of the computer code DENSJT.
[14]J. N. Ginocchio and M. M. Yen, Nucl. Phys. A239, 365 (1975); G. D. Lougheed and S. S. M. Wong, Nucl. Phys. A243, 215 (1975), and references therein.
[15]K. F. Ratcliff, Phys. Rev. C 3, 117 (1971).
[16]F. S. Chang and A. Zucker, Nucl. Phys. A198, 417 (1972).
[17]J. N. Ginocchio, Phys. Rev. C 8, 135 (1973), and Phys. Rev. Lett. 31, 1260 (1973); S. A. Ayik and J. N. Ginocchio, Nucl. Phys. A221, 285 (1974).
[18]S. A. Ayik and J. N. Ginocchio, Nucl. Phys. A234, 13 (1974); S. A. Ayik, Ph.D. thesis, Yale University, 1974 (unpublished).
[19]G. D. Lougheed and S. S. M. Wong, Nucl. Phys. A246, 29 (1975); J. B. French, Rev. Mex. Fis. 23, 189 (1974).
[20]Computer codes which will give more accurate values for the operator moments are being developed by a University of Rochester team and separately by one of the authors.
[21]F. S. Chang, J. B. French, and T. H. Thio, Ann. Phys. (N.Y.) 66, 137 (1971).

Observation of Resonances in the Radiation Pressure on Dielectric Spheres

A. Ashkin and J. M. Dziedzic
Bell Telephone Laboratories, Holmdel, New Jersey 07733
(Received 20 April 1977)

We report an experimental check of the Mie-Debye theory for the variation of radiation pressure on dielectric spheres with wavelength and size using optical-levitation techniques. Sharp resonances are observed which are shown to be related to dielectric surface waves. They permit particle-size measurement to a precision of 1 part in 10^5 to 10^6.

We report the first observation of the variation of the radiation-pressure force on transparent dielectric spheres with wavelength and size. We use a technique based on optical levitation which we call force spectroscopy. The measured force shows a regular series of sharp optical resonances which are in excellent qualitative agreement with the limited force data presently available from Mie-Debye theory. This resonant behavior can provide the most precise check on Mie scattering theory and also a way of measuring sizes of spheres to an accuracy exceeding that of present far-field scattering techniques by at least two orders of magnitude. These resonances are thought to be due to dielectric surface waves. This view is strongly supported by the observation of the scattered-light distribution in the near field. Both the resonant coupling of light striking the sphere edge and its subsequent isotropic tangential scattering are seen. These measurements should stimulate more precise calculation of radiation pressure and of the scattered-light distributions in the near field. Resonant effects are also of interest for the interpretation of particle scattering processes using optical models.

The Mie theory[1] for scattering of light by a sphere large compared to a wavelength is the best understood and most carefully checked example of scattering of waves by a particle. It is the basis of a vast theoretical and experimental literature and is widely used for particle-size measurement.[2] Using Mie theory, Debye[3] calculated expressions for the radiation-pressure force on a sphere as a function of the size parameter $X = 2\pi a/\lambda$, where a is the radius and λ the wavelength. Recently Irvine[4] made the first computer evaluation of the radiation pressure and extended computations of the total scattering cross section to large values of X using relatively high resolution (up to $\Delta X/X = 10^{-4}$). At low X, calculations of the total scattering cross section for low-loss spheres show the well-known "ripple structure"[5] as X is varied. Ripples are experimentally observed in measurements of the total scattering cross section for small x,[6] in far-field radar backscatter,[7] and in 90° scattering.[8] They are attributed to dielectric surface waves as originally proposed by Van de Hulst.[5] Irvine[4] shows that the ripple structure is larger on the radiation pressure than on the total scattering cross section and that at large X and high index n it sharpens dramatically and eventually becomes an unresolved sequence of resonances. Unfortunately he did not increase the resolution of his calculation further, since recent advances in the study of radiation pressure have now made possible the observation of Mie-Debye resonances with a resolution exceeding these existing calculations.

