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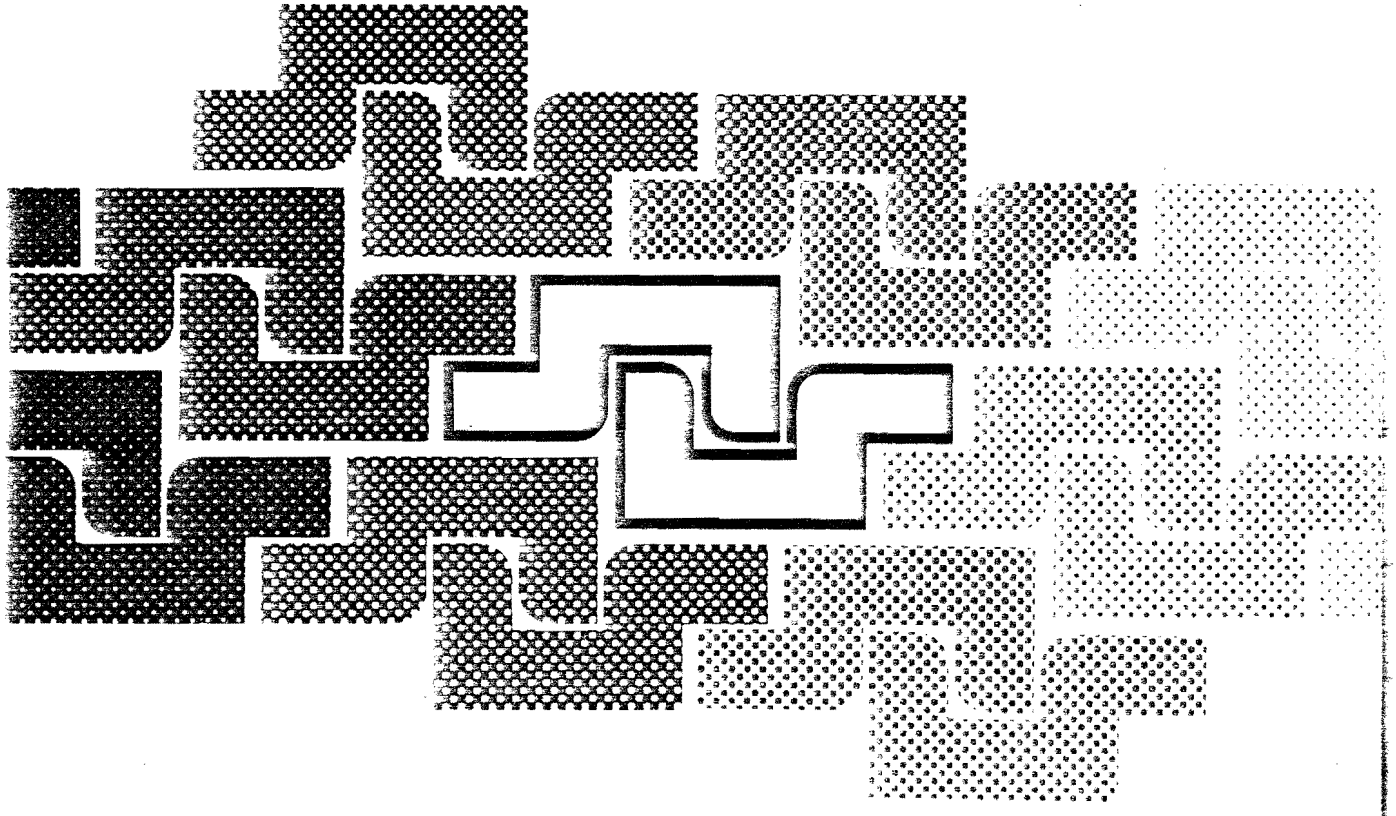

Microscopic Simulations of Complex Flows

Edited by

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NATO ASI Series

SIMULATION OF MECHANICAL DEFORMATION *VIA* NONEQUILIBRIUM MOLECULAR DYNAMICS*

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ABSTRACT

We are developing two- and three-dimensional pair-force and embedded-atom simulations of mechanical deformation processes--indentation, machining, and inelastic ballistic-impact collisions--related to current nanometer machining practice. Here we describe these problems and their implementation using both mainframe and parallel-processor computers.

