Applications in physics research

Laser developments are benefiting work in many other fields. Examples are in nonlinear spectroscopy, time and distance measurement, and Raman and Rayleigh scattering.

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Now firmly established as a versatile tool in many branches of science, the laser has taken little more than a decade to develop from a research project itself into an instrument for the support of other research. The number of laser applications in industry, technology and pure science is now so great that no one article could possibly cover them all. Here I will be concentrating on scientific applications only, but even so I must adopt a limited scope. The choice of topics has been restricted by posing the question: Of the many scientific investigations conducted with help from lasers, which ones are feasible only because lasers are available? Applications that satisfy this criterion should be the ones that best emphasize the unique capabilities of the laser as a tool of science.

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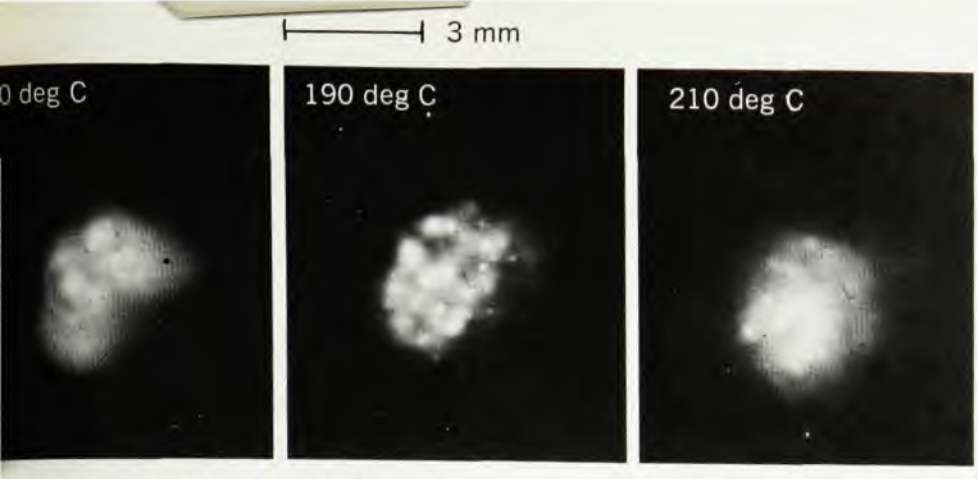
Most of my examples are taken from the fields of nonlinear optics and nonlinear spectroscopy, time and distance measurement, and light scattering. Fuller details than I can give here will be found in the references, which include general reviews as well as articles on specific methods and results mentioned in the text. An overview of the whole field of lasers and their applications will be found in four books, listed in reference 1, that I recommend particularly.

Nonlinear optics

We can characterize the phenomena of nonlinear optics by optical susceptibilities dependent on light intensity. Although the vast majority of the effects can only be observed with laser beams of great brightness, it does not follow that nonlinear optical effects are small or weak. On the contrary, the phenomena are often striking both in their qualitative and in their quantitative aspects.

Nonlinear optics includes the following effects: harmonic generation (up to the fifth so far); three- and four-frequency parametric interactions; multiphoton absorption and emission; optical bleaching; stimulated light scattering (Raman, Brillouin, Rayleigh, Rayleigh wing, concentration);[2] self-focusing, self-defocusing, and self-phase-modulation;[3] photon echoes and self-induced transparency,[4,5] and optical breakdown.[6] In addition to these topics, which are clearly recognized as parts of nonlinear optics, there are others that properly belong here as well. The laser oscillator is itself a nonlinear device, and its operation and noise properties can be thoroughly understood only in terms of its nonlinear behavior.[7] There is also a growing field of nonlinear spectroscopy; this technique depends entirely on laser sources and has a resolution often far greater than may be obtained by linear techniques.[8]

The recognition that certain optical materials have large nonlinear susceptibilities required generalization of the well known Fresnel laws of reflection and refraction to include reflection and refraction at the boundary between



Self-focusing of light in potassium vapor. In these photographs of the output window of a heated potassium-vapor cell, Raman-shifted light at 7658 A is focused by the saturable dispersion of the 7665-A resonance line of potassium. The self-focusing length decreases with increasing temperature because the strength of the nonlinearity increases with the number density of potassium atoms. Figure 1

linear and nonlinear media. Maxwell's equations for the electromagnetic field are, of course, perfectly applicable to nonlinear as well as to linear effects. But considerable effort has gone into determining the proper form for writing down the nonlinear polarization, which incorporates specific nonlinear effects into the wave equation.[9] The various nonlinear susceptibilities represent new material parameters to be measured and correlated with the other optical constants.[10] The problem of calculating nonlinear susceptibilities (for example, for second-harmonic generation) from first principles is formidable, but some progress has been made, particularly for semiconducting materials.[11]

