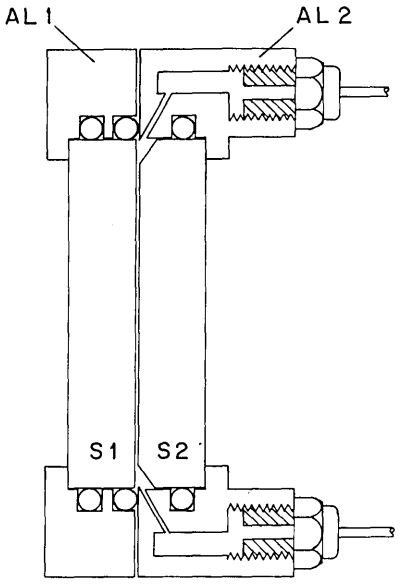

Fig. 1. Cross-sectional view of dye cell showing aluminum rings $AL1$ and $AL2$, substrates $S1$ and $S2$, the sealing O-rings, and the Swagelok connectors.

the entire cell could be repositioned, and another portion of the mirror could be used. In this way a single mirror could continue to be used following several damaging events.

The cell, shown in Fig. 1, consists of two 5.08-cm diam, 0.95-cm thick BK7 substrates, $S1$ and $S2$, mounted in aluminum rings, $AL1$ and $AL2$. Substrate $S1$ is coated on its inner surface to reflect 50% at 1064 nm. It is mounted in ring $AL1$, which has two O-ring grooves, one near its surface and the other along the side. (Although the side O-ring is redundant, it serves as a second seal to prevent dye solution leakage.) The inner reflective surface of $S1$ is flush with the flat surface of $AL1$, while the surface O-ring protrudes about 40 μm above this surface.

The second substrate $S2$ is mounted so that its inner surface protrudes 25 μm above the surface of $AL2$, and it is sealed with a side O-ring. Special care must be taken when machining the aluminum rings so that the rings fit snugly and the substrate surfaces have the proper relationship to the ring surfaces. Each individual substrate must be measured precisely before the machining is begun, as substrate dimensions are not generally held to close tolerances by the manufacturers.

All O-rings are 5.08-cm diam and 3.2-mm thick Viton, a material which is tolerant of the dichloroethane dye solvent. The substrate $S2$ was a 30-ft. wedge, and both surfaces of $S2$ were antireflection coated at 1064 nm. Before mounting, opposite edges of the inner surface of $S2$ were beveled off at 45° using a carborundum cutting wheel, while the substrate was held in a simple wooden jig. The optical coatings were protected during the grinding with masking tape. The bevels are each about 1.9 cm long on opposite sides of the surface.

When $S2$ is mounted in $AL2$ the beveled edges leave a small portion of the inner wall of $AL2$ exposed, and small holes (56 drill) were drilled at 25° to the normal into these two surfaces. The holes extend until they meet 0.32-cm holes drilled into the back surface of $AL2$. At the back surface these larger holes are further drilled out and tapped to accept $\frac{1}{16}$-in. × $\frac{1}{16}$-in. (1.5-mm × 1.5-mm) male stainless steel Swagelok connectors. The connectors couple to 18-gauge Teflon tubing, which carries the dye solution to and from the circulating pump and reservoir.

Thin Teflon spacers are placed between the substrates, and the two rings are drawn together by three 8-32 screws, which pass through clearance holes in $AL1$ and are threaded into $AL2$. The spacers are 50 μm thick and only block a small portion of the aperture. When the two rings are together, the middle O-ring contacts the edge of $S1$ and the face of $AL2$. A second O-ring also provides a backup seal on the circumference of $S1$. The beveled edges of $S2$ provide a small space for the dye to enter and exit the cell beneath the O-ring by way of the angled holes drilled in $AL2$. Another O-ring seals the circumference of $S2$, and thus a thin closed cell is formed between the substrates.

The o.d. of the aluminum rings is 3 in. (7.62 cm) so they fit directly into a 3-in. mirror mount.[6] Slight machining was done on the mount so that the Swagelok connectors would clear. The cell is firmly held by two set screws in the side of the mount. The compact cell protrudes only 2.41 cm beyond the outer surface of the mount.