1. INTRODUCTION

Nanometer technology is an engineering term for the design and fabrication of precision parts with spatial tolerances comparable to microscopic interatomic spacings. Applications include the transmission of electromagnetic waves through large accurately-shaped optical structures as well as the electromagnetic storage of information using nano- (as opposed to micro-) circuitry.

Macroscopic thermodynamics, hydrodynamics, and solid mechanics describe equilibrium and nonequilibrium states of continua. These disciplines make up "continuum mechanics". Their application, through the solution of *partial* differential equations, is the traditional approach to designing and fabricating engineering structures. Conservation of mass, momentum, and energy are fundamental to either the macroscopic

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continuum aproach or to the alternative microscopic atomistic approach. The distinguishing feature of the macroscopic approach is the use of "constitutive equations" which describe the response of particular materials to gradients of the fundamental conserved quantities.

The alternative microscopic approach follows the history of individual atoms by solving the *ordinary* differential equations of motion. The distinguishing feature of the microscopic approach is the use of action-at-a-distance force functions acting among sets of mass points. The macroscopic continuum approach can be thought of as describing averages of microscopic atomistic properties to obtain constitutive properties for *regions* containing many atoms (at least millions). But when the *scale* of a part or feature is *small*, as at a crack tip, dislocation, vacancy, or impurity, the continuum approach fails[1]. This failure shows up in a fundamental way in what is called "size dependence", the failure of material properties to obey simple scaling relationships. If stress were solely a function of strain then the time-and-space-dependence of the macroscopic equations of motion could be scaled. Structures could be built with confidence based solely on the performance of scale models. The fact that large specimens break under smaller stresses than do small specimens of identical shape indicates the presence of a characteristic length (the atomic spacing).

The computer industry is profitting from miniaturization, and the result is a scaleup in the size of simulations it is feasible to make. It does not seem possible to gain much speed by further reduction of the fundamental computer cycle time. But what *can* be done is to increase, greatly, and at little cost, the number of degrees of freedom being treated. The increase makes the difference between a few-body caricature and a truly many-body *simulation*. Many small computers, working in parallel, can now accomplish the same tasks as can the large computers, but 1000 times more cheaply and in the same clock time.

Because the crystal dislocations fundamental to plastic flow have long-range interactions, flow results can be affected by boundaries lying many atomic diameters away. For this reason relatively large (nanometer-scale) simulations are necessary to the atomistic simulations of flow and failure processes in materials science. Such simulations are becoming feasible. Because multiprocessors with millions of processors are on the horizon there is no doubt that billion-particle simulations will eventually be carried out, but still only for times up to about a microsecond. This is exactly the physical scale required for an understanding of shockwave deformation, fracture, and high-strain-rate plasticity. It is because large-scale simulations are becoming a reality that we feel it worthwhile to develop computational tools for addressing these problems.

In this paper we describe our progress and plans.

2. SIMULATION METHODS

Forces and boundary conditions, together with a computer, graphic output, and an integration alogrithm, are the requirements for an atomistic simulation. Hooke's-law and Lennard-Jones forces are instructive and useful models for two-body "pair" forces, and we began with these two. The Hooke's-law crystal obeys exactly the same motion equations as does a two-dimensional bilinear finite-element representation of an elastic continuum[2],

for which many analytic and numerical solutions are available. The anharmonic Lennard-Jones potential has a known and relatively-simple phase diagram and has been the subject of hundreds of equilibrium and nonequlibrium simulations ever since the development of the Monte Carlo method at Los Alamos after the Second World War.

In the past ten years there has been increasing emphasis on simulating "real materials". Only a part of this increasing emphasis comes from the shift toward applied and away from basic research. Improvements in computation and experiments are responsible too. Increasing computer power makes it feasibile to use ten-parameter potential functions with angle-dependent forces in combination with disordered structures and sophisticated boundary conditions to model such diverse materials as water, glasses, and proteins under both equilibrium and nonequilibrium conditions. Ever-more-precise experimental tools, such as the field-ion, force-balance, and scanning-tunneling microscopes are providing ever better tests to challenge such simulations.