Most of the effects in the list above are now fairly well understood, both experimentally and theoretically. However, in several cases the understanding has come only after considerable effort. There are two reasons for this. First, it has sometimes happened that several nonlinear phenomena occur simultaneously in a given experiment, thus complicating the understanding of each (for example, self-focusing and stimulated scattering). And second, considerable time passed before we understood that initial and boundary conditions play different roles in nonlinear and in linear problems. A good example is the great improvement in experiments on harmonic generation, stimulated scattering, and self-focusing brought about by the use of single-mode laser pulses. Similarly, the stimulated Raman effect is better understood from experiments on amplifiers than on oscillators.[12]

For most of these nonlinear effects there is now agreement on the important physical mechanisms involved, as well as agreement on the nonlinear differential equations that govern the phenomena. But solving these equations for anything like a general range of initial and boundary conditions has proved very difficult. The standard response to this impasse is to use a computer for numerical solutions of particular cases. However, some analytical solutions to nonlinear optics problems have been found,[5,13] and they perhaps justify the hope that attention to these problems by experts in applied mathematics and analysis would pay rich dividends. One direction for future work in nonlinear optics, therefore, will be increasingly sophisticated mathematical approaches to the coupled, nonlinear, partial differential equations involved.

There is room for experimental progress as well. For example, the problem of self-focusing and self-trapping is the subject of considerable controversy at present. Do self-trapped filaments of light exist and, if so, under what experimental conditions? If not, what happens to the light when it leaves the self-focused region?[14]

An experiment that may help to answer these questions involves the self-focusing of light in potassium vapor[15] (see figure 1). The nonlinearity responsible for the focusing arises, crudely speaking, from the saturation of the normal dispersion associated with a resonance transition of potassium. On the high-frequency side of such a transition, the index of refraction increases with increasing intensity, leading to self-focusing. In contrast to other self-focusing mechanisms, however, the dynamic response of the two-level system is known analytically at all powers. Moreover, the size of the focused spot at its smallest is large enough that there is hope of describing the entire focal region with a scalar wave equation. Finally, no other nonlinear effects are observed to accompany the self-focusing.

Nonlinear optical spectroscopy

It is well known that the spectral linewidth of optical transitions in the infrared and visible regions is often orders of magnitude greater than the fundamental limit set by lifetime broadening. The width may be determined by instrumental effects or at best by the Doppler effect in gases and by inhomogeneous strain distributions in solids. These lineshapes obscure structure in the spectrum resulting from molecular rotation, isotope shift, hyperfine splitting, or the Zeeman and Stark effects. However, when an inhomogeneously broadened line is irradiated by monochromatic laser fields strong enough to produce an appreciable nonlinear response, the width of the resulting absorption or emission line is primarily determined by lifetime effects. That is, the frequency-dependent nonlinear polarization of a two- or three-level system may have structure that is a thousand-fold narrower than the inhomogeneous width. It is, therefore, possible to uncover details of the spectrum not evidenced by ordinary, linear spectroscopy.

For atomic and molecular gases, this nonlinear response is exploited in three related spectroscopic techniques: optical double resonance,[8] level-crossing, and Lamb-dip.[16] The first method involves two laser beams of frequency ν_1 and ν_2 travelling in the same direction through a gas sample. The beams excite two transitions, of frequency f_1 and f_2, that share a common upper or lower level. When either the lasers or the absorbers are tuned so that $\nu_1 - \nu_2 = f_1 - f_2$, a nonlinear resonance is observed in the transmitted power. This resonance occurs even for $f_1 - f_2 = 0$, in which case only a single travelling wave is required. This is the zero-field level-crossing signal and is observed as the two transitions are tuned, by Stark or Zeeman effect, until they coincide within their homogeneous linewidths with the laser frequency. Last, if the travelling waves are replaced by a monochromatic standing wave, a narrow tuning dip, called the Lamb-dip, will occur in absorption (or emission) at the Doppler peak.

