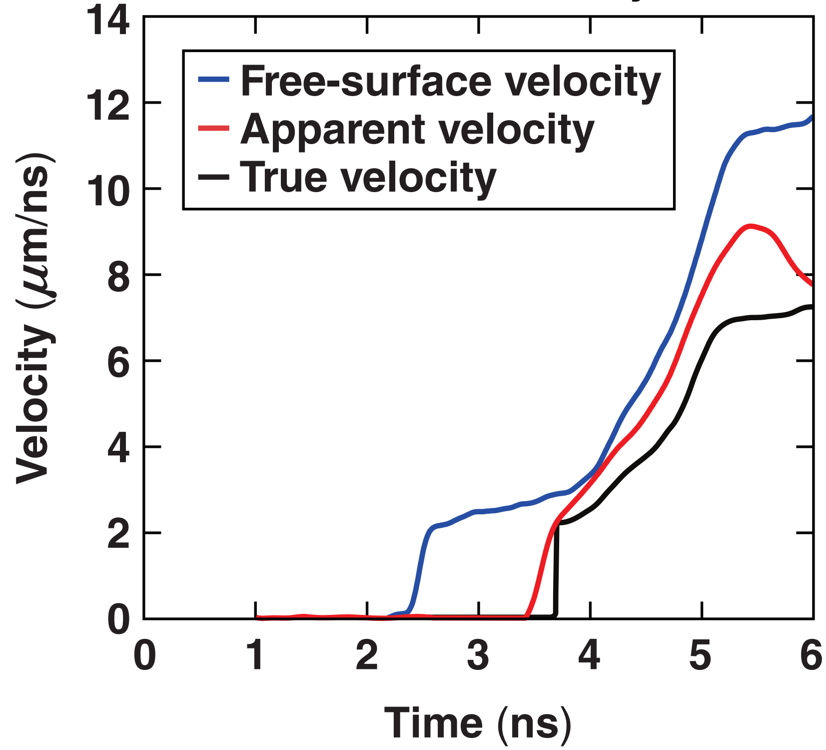


The applied pressure is forward propagated through the LiF window to infer the true particle velocity

Shot 57575:
Interface characteristics



Shot 57575:
Interface velocity

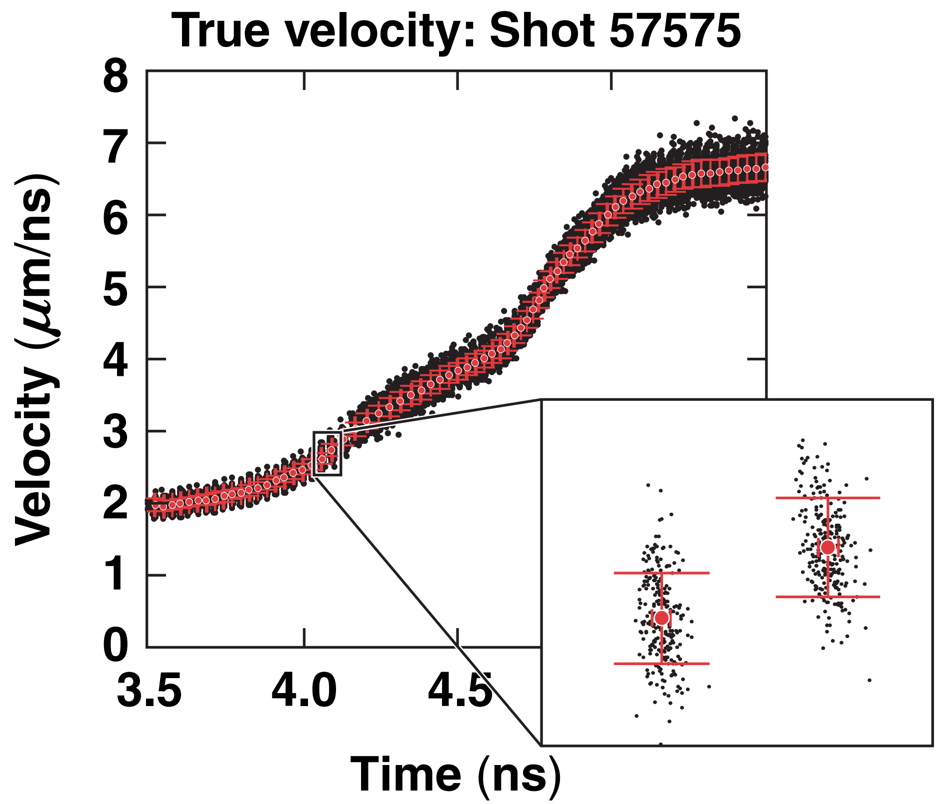


The Monte Carlo technique is used to estimate errors associated with the calculated true velocity

