The Long and Short of Lasers (I): The Far Infrared

In the years since the invention of the laser, researchers have extensively exploited its ability to generate coherent light in the near infrared and visible regions of the optical spectrum in both continuous wave (cw) and pulsed modes. In this wavelength range, there are, for example, pulsed lasers that have high peak power (10^{12} watts) or that have ultrashort pulse lengths (less than 10^{-12} second) and cw lasers that produce a megawatt of optical power. Many lasers have a high degree of spatial coherence (necessary for holography). Some lasers are highly monochromatic (emit over a frequency range as narrow as 1 kilohertz), while others can be tuned over a range of frequencies. Still others are miniaturized (for solid state circuits). Recently, however, laser scientists have increasingly been turning their attention toward the far infrared (roughly 50 micrometer to 1 millimeter wavelength) and the vacuum ultraviolet (about 200 to 10 nanometers wavelength).

The far infrared (often also called the submillimeter wave region) has for years been the stepchild of optical spectroscopists because of a dearth of bright light sources, efficient optical materials, and sensitive detectors with fast responses. The use of Fourier transform spectroscopy and the development of a variety of improved detectors in recent years has helped some. But the arrival of far infrared lasers and coherent sources derived from lasers is having, in the words of one enthusiast, a revolutionizing effect on submillimeter wave spectroscopy, permitting measurements of the properties of solids, liquids, and gases which were never before possible. Other applications projected include the study of the properties of plasmas created in controlled fusion experiments (plasma diagnostics), sensitive detectors (double heterodyne detectors) for astronomers and communications, and improved air pollution detection systems. Perhaps one indicator of the interest stimulated by the evolution of far infrared lasers is that, at the American Optical Society meeting held in Washington, D.C., in April, the first steps were taken toward the formation of a technical group on submillimeter waves.

Far infrared lasers appeared in the middle 1960's with one development being a water vapor laser that emits

gaw 1062 light at 78 micrometers. However, the electric discharge that excited (pumped) the laser medium (in which a voltage pulse is applied along the length of the tube containing the vapor) unfortunately had a tendency to break bonds in many of the other molecular gases that were tried, and in effect destroyed the laser medium. As a result, only a few far infrared lasers of this sort had been identified until T. Y. Chang and T. J. Bridges at Bell Laboratories, Holmdel, New Jersey, came upon the idea of optically exciting the molecules in the gas (which they reported in 1970) during experiments with methyl fluoride. Upon noting the strong absorption of carbon dioxide laser light by the methyl fluoride gas, the Bell scientists realized the potential for creating laser action in methyl fluoride. When absorption of the carbon dioxide light excites the methyl fluoride from one vibrational state to a given rotational level in a higher vibrational state, light is given off at 496 micrometers (and several other wavelengths) as a result of transitions between rotational levels in the higher vibrational state. At present, 282 laser lines in 18 different gases have been identified in the far infrared at a number of laboratories, thus providing a rich variety of both cw and pulsed laser sources for molecular spectroscopy.

Compact Gas Laser

Efforts are now under way to improve the usefulness of optically pumped molecular gas lasers. Workers at the Aerospace Corporation, El Segundo, California, are using metal waveguides (similar to the metal waveguides long used in microwave systems) with mirrors at each end to form the optical cavity of the laser. (Normally, the optical cavity of a gas laser consists only of the two mirrors at the ends of a long glass tube containing the gas.) This would permit a more compact laser than heretofore in use and possibly also an increased portability and lower cost. Similar work has been carried out at Osaka University in Japan.

H. R. Fetterman and C. D. Parker of the Massachusetts Institute of Technology, Lincoln Laboratory, Lexington, and H. R. Schlossberg of the Air Force Cambridge Research Laboratories, Bedford, Massachusetts, have

demonstrated that the need for close matching of the pump and the molecular absorption frequencies can be largely overcome by the application of a high electric field to the molecular gas. This technique of shifting the vibrational absorption into coincidence with the carbon dioxide pump is a general method which greatly increases the number of cw lines available.