Examples of measurements that have been made by these nonlinear techniques include:

- precision determinations[17] of the electric dipole moments of both ground and excited vibrational states of CH_3F (see figure 2)
- g-values, radiative lifetimes, and hyperfine structure splitting of excited states of Xe[18]
- molecular rotational magnetic moments and relaxation rates[19] in both ground and excited vibrational states of CH_4.

In the optical double-resonance experiments on CH_3F at 9.5 microns the resolution was about 10^8, corresponding to a 100-kHz linewidth. The precision obtained in measuring the dipole moments was about one part in 2000, which compares quite favorably with the level of precision derived in microwave and molecular-beam resonance spectroscopy. Such measurements will, therefore, provide new information for testing *ab initio* calculations[20] of molecular

quantities. Finally, Stark spectroscopy in the infrared (both linear and nonlinear) can be used to give unique vibrational-rotational line assignments in systems where this has not been possible in the past.

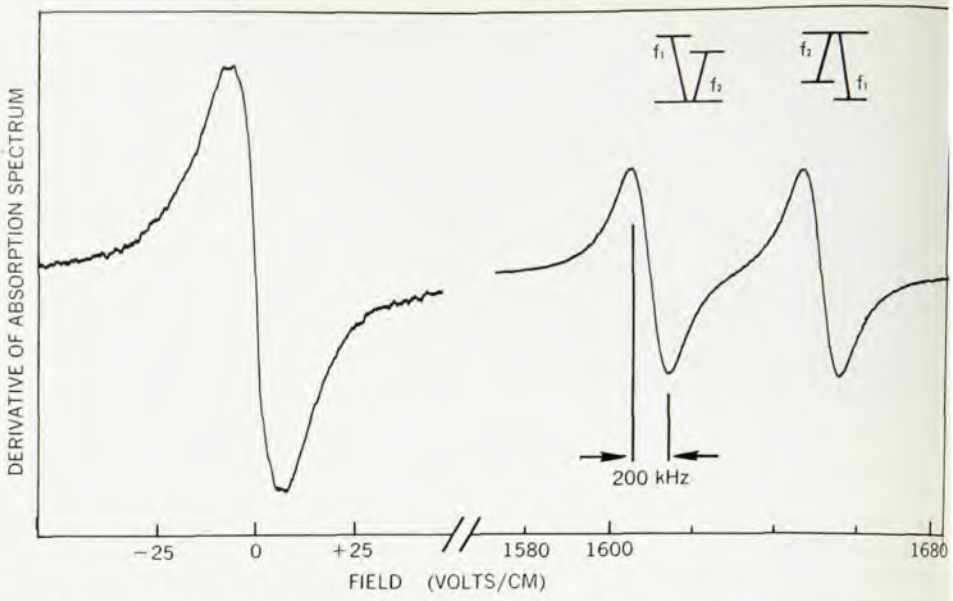
These ultrahigh resolution, nonlinear techniques have been applied primarily to gases in the infrared, where it is expected they will add a new dimension to atomic and molecular spectroscopy. They are also of great importance in the production of laser wavelength and frequency standards, which we will discuss next.

Time and distance

Any significant improvement in the precision with which either time or distance can be measured has always had important consequences in science, and the development of lasers as high-precision standards of wavelength and frequency is well under way in several countries, including the US and Japan. We should recall first that until very recently optical frequencies have not been measurable at all. Instead, wavelengths have been measured with an interferometer or grating and the frequencies calculated using the experimentally known value of the speed of light. The precision to which c is known, however, is lower than that with which precise wavelength measurements can be made. Now, however, we can *measure* laser frequencies as high as 6×10^{13} Hz by beating the laser first with other lasers and finally with a microwave source that can be stabilized by an atomic clock.[21]

With the ability to measure independently the wavelength and the frequency of a well stabilized laser we can significantly improve the determination of the velocity of light. This in turn makes it conceivable that the standards of length and time will be made interdependent via a defined value of c. There are also, of course, clear uses for accurate optical frequency measurements in connection with the high-resolution, nonlinear spectroscopic techniques I have described above. Two other examples of time and distance measurement by lasers are laser ranging of the moon and picosecond pulse measurements of very fast processes in molecules and condensed systems. In this section I will deal with each of these topics except the last, ultrafast pulses, which is to be the topic of a forthcoming article in *physics today* by Anthony J. DeMaria of United Aircraft Research Laboratories.