Monte Carlo errors

- Experimental diamond isentrope
  - standard deviations provided*
- LiF EOS (*SESAME* 7271)
  - assumed 10% error in pressure
- Gap thickness between diamond anvil and LiF window

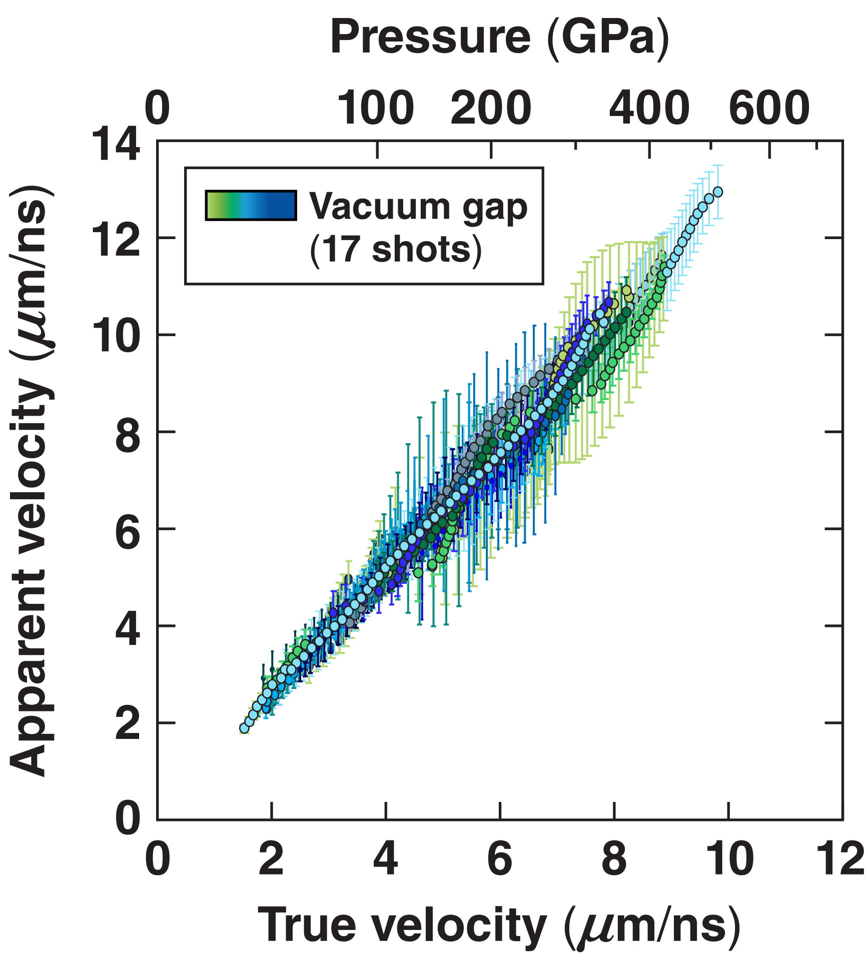
Timing errors (of the order of the etalon delay) were incorporated following the Monte Carlo routine