Pair potentials can describe simple materials, such as the rare gases, relatively well. But pair potentials cannot describe directional chemical bonds, or account for the observed large differences between the elastic moduli C_{12} and C_{44}, or reproduce the differences between the cohesive energy and the vacancy energy. Daw[3] had the idea of introducing a local many-body potential that incorporated these features at low cost, the "embedded-atom" potential.

Pair forces have been studied for nearly a century and the corresponding macroscopic properties are fairly well understood. Because the embedded-atom idea is relatively new there is not yet a correspondingly good understanding of the correlation between potential parameters and macroscopic properties such as yield and fracture toughness. But this will come with experience. We have followed Daw's lead in investigating this low-cost approach to metal simulation.

The basic idea is to calculate **Particle i**'s contribution, $\phi(r_i)$, to the coordinate-dependent potential energy $\Phi(\{r_k\})$ as a function of the density at that particle due to the influence of the neighboring particles at $\{r_j\}$:

$$\rho_i = \Sigma\rho(r_{ij}).$$

Particle **i** can be thought of as being "embedded" in a field provided by its neighbors. So long as this embedding is viewed as a pragmatic and phenomenological low-cost procedure for avoiding prohibitively-expensive quantum simulations, it represents the best method for predicting the properties of metals and their defects.

Boundary conditions are dictated by the corresponding experimental situation. The boundaries can either be free of forces, or they can be subject to prescribed time-dependent displacements or forces. In Vineyard's pioneering work[4] viscoelastic boundaries were used to absorb the wave energy incident on walls. In continuum mechanics corresponding "quiet" boundaries are sometimes used. The main advance in treating boundaries since Vineyard's work has been Nosé's reversible and deterministic method[5] for introducing temperature and stress into the microscopic equations of motion. We have incorporated this method in our work.

The simplest reasonable integration method for conservative systems is the time-reversible centered second-difference algorithm in which the accelerations are replaced by differences:

$$d^2\mathbf{r}/dt^2 \equiv [\mathbf{r}(t+dt) - 2\mathbf{r}(t) + \mathbf{r}(t-dt)]/(dt)^2.$$

But because many interesting equations of motion, such as Nosé's, are *first*-order in time, rather than *second*, and also because the same accuracy can be obtained easily with fewer force evaluations, and significantly reduced computer time, we prefer the classic "Fourth-Order" Runge-Kutta approach. With Runge-Kutta[6,7], four easy-to-program estimates of $\mathbf{dr}/dt$ and $\mathbf{dp}/dt$ are averaged, with a resulting error, over a fixed interval of time, proportional to the *fourth power* of the timestep. For the simple second-difference scheme above the integration error over a fixed time interval is larger, varying as the *square* of dt.

The recent book **Numerical Recipes**[7] claims that the *Bulirsch-Stoer* integration method is likely the best for integrating ordinary differential equations. This method uses a relatively-large timestep (a complete vibrational period for an oscillator, for instance) and evaluates a series of approximate integrals over the interval, using 2, 4, 6, 8, 12, 16, 24, ... function evaluations. These results are then extrapolated, using a Padé (or continued-fraction) approximant, to the limit of an infinite number of evaluations.

On the strength of this recommendation, we investigated the Bulirsch-Stoer integrator. As is usual, such an investigation is hampered by the failure of common library routines to allow either the order or the timestep to be specified. In "packaged software" these parameters are varied internally, at some "overhead" cost, in order to minimize the error. But, because most molecular-dynamics simulations proceed with a roughly constant degree of anharmonicity there is no reason to consider varying the order or timestep to improve accuracy. Diligent comparison of the packaged International Mathematics and Statistics Library (IMSL) Bulirsch-Stoer routine "IVPBS" with the **Numerical Recipes** version showed that the two do indeed, apart from the error and order controls, implement precisely the same method.

A detailed investigation for the one-dimensional harmonic oscillator (See also Reference 6) showed that Bulirsch-Stoer approach, with optimized timestep and number of force evaluations, matched the performance of the popular Gear integrator when the required maximum coordinate error was set at one part per million. This level of error required about one hundred force evaluations per oscillator period. With fewer evaluations the Bulirsh-Stoer performance degrades much more rapidly than the others. Encouraged by the oscillator results we applied the same technique to Lennard-Jones crystals. Here the Bulirsch-Stoer performance is disappointing, even relative to the simple Runge-Kutta integrator, and we therefore abandoned the Bulirsch-Stoer technique.