The problems of making an optical frequency standard fall roughly into two categories. The first is the production of an oscillator of the requisite stability and resetability; the second is the actual measurement of the frequency in terms of standards at lower frequencies—perhaps in terms of the cesium clock or the hydrogen maser.



Nonlinear infrared absorption spectrum of CH_3F transitions tuned into resonance by a Stark field. Excitation is by two 9.5-micron CO_2 lasers with a fixed frequency difference of 39.63 MHz. At the left is the zero-field level-crossing resonance. On the right are two optical double resonances for excited and ground vibrational levels respectively. The frequency difference $f_1 - f_2$ is equal to the laser frequency difference at resonance. These lines would normally be buried within a Doppler width of 66 Hz, but they are well resolved here and give precise electric dipole moments in the two states. Figure 2

The approach to the stability problem that currently appears most promising is to lock the He-Ne laser line at 3.39 microns to a very narrow absorption line of methane vapor—the $P(7)$ line of the ν_3 band.[22] The extremely narrow effective width of the methane line (about 150 kHz half width at half maximum) is obtained by utilizing the nonlinear response of the saturated absorption of the transition in the (relatively) high laser field. As we saw in the discussion on nonlinear spectroscopy, the nonlinear response may be much narrower than the Doppler width of the line. In this case it is limited only by collisions and by the time of flight of a molecule through the laser beam. This particular line in CH_4 has been found to have a very small Stark shift and only a small magnetic splitting in the earth's field. Moreover, the pressure shift of the line center is so small that so far it has been impossible to measure it. These features have made it possible to lock the laser to the methane-saturated absorber with a resetability of better than one part in 10^{11}. This is some 300 times better than the resetability associated with the present primary standard of length, the orange-red line of Kr^{86} at 6057 Å emitted from a lamp operated under carefully defined conditions. The output of the krypton lamp is, of course, spontaneous emission. The advantages of the stimulated-emission source arise because its linewidth is intrinsically narrow and its intensity is great enough to saturate an absorption band.

Experiments are being carried out to compare interferometrically the methane-stabilized He–Ne laser with the krypton standard.[23] Initially this comparison will serve to determine the absolute wavelength of the laser emission to the precision of the standard. That the laser is more precise than the standard is a situation which, in the end, can only be dealt with by fiat.

Measuring the frequency of the stabilized laser requires techniques very different from those used to measure its wavelength. The process is involved and requires great care. However, it now appears clear from the work of several groups that measurement of the frequency and wavelength of a stabilized laser can be carried out with sufficiently high accuracy to permit a significant increase in the precision with which c can be measured.

For a frequency measurement the laser must be compared with a standard whose frequency is more than a thousand times lower. Clearly the process of beating frequencies together must be carried out in a number of steps, and over the past few years there has been gradual progress in extending laser frequency measurement to higher and higher regions. At the moment the highest frequency measured is the 5.2-micron line of the CO laser.[24] It is the $P(13)$ line of the 7–6 vibrational band and has been found to have a frequency of 58 024 341 ± 55 MHz. (This measurement was not made under conditions where accuracy was the principle aim; it is quoted to indicate the progress that has been made in high-frequency measurement.)

To beat a laser source with another

frequency one must have a mixer that responds at frequencies as high as that of the laser. Remarkably, such mixers do exist. They consist of a metal-to-metal diode formed by contacting a pointed tungsten whisker to a polished nickel post.[25] The whisker, which may be several millimeters long and a few microns in diameter, has an etched point of about 500-Å radius. The resulting diode permits currents to flow at frequencies at least as high as 6×10^{13} Hz, and the upper limit of the diode's frequency response is not yet known. Nor is the precise mechanism by which the whisker diode operates yet known, but it is believed to involve electron tunneling in a metal-oxide-metal layer.

The steps by which the frequency measurement at 5.2 microns was made illustrate the new technique and its problems. The 5.2-micron laser beam was mixed in a whisker diode with a CO_2 laser beam at about 10.4 microns. The nonlinear response of the diode at 10.4 microns generated its second harmonic, which was close to the CO laser frequency. The difference between these two frequencies was a microwave frequency, which was mixed in the same diode with a known microwave signal. In this way the frequency of the 5.2-micron laser was measured with respect to that of the 10.4-micron CO_2 laser. The absolute frequency of the CO_2 laser was calculated from the accurately known rotational constants of CO_2 coupled with measured values of the CO_2 laser frequency at 10.6 microns.[26] The latter was measured by a mixing technique (using a similar diode) involving two water-vapor laser lines at 78 microns and 28 microns and a klystron of known frequency. Similarly, the frequencies of the water-vapor lines had been measured by still another experiment involving lower frequencies.