E19155

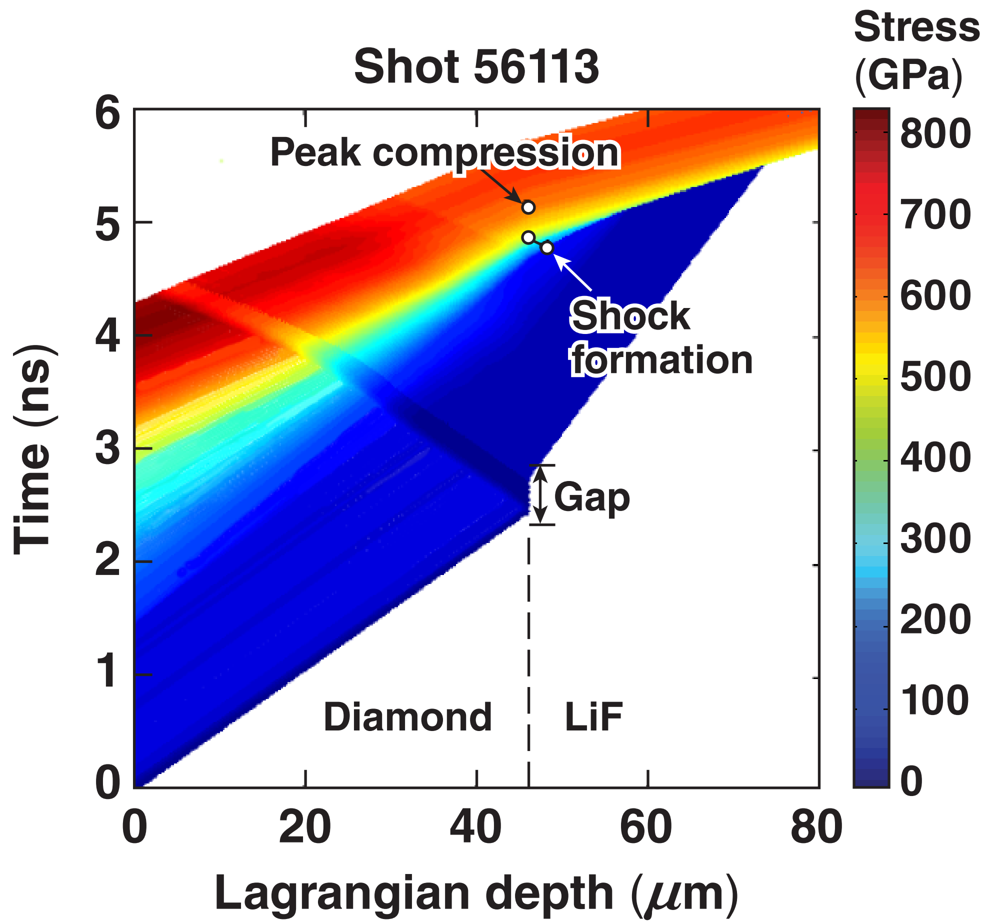
*D. K. Bradley *et al*., Phys. Rev. Lett. 102, 075503 (2009).

The apparent velocity depends linearly on the true particle velocitiy for 17 experiments