Today the Lawrence Livermore National Laboratory emphasizes "Supercomputing", computing with mainframe machines costing tens of millions of dollars. But, for molecular dynamics, this approach has the look of a dinosaur. Interesting low-cost alternatives are becoming available. The SPRINT (Systolic Processor with Reconfigurable Interconnection Network of Transputers) computer[8] is about 1000 times cheaper than a state-of-the-art CRAY. It was developed as a doctoral thesis project at the Lawrence

Livermore National Laboratory under the auspices of the University of California's Davis/Livermore Department of Applied Science.

The SPRINT has 64 "Transputer" chips, each a 32-bit microprocessor with about three times the speed of a VAX-11/780. The processors can be connected up in a variety of topologies. In the checkerboard configuration appropriate to two-dimensional molecular dynamics simulations the SPRINT performed with a speedup over a single transputer between 16 and 32, carrying out 1000-atom molecular dynamics simulations at the same clock speed as a CRAY-1 computer. The loss of speed, relative to that of a single transputer, is due to the need for communication of information among the processors. Because these transfers only involve neighboring parts of the problem, the overall efficiency of the multiprocessor should remain roughly unchanged with increasing problem size.

The rapid display of computed results is still a bottleneck, even at Livermore. A thousand-frame ten-thousand particle movie runs for less than a minute, but requires the processing of about 10^8 particle or pixel coordinates. This is too much information for a single tape to hold. The portability and lower cost of videotape make that medium preferable to conventional movies. To make a finished videotape requires several hours of processing effort, with a considerable portion of that time devoted to transporting data from one machine to another. (CRAY to VAX to STELLAR to a tape editor, at Livermore.) But the results are worthwhile. Cooperative motion and correlations that would be difficult to see in still pictures stand out in movies.

Though presentday mathematicians may claim that Poincaré "understood" the complexity of chaotic mechanics through his analysis of the intricacies of homoclinic points, there is no doubt that a short videotape of the Lorenz attractor provides a relatively effortless and accessible understanding every bit as sure and reliable as Poincaré's.

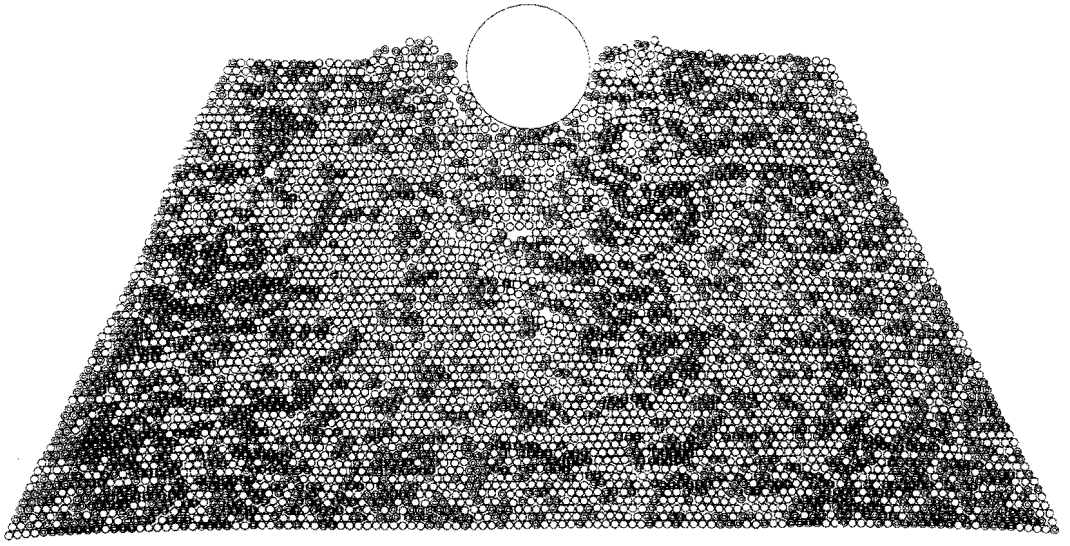

FIGURE 1. Indentation of 5430 Lennard-Jones atoms at about two-thirds the melting temperature ($kT/\varepsilon = 0.30$). The indentor speed increases linearly with time, up to a maximum value of $(\varepsilon/m)^{1/2}$, and then decreases linearly back to zero.