These high-frequency measuring techniques have been used to give accurate values of the frequency differences between different lines of a given laser. From these measurements one can obtain very precise values for the constants that give the energy levels of the laser molecule in terms of its rotational and vibrational quantum numbers. Frequency-mixing experiments have also been used to decide between conflicting measurements of the wavelengths of the water-vapor laser line at 28 microns.

Moon ranging

The wavelength and frequency measurements described above rely on the ability of lasers to operate with an extremely narrow spectrum. But lasers can also be made to operate with large absolute bandwidths, and in this regime they can produce intense pulses with durations from tens of nanoseconds down to a few picoseconds. The nanosecond-long pulses are being used to measure the range of the moon to an accuracy of a foot or so. The lunar-ranging experiment, which has been described in a number of recent articles,[27] is possible only because of the availability of pulsed lasers of high brightness and energy. The laser used at the MacDonald Observatory is essentially diffraction limited and emits about 5 joules in a pulse typically 4 nanoseconds long. Even so, the return signal from the retroreflector on the lunar surface is so weak that, on the average, one detected photoelectron is expected for each transmitted pulse. Clearly, no nonlaser source is conceivable for this experiment. Sophisticated timing and detection circuits, coupled with a narrow-band optical filter, make it possible by averaging over a number of pulses to measure the one-way distance to the reflector with an error of ± 15 cm.

The range to the moon is about 3.7×10^{10} cm, and therefore the range determination in a given measurement has an accuracy of one part in 2.5×10^9. Before these measurements the error in the predicted range for a given measurement was about one part in 2.5×10^6 ($\pm$ 1 microsec in a round-trip time of 2.5 sec).

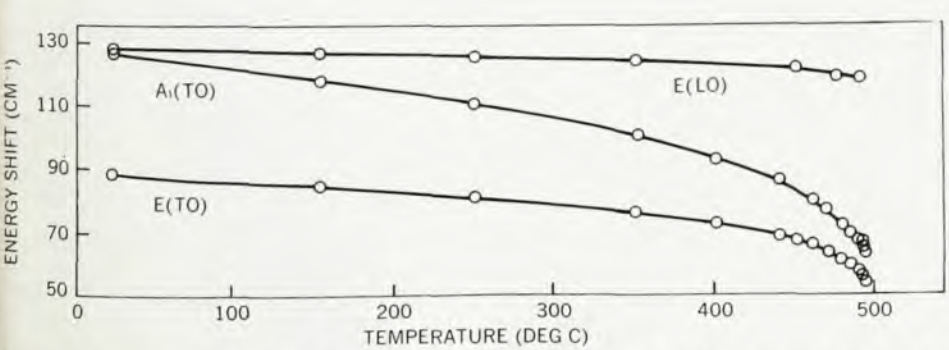
The improvement we expect in our knowledge of the lunar orbital parameters, however, is not so great as this factor of a thousand suggests. Nevertheless the improvement will be substantial; it is between a factor of ten and a hundred depending on the quantity involved. A number of the desired measurements can be made only if data are available from at least three terrestrial observing locations. Similarly, determination of the libration of the moon requires several reflectors on the moon (there are now three). Improved measurements can be made both of astronomical and of terrestrial quantities; the list includes the mean earth–moon distance, the eccentricity of the lunar orbit, the angular position of the moon with respect to various fixed points, the rotational period of the earth, the motion of the pole and the rates of continental drift. Accurate measurement of the lunar orbit is expected to shed light on the current controversy between scalar-tensor and pure tensor theories of gravitation. Each of these determinations will require an extended period of observations, lasting from one to eight years.

Laser systems are now available that emit a single pulse, with a duration of several picoseconds, which can be amplified to have an energy content of about a joule. The extent to which such pulses can be used to improve the accuracy of these lunar measurements is not yet clear. At present the range determination is limited by the properties of the photomultipliers used. There is also some question about the degree of stretching to which a pulse only a few millimeters long will be subjected on a round trip through the atmosphere. Nevertheless, it is not unreasonable to expect that picosecond pulses will ultimately find use in these scientific ranging experiments.