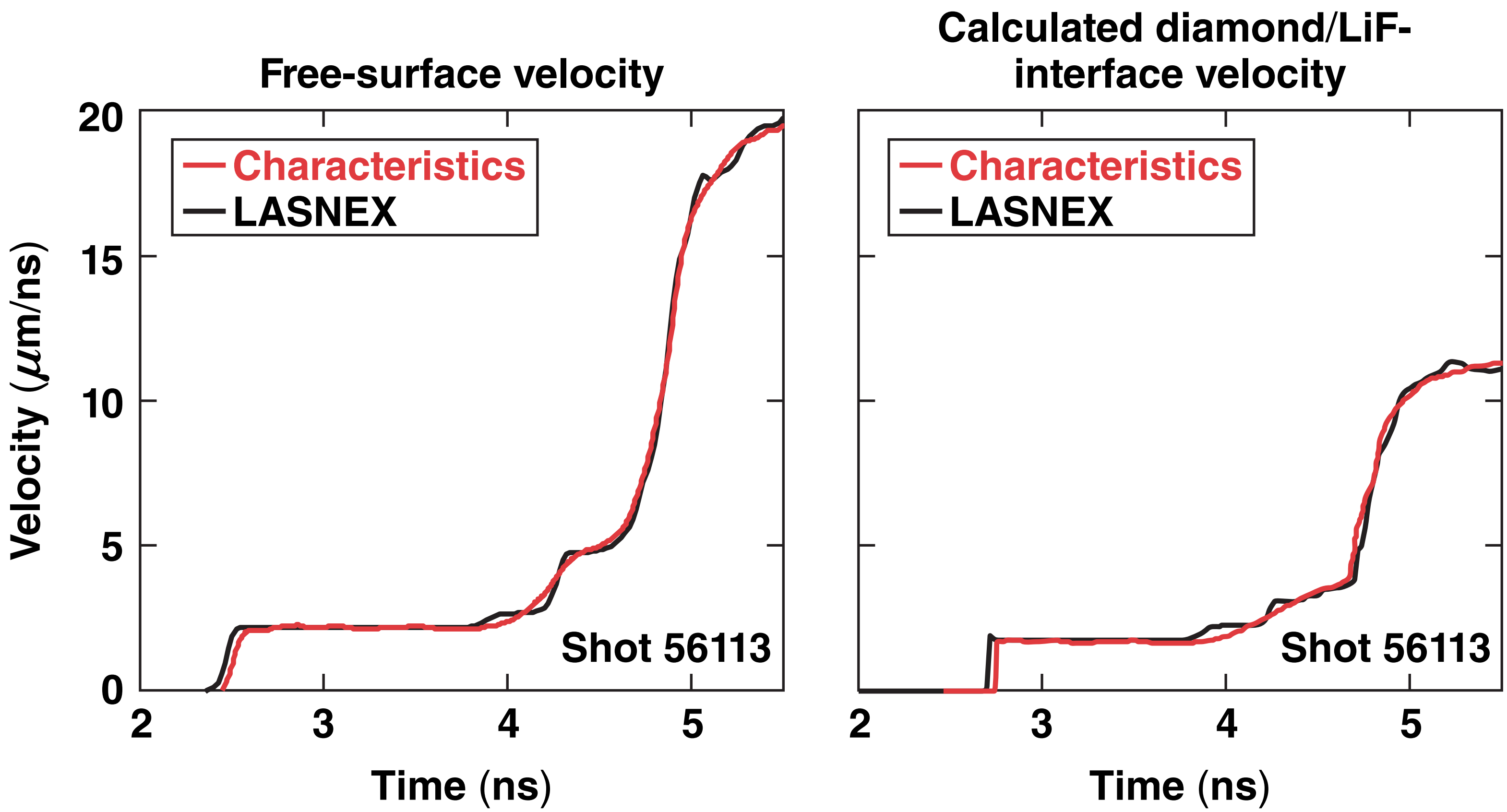


LASNEX simulations were performed to validate the method of characteristics and address concerns regarding shock formation in the LiF window

- LASNEX simulations tested the effects of shock formation and gap thickness on the calculated interface velocity
- It was found that the gap thickness and shock formation do not significantly affect the interface velocity
- Excellent agreement was observed between LASNEX simulations and the method of characteristics



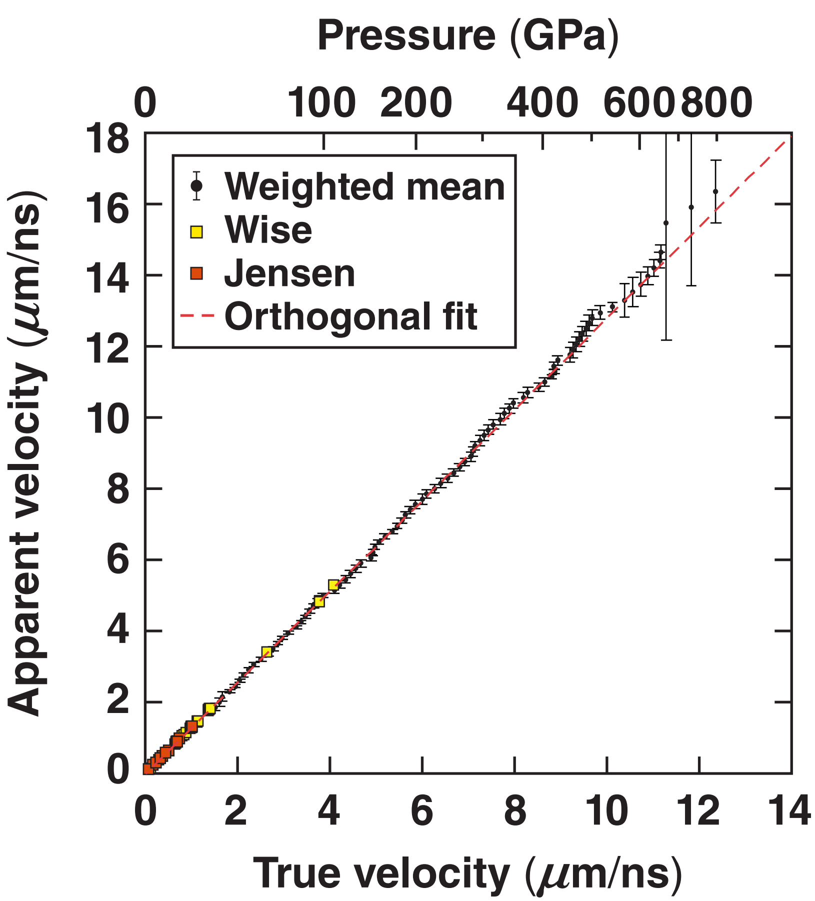
LASNEX simulations show that shock formation in the LiF window does not perturb interface measurements