The cell has proved to be completely reliable in operation: a laser mode-locking success rate of 80% is obtained,[5] and the mirror alignment is stable for a period of several days. Dye is pumped through the cell between laser shots, and no leakage of dye has been observed. Mode-locked pulse trains are produced with high reliability using this contacted dye cell-front reflector combination. The laser cavity in which this dye cell was tested is a generalized confocal configuration, which uses a spherical rear reflector and intracavity negative lens to compensate for thermal lensing.[4]

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Variational principle for scattering of light by dielectric particles

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Purcell and Pennypacker[1] (whose notations we follow) treat a dielectric particle as an aggregate of N polarizable elements (called atoms) mounted on a cubic lattice. The scattering problem reduces to solving a set of N simultaneous equations for dipole amplitudes $\mathbf{P}_i$, $i = 1,2,\ldots N$:

$$\frac{\mathbf{P}_i}{\alpha_i} - \sum_{\substack{j=1 \\ j\neq i}}^{N} \frac{\exp(ikr_{ij})}{r_{ij}^3}\left[k^2(\mathbf{r}_{ij}\times\mathbf{P}_j)\times\mathbf{r}_{ij} + \frac{(1-ikr_{ij})}{r_{ij}^2} \times (3\mathbf{P}_j\cdot\mathbf{r}_{ij}\mathbf{r}_{ij} - r_{ij}^2\,\mathbf{P}_j)\right] = \mathbf{E}_i, \quad (1)$$

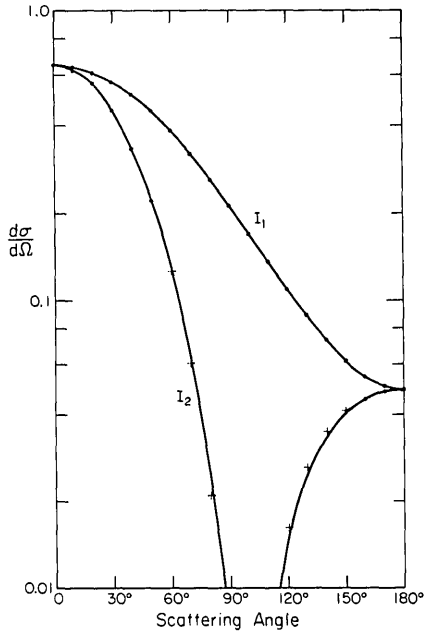

Fig. 1. Differential scattering cross section in units of k^{-2} at various scattering angles for a sphere composed of 4872 atoms. I_1 and I_2, respectively, refer to the components with electric field perpendicular and parallel to the scattering plane. The scattering parameter $2\pi a/\lambda$ equals 1.5, the bulk index of refraction is 1.33. The dots agree with Mie theory (solid lines) to better than 1%. The value for Q_{ext} is 0.321, compared with 0.322 given by Mie theory.

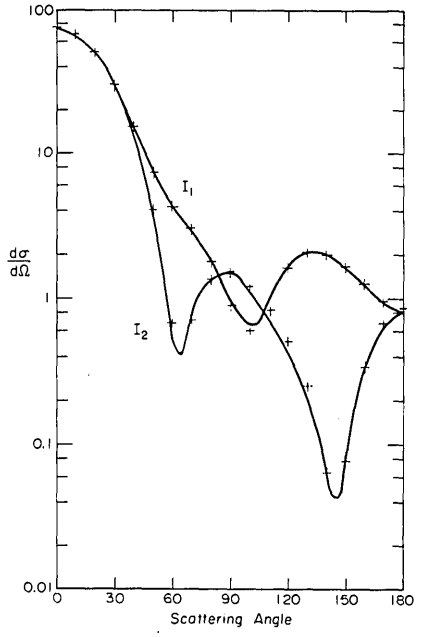

Fig. 2. Same as Fig. 1 for a sphere composed of 15,600 atoms, scattering parameter = 3, and bulk index of refraction = 1.7–0.1i. Q_{ext} and Q_{scat} from this computation are 3.80 and 2.74, respectively. The corresponding values given by Mie theory are 3.78 and 2.70.