Indeed, with focused laser beams of modest powers one can use radiation-pressure forces to

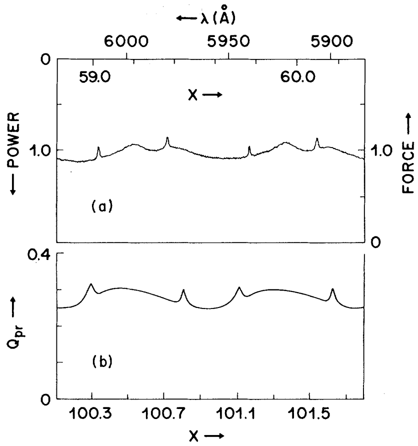

FIG. 1. (a) Measured power for levitation and approximate radiation-pressure force vs λ and X for a drop of $n=1.40$. (b) Theoretical force Q_{pr} vs X, $n=1.33$.

stably trap, levitate, and manipulate micron-sized dielectric spheres.[9] Particles levitated against the forces of gravity act as sensitive probes for various applied forces,[10] e.g., electrical, gravitational, and optical. Recently an electronic feedback scheme[11] was devised to lock a levitated particle's height to an external reference by controlling the laser power. The laser power needed to hold the height constant is a continuous measure of any variations of force on the particle. Thus to measure the wavelength variation of the radiation-pressure force we use the feedback stabilizer with a tunable dye laser as the levitating laser. We then simply record the light power needed to hold the sphere at fixed height as λ is continuously tuned by a motor-driven birefringent plate rotating inside the dye-laser cavity. We call this technique of force measurement radiation-pressure–force spectroscopy. For most experiments we used liquid drops as test spheres because of their remarkable perfection. Drops were made from highly transparent, low-vapor-pressure silicone oil[10] having $n = 1.40$ and somewhat more volatile, index-matching oils with indices up to 1.53. To levitate drops in the size range of 4 to 30 μm required dye-laser powers in the range of 1 to 400 mW. Since the theory is calculated for plane waves, particles were held at a height in the beam where the intensity variation of the Gaussian beam across the particle was very small.

Figure 1(a) is a tracing from the chart recording of the levitating power vs λ for an 11.3-μm-diam silicone oil drop, shown upside down (with zero power on the top). Since the radiation-pressure force per unit power is proportional to the inverse of the levitating power this view gives a good approximate representation of the force variation vs λ for comparison with the theoretical computations of Irvine. The X scale is derived from the λ scale using an approximate value of a (± 5%) as measured with a microscope. Figure 1(b) shows Irvine's theoretical force per unit power, Q_{pr}, versus X for $n = 1.33$ calculated with a resolution $\Delta X/X \cong 10^{-4}$. The experimental resolution $\Delta\lambda/\lambda$ is determined by the dye-laser linewidth of $\frac{1}{4}$ Å. Thus $\Delta\lambda/\lambda \cong 4\times10^{-5} = \Delta X/X$. Although the data were taken for $n \cong 1.40$ and $X \cong 60$, the resemblance to the theory is striking. We find much sharper resonances and more structure to the force variation for other size ranges of silicone drops (i.e., different X) and for drops of higher index of refraction. Figure 2 shows a section of one such curve for a slightly volatile index oil of $n = 1.47$. Since a scan over the laser tuning range takes $\sim\frac{1}{2}$ h and it takes minutes to change drops we can easily map out the variation of force with X and n. A range of X from ~10 to 200 is readily accessible. We believe the measured force to be highly accurate. The response of the laser-power monitor is constant with λ. Other than weak interference ripples from optics in the beam path there are no artifacts of the tunable laser in the data. Since the force depends solely on X for a given n we can, as a check, obtain identical force curves, appropriately shifted in λ, for different drop

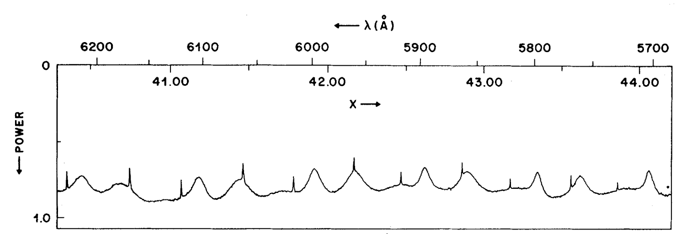

FIG. 2. Measured power for levitation vs λ and X for an approximately 8.0-μm-diam drop of $n=1.47$. The diameter decreased ~0.75% by evaporation as the wavelength was increased during the scan.

sizes. We can also get force-vs-X data using a fixed λ and a continuously varying radius a by measuring the levitating power as drops of the more volatile index oils slowly evaporate. These data are completely free of interference ripples from the optics. With evaporating drops we can rerun parts of the force curve by decreasing λ and letting X repeat itself as a decreases. With evaporation we can view the force variation over wide ranges of X and observe the evolution of successive groups of interleaved resonances of gradually diminishing sharpness as X decreases. This variation in sharpness is in part seen in Fig. 2. Irvine noted similar behavior in low-resolution calculations (see his Figs. 2 and 3).