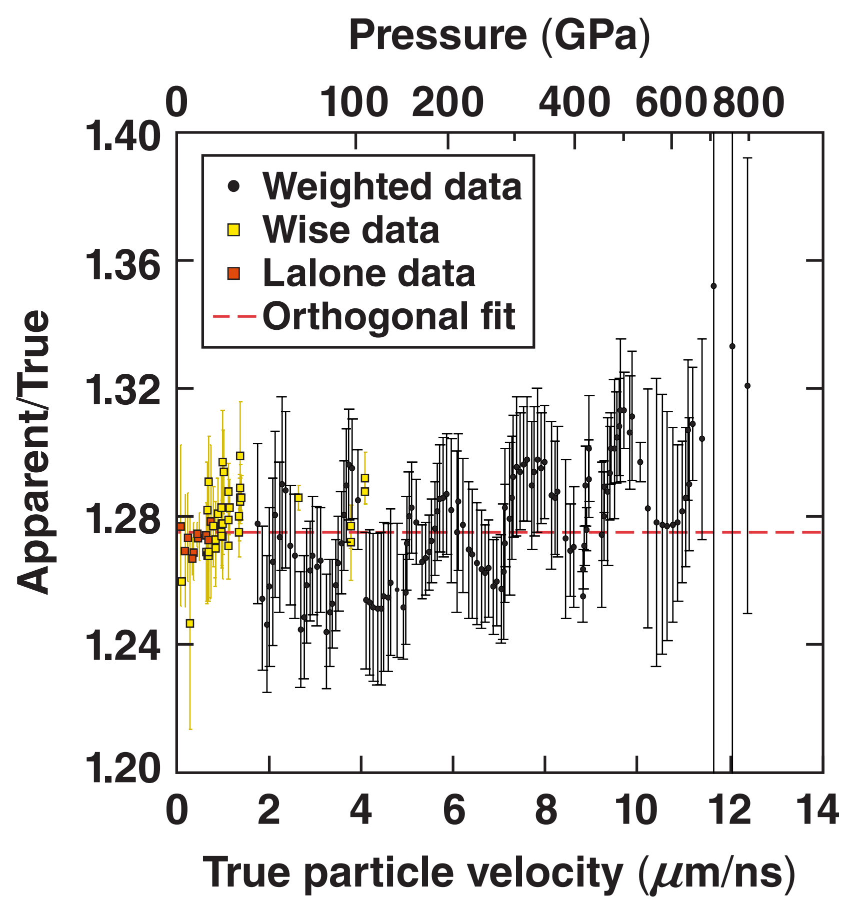


Agreement between LASNEX simulations and the method of characteristics validates this technique and uncertainties caused by the vacuum gap.

E19061

A second-order, orthogonal-polynominal regression determines the relation of the apparent and true particle velocities





E19203

Results

A linear dependence on refractive index is observed

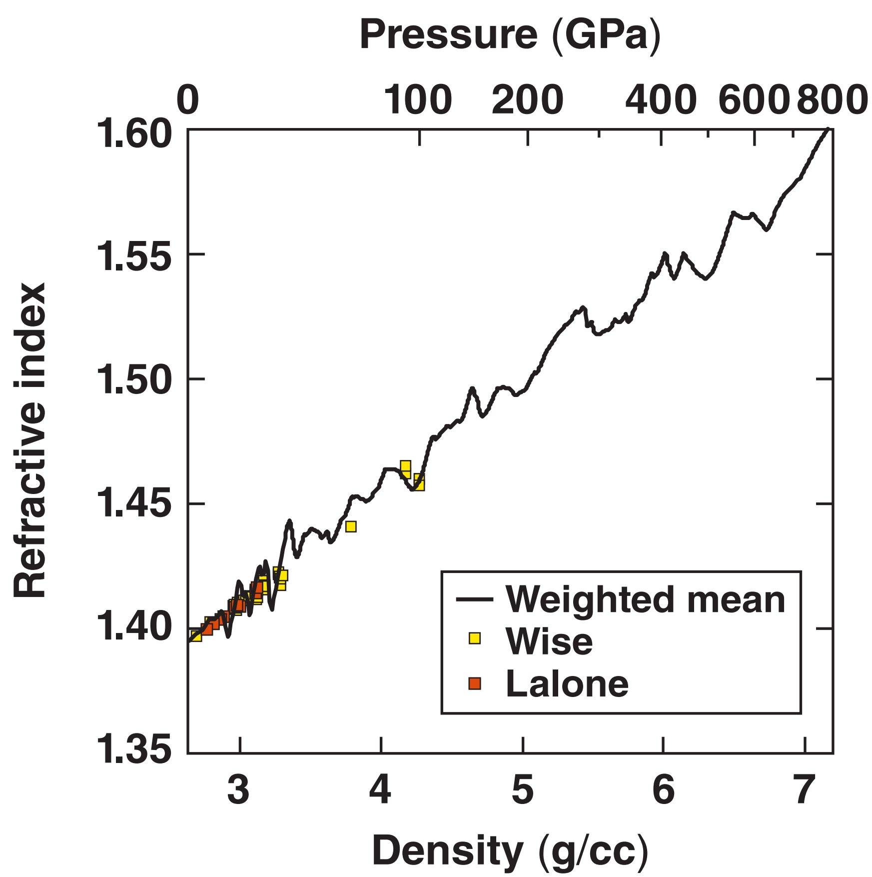
- Refractive index is determined from the Hayes formula*