Light scattering

There is a 50-year tradition of extracting physical data from light-scattering experiments. However, it is hardly an overstatement to say that lasers have revolutionized all aspects of light scattering. Not only have lasers drastically shortened the time required for traditional measurements, but they have made feasible new experiments as well. The examples I shall mention here lie in the areas of Raman scattering in solids and Rayleigh scattering in liquids, and they represent only a few of the many experiments that could be cited.[28]

In vibrational Raman scattering one observes light that has been inelastically scattered by the optic modes of vibration of a crystal or by the normal modes of a molecule. The polarization properties of the scattered light are of great importance in making assignments of the observed scattering frequencies to the proper modes. Raman work used to be hampered by the low intensity of sources previously available, and hence both the irradiation and scattered beams usually subtended large solid



Temperature dependence of the soft modes in $PbTiO_3$ as the ferroelectric phase is heated towards the transition temperature. E and A_1 denote the symmetry of the transverse (TO) and longitudinal (LO) modes, A_1 being totally symmetric and E doubly degenerate. Figure 3

angles as seen from the scattering region. Under such conditions it was very difficult to make unambiguous measurements of the polarization properties of the scattered light. Most of the polarization measurements were not carried out at all. This is particularly unfortunate because the Raman-scattering tensor has six independent components and is thus richer by a factor two in selection-rule information than infrared spectroscopy.

With a laser source and a double monochromator the experimenter can exploit fully the Raman selection rules, and this makes possible the unambiguous assignment of many weak, Raman active modes.[29] For example, the Raman spectrum of α-quartz has been worked out only recently, using these laser techniques.[30]

Ionic lattice vibrations couple strongly to electromagnetic waves of the same frequency and form a mixed elastic–electromagnetic excitation known as a "polariton." In the region of strong coupling the polariton frequency depends strongly on its propagation vector (**k**-vector). Moreover, the distinctive Raman scattering from polaritons can be seen only as small-angle, near-forward scattering (where the **k**-vector of the polariton is small). Conventional Raman sources are not bright enough to permit the angular resolution required, but laser illumination has made polariton scattering observable and allowed a study of the polariton dispersion curve in GaP and ZnO.[31]

The study of phase transitions is currently of great interest, and is characterized for displacive transitions by concern with "soft modes." A soft mode is a normal mode of the lattice whose frequency approaches zero at the phase transition. To observe this process by Raman scattering requires a technique with high sensitivity to broad lines near the exciting line. Here again, the combination of a laser and a double monochromator permits measurements not otherwise feasible. For example, the paraelectric to ferroelectric transition in the ferroelectric $PbTiO_3$ has been studied by Raman scattering.[32] In the ferroelectric phase, two transverse optical modes are strongly temperature dependent near the transition. The lower frequency can be followed with considerable accuracy from its room-temperature value, 89 cm^{-1}, to 55 cm^{-1} at the transition temperature of 490 deg C. The damping of this mode diverges as T_c is approached from below, but its frequency may still be determined accurately (see figure 3).

Optic modes of vibration fall into two classes, the longitudinal (LO) and transverse (TO) modes, and in noncentrosymmetric (piezoelectric) crystals these modes are Raman active. Because of anisotropy, the frequencies of the modes depend on their propagation direction **k** with respect to the crystal axes. By "the" LO and TO modes one means those that propagate along the principal directions of the crystal. Normally, when single crystals are used in Raman experiments the crystal is oriented so that **k** is along a principal direction. It is possible, however, to obtain the same information about "the" modes when polycrystalline powders are used. This is useful, as it is often difficult to obtain even small single crystals of materials of interest. In the tetragonal case the curves that give the LO and TO frequencies as a function of the angle θ between **k** and the c-axis have horizontal tangents at 0° and 90°, corresponding to "the" LO and TO frequencies. The density of states, giving the number of crystallites in a powder contributing to the scattering at a given frequency, has a singularity whenever $d\omega/d\theta = 0$. As a result there is a large peak in the scattered Raman radiation corresponding to the LO and TO modes. This technique has recently been demonstrated in the $PbTi_{1-x}Zr_xO_3$ system, a material in which there are para-, antiferro-, and three ferroelectric phases.[33]