Experiments were performed to understand the nature of the observed resonances. By simultaneously observing the force and the 90° scattered light in the near field from two orthogonal directions, the so-called parallel and perpendicular polarization directions, it is clear that the resonances fall into two interleaved sets depending on incident polarization. Next, if these resonances are due to Van de Hulst–type surface waves they should be excited by light striking the sphere edge tangentially. To check this we manipulated the drop with the feedback stabilizer away from its usual plane-wave position to a location at the focus of a strongly convergent beam where the beam completely misses the edges of the sphere and the sphere is supported solely by light passing through its axis.[11] In this case all sharp resonances disappear and we are left with a broad force variation of period $\Delta X \cong \pi/2n$ due to interference from the weak front- and back-surface reflections of the sphere. This is strong evidence for the surface-wave picture. The fact that edge illumination can cause strong surface-wave resonances at first sight violates one's intuition based on the high reflectivity of rays striking the sphere at nearly tangential incidence. However, viewing the sphere as a resonant, low-loss optical cavity, it is clear that the buildup of internally circulating light can modify the reflectivity sufficiently to give strong coupling into the sphere. Light thus coupled into the sphere should then be scattered isotropically, emerging tangentially. This suggests looking for direct visual evidence of surface-wave coupling and tangential scattering by near-field observations. Indeed tangential scatter is clearly seen in the backward direction. Not only do the edges of the sphere brighten at resonance but we can distinguish perpendicular and parallel resonances. When viewing in the forward direction [Fig. 3(a)] we can see the coupling process by using a crossed polarizer placed in front of the near-field image of the sphere. This blocks almost all transmission when off resonance [see Fig. 3(b)]. When tuned on resonance for one of the polarization components, the sphere by absorbing the resonant component of the light striking the edge and passing the orthogonal nonresonant polarization, in effect acts as a rotated polarizer inserted between two crossed polarizers and gives transmission. Since the effective angle of the sphere "polarizer" varies with azimuth and the absorption occurs only near the sphere edge, we expect four arcs of transmission with the maximum intensity at 45°. This is what is observed; see Figs. 3(c) and 3(d).

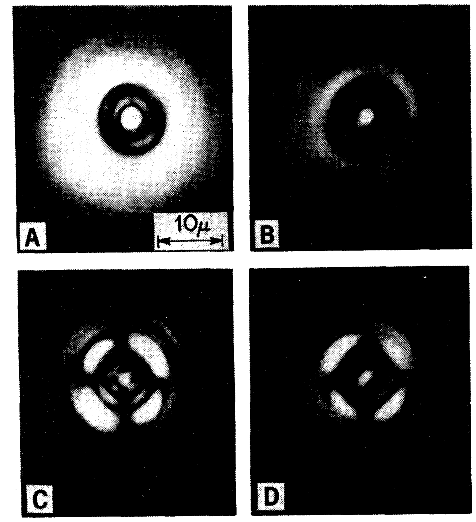

FIG. 3. Near-field views in the forward direction of an 11.3-μm-diam levitated drop: (a) no polarizer (exposure $E = \frac{1}{16}$ sec); (b) through a crossed polarizer, off resonance ($E = \frac{1}{4}$ sec); (c) through a crossed polarizer, on resonance for parallel polarization ($E = \frac{1}{8}$ sec); (d) through a crossed polarizer on resonance for perpendicular polarization ($E = \frac{1}{8}$ sec).

Consider the accuracy of size measurement and the related question of the accuracy of Mie-Debye theory. Assuming that we had a high-resolution curve of theoretical force vs X for the given n, we could then determine a by identifying each of a sequence of measured resonances with its corresponding X value. This comparison is unambiguous because of the distinctiveness of the shape variations of the force curve (see Fig. 2). From the theoretical X for each resonance and the measured λ we get a by using $a = X\lambda/2\pi$. The accuracy of each radius determination, $\delta a/a$, is equal to $\delta\lambda/\lambda$, the accuracy of the wavelength determination. Since we have observed resonant widths of $\sim\frac{1}{4}$ Å and can determine the resonance peak to $\delta\lambda \cong \frac{1}{5}$ of full width, we expect an accuracy $\delta a/a \cong (\frac{1}{20}\ \text{Å})/(6000\ \text{Å}) \cong 1{:}10^5$. The extent to which a values from different resonances are the same tests the internal consistency of Mie-Debye theory. In the absence of theoretical curves we determined the ratio of radii of two silicone drops roughly 13.1 μm in diameter by comparing a sequence of ten sharp resonances. These data give the same ratio 1.007 22 for all ten resonances to a rms accuracy of $\pm 0.000\,03$. The precision of 3 parts in 10^5 can be improved. It still shows the high accuracy and internal consistency of the resonance method. Even higher accuracies are possible in measuring size changes of a single particle. By tuning to a point of steep slope on the side of a sharp resonance we have easily detected changes of $\sim 10\%$ of the resonance height as a volatile drop evaporated. This corresponds to a change $\delta X \cong \frac{1}{10}$ of the resonance width which gives a sensitivity $\delta a/a = \delta X/X$ of 1 part in 5×10^5. If $a = 5$ μm, then $\delta a \cong 0.1$ Å. Since δa is considerably less than a monolayer this is interpreted as an average radius change. For comparison the accuracy of standard Mie-theory radius measurements[12] based on the far-field angular distribution of scattered light is only $\sim 1\%$ using Mie fringe-counting techniques. By comparing intensities with theoretical distributions this can be improved to $\sim 1{:}10^3$.