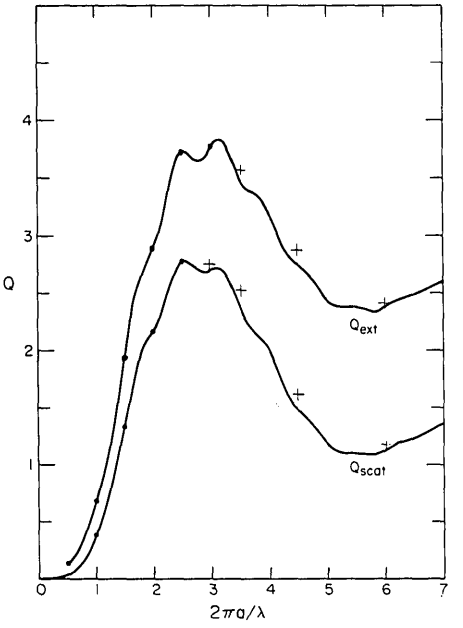

Fig. 3. Extinction and scattering efficiencies computed for a sphere composed of 4872 atoms. The last two points were calculated using a sphere composed of 15,600 atoms. The bulk index of refraction is 1.7–0.1i. The dots agree with Mie theory (solid lines) to better than 1%.

where $\mathbf{E}_i$ denotes the incident electric field at the ith atom whose polarizability is α_i. The theory has been applied to nonspherical and inhomogeneous objects with dimensions smaller than or comparable to the wavelength of incident light.[1–3] Further progress requires a better numerical approach. Let us write Eq. (1) in a compact form:

$$AP = E, \tag{2}$$

where $P = (\mathbf{P}_1, \mathbf{P}_2, \ldots \mathbf{P}_N)$, $E = (\mathbf{E}_1, \mathbf{E}_2, \ldots \mathbf{E}_N)$, and A is a $3N \times 3N$ symmetric matrix. It is of interest also to consider the scattering problem for a light beam incident from the opposite direction. In this case we replace the RHS of Eq. (1) by $\mathbf{E}_i^*$, the complex conjugate of $\mathbf{E}_i$. Let the solution be $P^\dagger = (\mathbf{P}_1^\dagger, \mathbf{P}_2^\dagger, \ldots \mathbf{P}_N^\dagger)$.

$$AP^\dagger = E^*. \tag{3}$$

Note that in Eq. (3) $E^* = (\mathbf{E}_1^*, \mathbf{E}_2^*, \ldots \mathbf{E}_N^*)$ is the complex conjugate of E, but $P^\dagger$ as defined by Eq. (3) is not the complex conjugate of P. Let us define a pair of functionals

$$H(P,P^\dagger) = \tfrac{1}{2}\left(\tfrac{1}{2}\langle P, AP\rangle - \langle E, P\rangle - \tfrac{1}{2}\langle P^\dagger, AP^\dagger\rangle + \langle E^*, P^\dagger\rangle\right), \tag{4}$$

$$K(P,P^\dagger) = \tfrac{1}{2}\left(\langle P^\dagger, AP\rangle - \langle E, P^\dagger\rangle - \langle E^*, P\rangle\right), \tag{5}$$

where P, $P^\dagger$ are now any two arbitrary $3N$-vectors, and the pointed brackets denote vector product. Applying an infinitesimal variation to Eqs. (4) and (5) and using the symmetry of A, we have

$$\begin{aligned}\delta H(P,P^\dagger) &= H(P+\delta P, P^\dagger + \delta P^\dagger) - H(P,P^\dagger)\\ &= \tfrac{1}{2}\left(\langle AP - E, \delta P\rangle - \langle AP^\dagger - E^*, \delta P^\dagger\rangle,\right.\\ &\qquad \left. + \tfrac{1}{2}\langle \delta P, A\delta P\rangle - \tfrac{1}{2}\langle \delta P^\dagger, A\delta P^\dagger\rangle\right),\end{aligned} \tag{6}$$

$$\begin{aligned}\delta K(P,P^\dagger) &= K(P+\delta P, P^\dagger + \delta P^\dagger) - K(P,P^\dagger)\\ &= \tfrac{1}{2}\left(\langle AP - E, \delta P^\dagger\rangle + \langle AP^\dagger - E^*, \delta P\rangle + \langle \delta P^\dagger, A\delta P\rangle\right).\end{aligned} \tag{7}$$

Therefore, $H(P,P^\dagger)$ and $K(P,P^\dagger)$ are stationary if and only if Eqs. (2) and (3) hold. The physics becomes clear in the long wavelength limit.

$$\lim_{k\to 0} H(P,P^\dagger) = \lim_{k\to 0} \mathrm{Im}K(P,P^\dagger) = 0, \tag{8}$$

$$\lim_{k\to 0} \mathrm{Re}K(P,P^\dagger) = \tfrac{1}{2}\sum_{i=1}^{N}\frac{\mathbf{P}_i\cdot\mathbf{P}_i}{\alpha_i} - \sum_{i=1}^{N}\mathbf{P}_i\cdot\mathbf{E}_i + \sum_{i=1}^{N}\frac{1}{\alpha_i}\sum_{j=i+1}^{N} \times\left(\frac{\mathbf{P}_i\cdot\mathbf{P}_j}{r_{ij}^3} - \frac{3\mathbf{r}_{ij}\cdot\mathbf{P}_i\,\mathbf{r}_{ij}\cdot\mathbf{P}_j}{r_{ij}^5}\right). \tag{9}$$