Investigators at the University of Illinois, Urbana, and the Massachusetts Institute of Technology (MIT), Cambridge, are working on high power optically pumped molecular gas lasers. The Illinois group (T. K. Plant, L. Newman, T. A. DeTemple, E. J. Danielewicz, and P. D. Coleman) obtained very high pulse peak powers (100 kilowatts) from methyl fluoride pumped by a 10-megawatt pulsed carbon dioxide laser in what Coleman called abnormal spontaneous radiation (Dicke superradiance). In this mode of operation, the mirrors of the laser optical cavity are removed, but the high pumping power may cause the molecular dipoles to, in effect, line up and emit light coherently as one giant dipole. At MIT's Francis Bitter National Magnet Laboratory, Fielding Brown of Williams College, Williamstown, Massachusetts, obtained 500 watts (peak) from a methyl fluoride laser (pumped by a 20-megawatt pulsed carbon dioxide laser) in which a cylindrical lens was used to focus the pumping light into a line along the axis of the tube containing the gas [transverse optical pumping (TOP)]. This method of optical excitation permits obtaining a higher power from a normal laser (one with mirrors) than can be obtained with the excitation through the end of the tube. as is the usual custom. A high quality laser pulse (line shape) is also obtainable from these TOP lasers, according to Brown, who is now working on ways to step up the power output achievable.

Despite the admitted virtues of optically pumped molecular gas lasers, spectroscopists are nevertheless limited by the fact that the far infrared laser emissions come only at the discrete wavelengths dictated by the energy level structures of the molecules, and these are not evenly spaced throughout the submillimeter region. A tunable laser one that could be made to emit any desired wavelength—would impart to the experimenter a much greater flexibility than he now has. There are a variety of schemes for obtaining tunable coherent light in the far infrared, but none are true submillimeter lasers. Instead, the light from visible or near infrared lasers is converted by one of a number of nonlinear optical processes into far infrared radiation. Since the frequency conversion process preserves the properties of the light (such as its coherence), the resulting submillimeter light sources are described as coherent sources.

Whenever a light wave passes through a medium, a polarization is created by the electric field of the light, which has a magnitude proportional to that of the field and has the same frequency. But, because of the intense light possible with lasers, the polarization in some crystalline materials is no longer simply linear, but has components proportional to some higher power of the field. For example, when two laser beams of different frequencies are incident upon such a medium, a polarization proportional to the square of the field will oscillate with a frequency equal to the sum or the difference of the frequencies of the two incoming waves, and will reradiate light with this new sum or difference frequency. (It is the difference frequency radiation which is of interest in the far infrared region.) As the original light waves and the reradiated wave pass through the medium, it is important that they all travel at the correct velocity to "keep in step," so that the intensity of the generated wave can build up. This condition, called phase matching, is determined by the index of refraction of the medium. Since, in general, the index of refraction is different for different wavelengths (dispersion), a number of tricks are employed to achieve phase matching, including the use of birefringent materials and noncollinear beams (the two waves to be mixed do not travel parallel to one another). Much of the early work of this type was done by C. K. N. Patel and others at Bell Laboratories.

Benjamin Lax and R. L. Aggarwal of the National Magnet Laboratory and Fetterman and P. Tannenwald of the Lincoln Laboratory are now working on what they describe as a quasitunable far infrared coherent source. Others in the group are N. Lee, G. Favrot, and B. Clifton. In their experiments, light beams from two carbon dioxide lasers at slightly different wavelengths are incident upon a nonlinear crystal, such as gallium arsenide or 7 JUNE 1974

cadmium telluride, cooled to 80°K. In this case, the nonlinear process produces far infrared light with a frequency equal to the difference between the frequencies of the carbon dioxide lasers. Phase matching is achieved by varying the angle between the two incident light beams (noncollinear phase matching). Tuning is possible because there are about 80 laser lines between 9.2 and 10.8 micrometers available from each carbon dioxide laser (by adjusting a grating in the optical cavity), giving a total of about 3200 possible difference frequencies that are approximately equally spaced in frequency in the wavelength range 70 micrometers to 2 millimeters.