The high precision of force spectroscopy is useful for measurement of evaporation and condensation; study of the deposition of monolayers; optical detection of drop oscillations and minute drop distortions; and precise determination of electronic charge in a Millikan experiment. At higher values of n we expect even narrower resonances. Eventually drop distortion due to the radiation pressure itself should broaden the resonances. Perhaps we can then use highly precise glass spheres. An understanding of Mie-Debye resonances is clearly necessary for accurate measurement of radiation pressure and for proper formulation of optical analogs[13] of molecular, nuclear, and atomic scattering.

In conclusion we believe that radiation-pressure–force spectroscopy can provide the most exacting test of Mie theory. We have visually observed dielectric surface waves and demonstrated measurement of sphere sizes to an accuracy of 1 part in 10^5 to 10^6.

[1]G. Mie, Ann. Phys. (Leipzig) 25, 377 (1908).

[2]H. C. van de Hulst, *Light Scattering by Small Particles* (Wiley, New York, 1957); M. Kerker, *The Scattering of Light and Other Electromagnetic Radiation* (Academic, New York, 1969).

[3]P. Debye, Ann. Phys. (Leipzig) 30, 57 (1909).

[4]W. M. Irvine, J. Opt. Soc. Am. 55, 16 (1965).

[5]See van de Hulst, Ref. 2, Chap. 17; and Kerker, Ref. 2, Chap. 4.

[6]W. Heller, in *Electromagnetic Scattering,* edited by M. Kerker (MacMillan Co., New York, 1963); and Kerker, Ref. 2, Chap. 7.

[7]P. S. Ray and J. J. Stephens, Radio Sci. 9, 43 (1974).

[8]R. A. Dobbins and T. I. Eklund, Appl. Opt. 16, 281 (1977).

[9]A. Ashkin, Phys. Rev. Lett. 24, 156 (1970); A. Ashkin and J. M. Dziedzic, Appl. Phys. Lett. 19, 283 (1971), and Science 187, 1073 (1975).

[10]A. Ashkin and J. M. Dziedzic, Phys. Rev. Lett. 36, 267 (1976), and Appl. Phys. Lett. 28, 333 (1976).

[11]A. Ashkin and J. M. Dziedzic, Appl. Phys. Lett. 30, 202 (1977).

[12]H. H. Blau, Jr., D. J. McCleese, and D. Watson, Appl. Opt. 9, 2522 (1970).

[13]V. Khare and H. M. Nussenzveig, Phys. Rev. Lett. 33, 976 (1974); U. Buck, Rev. Mod. Phys. 46, 369 (1974); H. C. Bryant and N. Jarmie, Ann. Phys. (N.Y.) 47, 127 (1968).

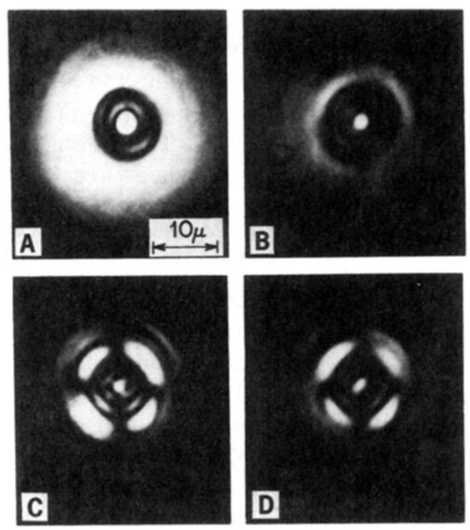

FIG. 3. Near-field views in the forward direction of an 11.3-μm-diam levitated drop: (a) no polarizer (exposure $E = \frac{1}{16}$ sec); (b) through a crossed polarizer, off resonance ($E = \frac{1}{4}$ sec); (c) through a crossed polarizer, on resonance for parallel polarization ($E = \frac{1}{8}$ sec); (d) through a crossed polarizer on resonance for perpendicular polarization ($E = \frac{1}{8}$ sec).