All of our simulations have been two-dimensional, but we are developing, for the SPRINT, the straightforward three-dimensional extension of these models. It appears that the current 64-processor SPRINT should be capable of efficient molecular dynamics simulations with 50,000 atoms. At Los Alamos Brad Holian and Art Voter expect to be able to treat one million particles on the 65,536-processor "Connection Machine".

3. SIMULATION RESULTS

We began our indentation studies with triangular and round indentors. See **Figure** 1. In testing real materials the yield strength is determined by dividing the applied load by the area of permanent deformation. We carried out sample two-dimensional calculations for a variety of indentor sizes and workpiece temperatures. We used both fixed-load and programmed-displacement boundary conditions. The resulting yield stresses (load divided by length) are *size-dependent*, still decreasing with increasing workpiece size for two-dimensional crystals with 5000 atoms. But the presence of many dislocations in the deforming crystal indicates that convergence should be possible in two dimensions. We intend to pursue convergence using a plane-strain (two-dimensional) representation of copper and to compare the resulting stress and strain fields with macroscopic finite-element simulations.

We followed our indentation studies with simulations of the diamond-turning process, in which strips only a few atoms thick are cut from a spinning workpiece through contact with diamond chips embedded in a resilient matrix. Computational specimens only a few atoms thick produced voids and extended defects at their bases, but larger specimens produced realistic chips, particularly when an embedded-atom model was used. The Lennard-Jones potential produced substantially more vapor (or dust?) in the cutting operation. A comparison of the two kinds of simulations is indicated in **Figure** 2. Note that in the evolution of these simulations a cutter radius of curvature was introduced. The embedded-atom potential we use has a functional form $\Phi = \Sigma\rho_i \ln\rho_i$, with $\rho(r)$ given by a parabola vanishing just short of the triangular-lattice second-neighbor separation. We intend to extend this work also to a more-realistic model for copper. In both the indentation and the cutting studies the peak deformation velocities were about one-tenth the sound velocity.

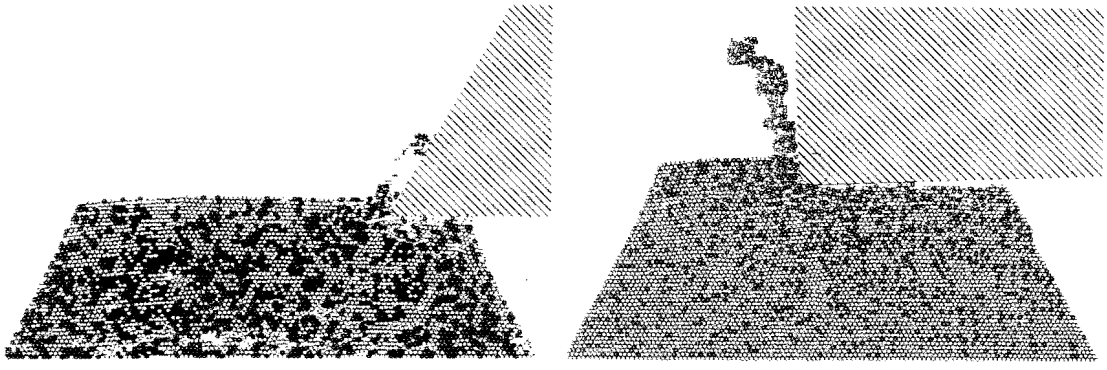

FIGURE 2. Cutting of Lennard-Jones (left) and Embedded-Atom (right) Crystals with a cutting tool moving at approximately one-tenth the sound velocity.

The simplest impact problem is the symmetric collision of identical particles. Ever since Newton's time the "coefficient of restitution" has been used to describe the result of such "inelastic" collisions, that is collisions in which the outgoing particle speed is less than the incoming one[9]. The ratio of these speeds *is* the coefficient. Because an explanation of the coefficient's magnitude is hard to find in the literature (despite considerable analysis by Rayleigh[10] and experiments carried out by Raman[11]) we carried out a short investigation. Initial trials revealed that roughly-circular disks, containing a few hundred to a few thousand atoms, bounce from a *mathematical* hard wall (at which incoming atomistic velocities are reflected) with a restitution coefficient approaching unity at low speeds, in agreement with Rayleigh's analysis. We obtained similar results for macroscopic continua using a finite-element approach.