$$\frac{dU_{app}}{dU_{true}} = n - \rho\frac{dn}{d\rho}$$

- For linear U_{app} (U_{true})**

$$n = a + b\rho$$

$$n = 1.2751\ (\pm 0.0082) + 0.0440\ (\pm 0.0022)\ \rho$$

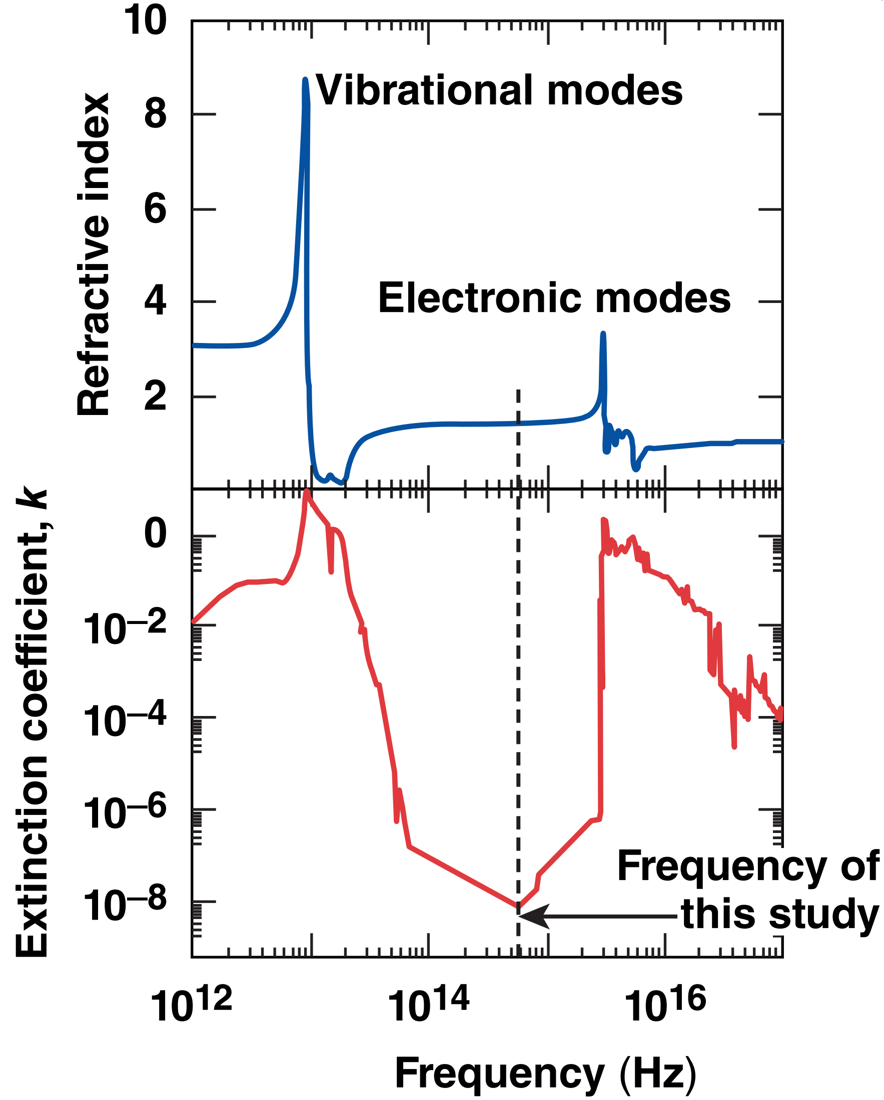


E19160c

* D. Hayes, J. Appl. Phys. 89, 6481 (2001).
** D. E. Fratanduono *et al.*, J. Appl. Phys. 109, 123521 (2011).