We recognize the RHS of Eq. (9) as the total energy (including self-energy) of a system of interacting dipoles. By a well known theorem in static electricity, this quantity is minimized[4,5]:

$$\lim_{k\to 0} \mathrm{Re}K(P,P^\dagger) = \text{minimum}. \tag{10}$$

Hence the variational principle

$$[H(P,P^\dagger),K(P,P^\dagger)] = \text{stationary}, \tag{11}$$

while equivalent to Eqs. (2) and (3) is a generalization of Eq. (10). The variational principle also unveils a subtle connection between energy and cross section:

$$\lim_{\delta K\to 0} \mathrm{Re}K(P,P^\dagger) = -\tfrac{1}{2}\sum_{i=1}^{N}\mathrm{Re}\mathbf{P}_i\cdot\mathbf{E}_i^* = \text{energy}, \tag{12}$$

$$\lim_{\delta K\to 0} \mathrm{Im}K(P,P^\dagger) = -\tfrac{1}{2}E_0\sum_{i=1}^{N}\cos kx_i P_{zi}'' + \sin kx_i P_{zi}' = \text{const}\times\sigma_{\text{ext}}, \tag{13}$$

where in Eq. (13) P_{zi}' and P_{zi}'' are real and imaginary parts of P_{zi}, and we use $\mathbf{E}_i = \mathbf{E}_0 1_z \exp(-ikx_i)$ and the optical theorem to identify the extinction cross section.[1] Equation (13) provides a stationary expression for evaluating σ_{ext}. The constant in front of σ_{ext} equals $-E_0^2/8\pi k$.

The existence of $H(P,P^\dagger)$ gives us the method of the steepest descent to solve Eqs. (2) and (3) separately. The following algorithm is adapted from Faddeev and Faddeeva.[6] Let $x_1, x_2, \ldots x_{i-1}$ be successive approximate solutions to Eq. (2). Then

$$x_i = x_{i-1} + \lambda_i s_{i-1}, \tag{14}$$

$$r_i = E - Ax_i, \tag{15}$$

$$s_i = r_i - \mu_i s_{i-1}, \tag{16}$$

where parameters λ_i and μ_i are chosen so that $\delta H(x_i,x_i^\dagger) = 0$, and s_i is conjugate to all previous s_j. One choice is

$$\lambda_i = \langle s_{i-1},r_{i-1}\rangle/\langle s_{i-1},As_{i-1}\rangle. \tag{17}$$

$$\mu_i = \langle r_i,As_{i-1}\rangle/\langle s_{i-1},As_{i-1}\rangle. \tag{18}$$

This is an exact iterative scheme and guarantees an exact solution (up to round-off errors) to Eq. (2) in $3N$ steps. We can similarly solve Eq. (3), but for particles with geometric symmetry $P^\dagger$ is simply related to P. Having obtained P, we can calculate the far field by summing over all the dipole amplitudes with appropriate phases. Results on scattering of light by dielectric spheres are shown in Figs. 1, 2, and 3. Comparison with Mie theory[7,8] and previous work[1] shows that we have indeed improved the numerical aspects of this simple and versatile theory.

In conclusion, we exploit two fundamental properties of the dipole interaction matrix A (symmetry with respect to reversal of incident beam direction and interchange of any pair of dipoles) to construct two functionals. The connection with the minimum energy theorem and cross section suggests that our numerical algorithm will search for a physical quantity, and hence it would be efficient and stable. In this Letter we have made the crudest use of the variational principle. The practical limit for the current version of our numerical scheme is a dipole array (with fourfold symmetry) of order $N \sim 10{,}000$ atoms. Since the phase difference between neighboring atoms should be $\lesssim 1/3$ rad for reliable results, this limits the scattering parameter $2\pi a/\lambda$ to $\lesssim 10$. The storage required is $\sim 20N$ real numbers, and the time is $\sim 1\times 10^{-6}N^2$ min on a CDC 7600. Note that the main advantage with the current method is that we do not need to store the A matrix with order N^2 elements.

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