But the collisions of particles with a mathematical "wall" seemed unrealistic. Why not simply allow two similar particles to collide? We did, and the results were amusing. *There was generally no "restitution" at all.* Instead the two particles would come together and stick, permanently cold-welded together. The dynamics underlying this phenomenon is reminiscent of the phase-space structure of the strange attractors of dynamical systems theory. The colliding two-particle system starts out with most of its kinetic energy directed. Then the collision occurs. Next, the anharmonicities and elastic wave reflections from the curved boundaries dephase, scatter, and thermalize the directed energy. The "Poincaré recurrence time" required for the momenta to again line up, allowing the particles to separate, is impossibly long. It is comparable to the time required for a similar number of particles in a box to simultaneously seek out one half or the other of the box. Thus the coefficient of restitution studies indicated rather strongly the importance of impurities on the surface. Without them restitution won't occur.

These collisional studies are not at all without applied interest. Rayleigh's calculations and Raman's pendulum experiments with "Hertzian-contact" dynamics showed that the stress reached between two colliding ball bearings is several kilobars, comparable to the yield strength of steel. Today it appears that the corresponding experiments, properly analyzed could be a useful source of high-strain-rate constitutive models. We found that our embedded-atom model, again with the potential function taken as $\Sigma\rho_i \ln\rho_i$, could plastically deform, as shown in the **Figure** 3, even with a relatively small kinetic energy, of order a few percent of the melting energy. We view this qualitative dependence of the deformation on the forcelaw as strong empirical evidence for the usefulness of the embedded-atom approach.

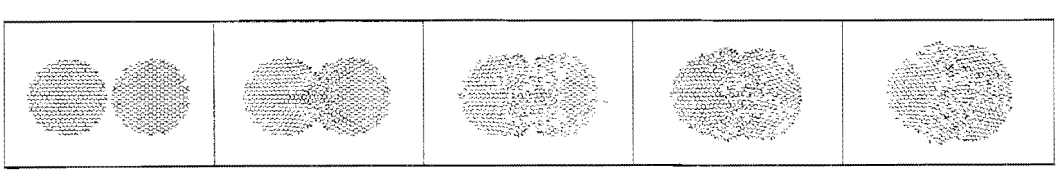

FIGURE 3. Snapshots of the low-speed inelastic cold-welding of two embedded-atom particles.

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REFERENCES

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1. D. Scott, W. W. Seifert, and V. C. Westcott, Sci. Am. **230** (May), 88 (1974).; E. Rabinowicz, Sci. Am. **218** (June), 91 (1968).
2. A. Hrennikoff, J. Appl. Mech. **8A**, 169 (1941).
3. M. S. Daw and M. I. Baskes, Phys. Rev. **B29**, 6443 (1984); M. S. Daw and S. M. Foiles, Phys. Rev. Letters **59**, 2756 (1987); S. M. Foiles, M. I. Baskes, and M. S. Daw, Phys. Rev. **B33**, 7983 (1986).
4. G. Vineyard, J. Appl. Phys., cover (August, 1959); J. B. Gibson, A. N. Goland, M. Milgram, and G. H. Vineyard, Phys. Rev. **120**, 1229 (1960).
5. S. Nosé, J. Chem. Phys. **81**, 511 and Mol. Phys. **52**, 255 (1984); W. G. Hoover, Phys. Rev. **A31**, 1695 (1985).
6. G. D. Venneri and W. G. Hoover, J. Comp. Phys. **73**, 468 (1987).
7. W. H. Press, B. P. Flannery, S. A. Teukolsky, and W. T. Vetterling, *Numerical Recipes* (Cambridge University Press, Cambridge, 1986).
8. A. J. De Groot, E. M. Johansson, J. P. Fitch, C. W. Grant, and S. R. Parker, IEEE Transactions on Nuclear Science, **34**, 873 (1987).
9. G. Barnes, Am. J. Phys. **26**, 5,9 (1958).
10. Lord Rayleigh, Phil. Mag. **11**, 283 (1906).
11. C. V. Raman, Phys. Rev. **12**, 442 (1918).