The effective-oscillator model is used to interpret the linear dependence of the refractive index and density

- Only electronic excitations are considered
  - ω is assumed to lie above the vibrational modes
- Changes in the refractive index caused by increases in density are due to a shift in the electronic resonance to lower frequency



E19270

E. D. Palik, *Handbook of Optical Constants of Solids* (Academic Press, Orlando, 1985).

The single-oscillator model accurately describes the dispersion of the refractive index

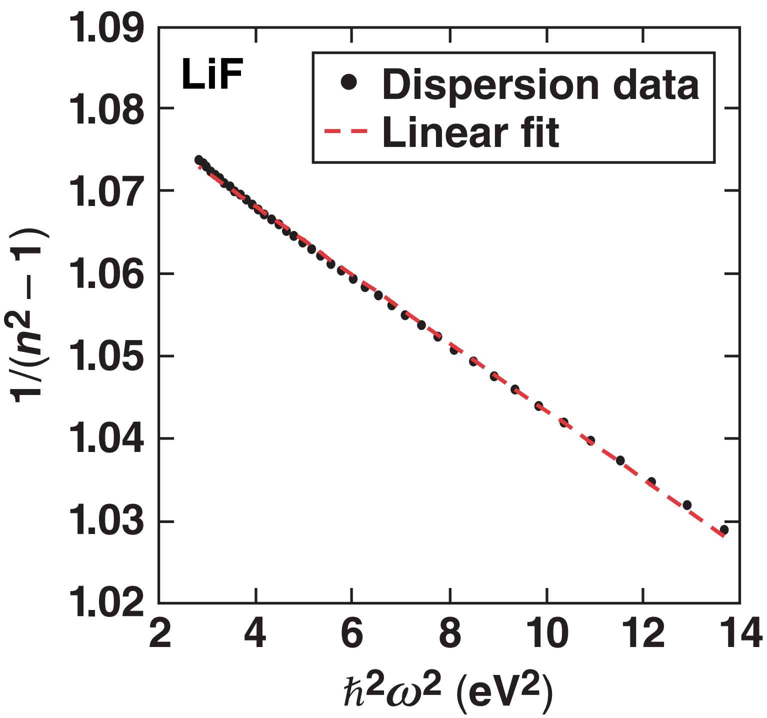
- Over 100 solid and liquid insulators obey this model

$$n^2 - 1 = \frac{E_d E_0}{E_0^2 - \hbar^2 \omega^2}$$

  – E_d is the dispersion energy

  – E_0 is the single-oscillator energy

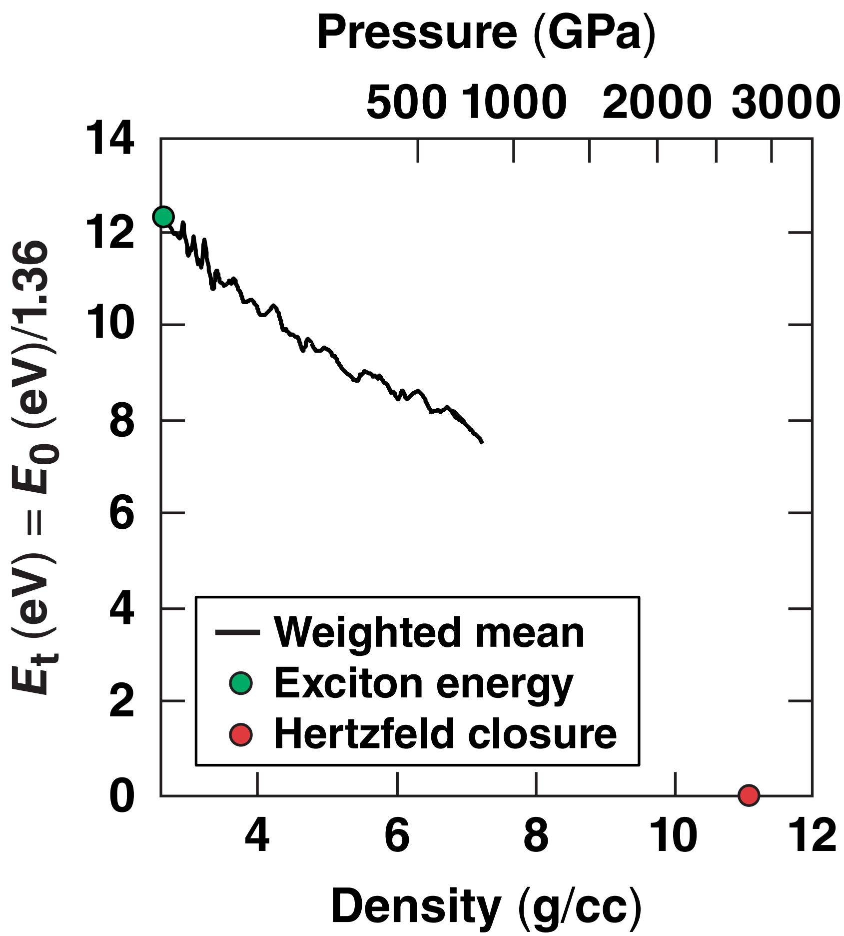
    - for the alkali-halides, E_0 is related to the excitonic energy (E_T) by $E_0 \simeq 1.36\, E_T$