Because of the large frequency shifts that are studied by Raman scattering, it is not necessary to have a spectrometer whose resolving power is much greater than 10–20 000. There are, however, other processes in condensed systems (for example, diffusion and critical fluctuations) whose detection *by light scattering* (Rayleigh scattering) requires vastly higher resolving power, of the order of 10^{13} or 10^{14}. The techniques of homodyne and heterodyne spectroscopy and the closely related technique of photoelectron correlation and counting distributions are all capable of furnishing this formidable resolution. Each of these methods measures an intensity

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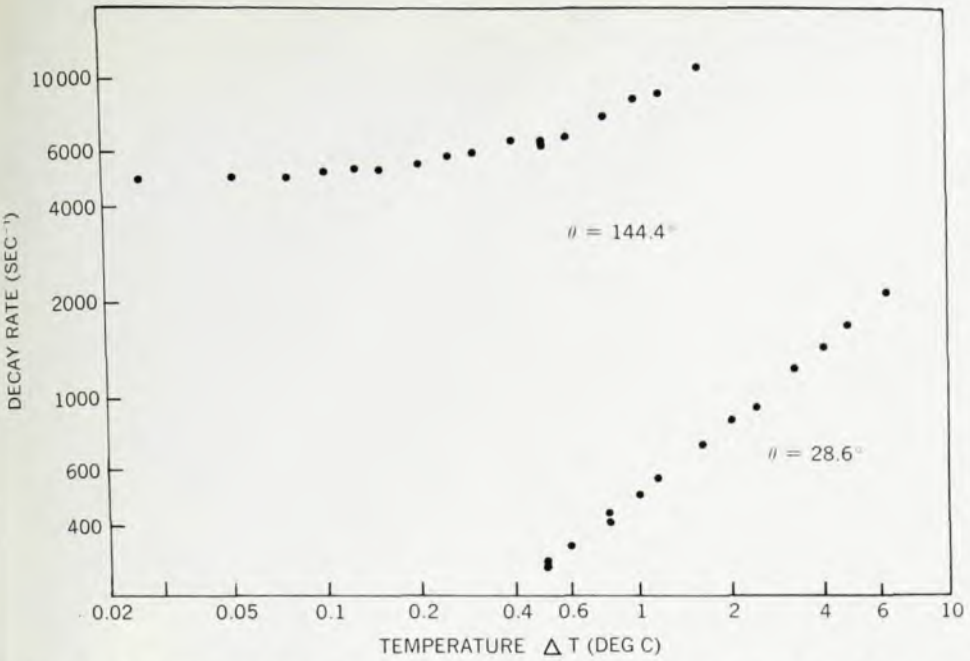

Decay rate of concentration fluctuations in a bromobenzene–water–acetone mixture, studied by light scattering at two angles θ. ΔT represents the temperature, $T - T_c$, above the critical point. From reference 35 (Bak). Figure 4

correlation function of the light scattered by fluctuations in the index of refraction due to some physical process of interest. A recent review[34] lists techniques and experiments in Rayleigh scattering, together with an extensive bibliography.

These light-scattering techniques have been applied to the study of critical phenomena. For example, detailed light-scattering studies have been made of the time-dependent density fluctuations near the gas–liquid critical point of such fluids as SF_6, CO_2 and Xe, and also of time-dependent concentration fluctuations near the critical mixing point of various binary and ternary liquid mixtures[34, 35] (see figure 4).

Great theoretical effort is being expended on the proper description of the temperature and spatial-frequency dependence of both the scattered intensity and the linewidth near a critical point.[36] Laser scattering techniques can be used to measure these variations in the hydrodynamic as well as the nonhydrodynamic regimes (that is, where the correlation length of the fluctuation is, respectively, less than or greater than the optical wavelength).

Light scattering from concentration fluctuations can also provide information on the hydrodynamic shape of macromolecules. Moreover, if there is independent knowledge of the molecular density and sedimentation rate, the scattering from concentration fluctuations can be analyzed to give the macromolecular diffusion constant.[37] This laser light-scattering method represents a giant step in the speed and accuracy with which diffusion-constant determinations can be made. The rate of agglomeration of macromolecules may also be measured by light scattering.

There are, of course, many more novel experiments and interesting techniques than the few I have been able to describe here. The number is growing all the time as a result of the continuing effort going into new and improved lasers. Laser sources will be increasingly important for linear spectroscopy in the infrared and far-infrared regions. At the other end of the spectrum vacuum-ultraviolet lasers[38] will be applied to photochemistry and photodissociation studies. And, only now beginning to be studied seriously, there are the possible uses of soft x-ray lasers.

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