- Previous studies on compressed H_2 and H_2O to 100's of GPa have shown that E_d is insensitive to changes in density

E19484

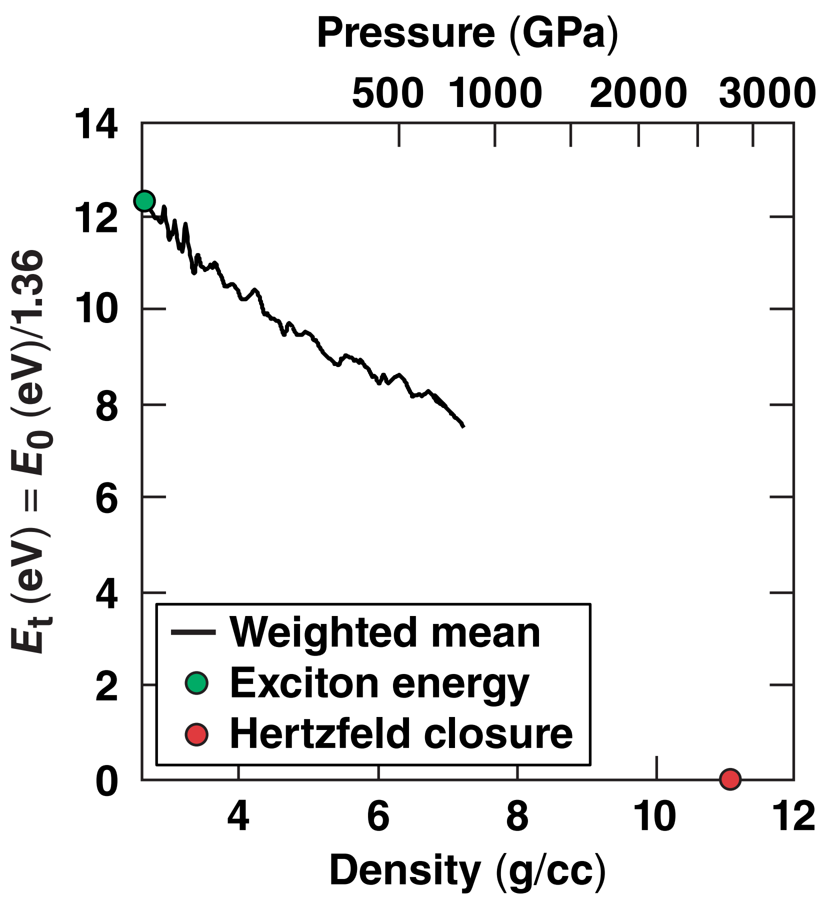
An effective-oscillator model suggests LiF will remain transparent to pressures above 4200 GPa



Extrapolation of these results suggests metallization will occur at ~4200 GPa.

E19485

The effective-oscillator model suggests LiF will remain transparent to pressures above the Goldhammer–Herzfeld metallization



	P_{GH} (GPa)	P_{OA} (GPa)	P_{BSC} (GPa)
LiF	2800	–	–
RbI	127	85	122
KI	125	115	155
CsI	105	111	100
Xe	150	–	132
He	–	–	11,200
Ne	–	–	158,000

GH: Goldhammer–Herzfeld criterion
OA: Optical absorption
BSC: Band structure calculation

High metallization pressure of LiF suggests that it will be a valuable window for high-pressure ramp-compression experiments.

E19163a

Summary/Conclusions

The refractive index of ramp-compressed LiF depends linearly on density up to 800 GPa

- Knowledge of LiF's compressed index of refraction is important for high-pressure equation-of-state (EOS) measurements
- The refractive index of shock-compressed LiF has previously been measured to 115 GPa*
- Ramp-compressed LiF is measured up to 800 GPa
  - LiF is observed to remain transparent over this range
- A single-oscillator model suggests that the band gap will close at pressures above 4200 GPa
  - these are the highest ever refractive-index measurements of an insulator

*J. L. Wise and L. C. Chhabildas, Sandia National Laboratories, Report SAND-85-0310C, NTIS (1985).