According to Aggarwal, the MIT scheme at present produces about 10^{-7} watt in a cw mode of operation (with 25 watts pumping power from each carbon dioxide laser), and several hundred milliwatts peak power in a pulsed mode—200 kilowatts (peak) in each pumping laser. Refinements, such as an improved nonlinear mixing configuration, are being studied now in the hope of raising the efficiency of the far infrared generation process.

Phase Matching in a Waveguide

D. Thompson and Coleman at the University of Illinois are also working on a quasi-tunable far infrared source that operates similarly to the MIT source, except that the nonlinear mixing process is achieved through the use of a dielectric waveguide made of gallium arsenide whose thickness is comparable to the wavelength of the light generated. Collinear phase matching (parallel incident beams) is possible because the dispersion of the waveguide itself plays a role analogous to that of the angle between incident laser beams in the MIT experiments. About 20 percent of the theoretically expected power output has been obtained.

At the University of California, Berkeley, K. H. Yang, J. R. Morris, P. L. Richards, and Y. R. Shen have developed a continuously tunable far infrared source between 52 and 500 micrometers. The Berkeley group begins with a ruby pulse laser, 30 megawatts (peak), which is used to pump a tunable dye laser, 600 kilowatts (peak), such as 3,3'-diethylthiatricarbocyanine iodide in dimethylsulfoxide. Dye lasers emit light over a broad range of wavelengths and are tuned to a particular wavelength through the use of gratings in the laser optical cavity. A special prism (Glan-Thomson prism) splits the light from the dye into two

separate beams with perpendicular polarizations. The orthogonally polarized beams then are tuned independently to different wavelengths by the gratings. Finally, a lithium niobate (or other) nonlinear crystal at room temperature produces far infrared radiation as the difference frequency of the two dye laser beams. A noncollinear phase-matching configuration (one of three configurations tried) resulted in the highest far infrared power output, up to 200 milliwatts (peak). Continuous wave operation is difficult because of the presently limited cw power of dye lasers in the visible region of the spectrum.

At Bell Laboratories, a third approach for generating continuously tunable far infrared light is being investigated by Van Tran Nguyen and Bridges. In the Bell experiments, light from a carbon dioxide laser is incident upon an indium antimonide crystal cooled to 15°K and in a magnetic field of about 5 teslas. In a magnetic field, the free electrons in a semiconductor fall into two groups depending on which of two spin orientations they have (the energy splitting is proportional to the magnetic field strength). Some of the incident light is converted to light of lower frequency by Raman scattering (due to excitation of electrons from the lower energy spin state to the higher one—that is, some of the energy of the light is lost in flipping the electrons from one spin to the other), so that two slightly different frequencies of light emerge. In a second indium antimonide crystal, also cooled and in a magnetic field, far infrared coherent radiation is generated from the two light waves by a nonlinear difference frequency process. In this case, however, the nonlinear process is due to creation of an electron spin magnetization wave oscillating with the difference frequency, rather than an electric polarization. Tuning is accomplished via the strength of the magnetic field, since the difference frequency corresponds to the energy separation between the two spin states.

So far, quasi-continuous tuning in the range 91 to 111 micrometers has been achieved with pulse peak powers up to 20 microwatts; the carbon dioxide source is 2 kilowatts (peak). True continuous tuning has been demonstrated as well, and, in the future, cw operation may also be possible through the use of a shorter wavelength pumping laser than carbon dioxide. Pumping with light having a photon energy near the energy needed to create free electrons in the semiconductor (bandgap energy) makes the Raman scattering process more efficient. Because of this resonant enhancement effect, the lower powers characteristic of cw lasers can be sufficiently converted to be observable.

Progress which is also being made in the development of lasers and coherent sources in the vacuum ultraviolet (where light is rapidly absorbed in the atmosphere and can be propagated only in a nonabsorbing medium, such as a vacuum) will be reviewed in a second article.—ARTHUR L. ROBINSON

Long-Range Weather Forecasting: Sea Temperature Anomalies

When the Arab oil embargo of last fall left the United States facing the possibility of serious shortages of heating oil, the question of how severe the winter would be was suddenly far from academic. One U.S. science official, reportedly under intense pressure to advise energy planners on the weather, made a desperate phone call to a West Coast meteorologist who is studying long-range weather prediction. After several days of hurried analysis of seasurface temperature patterns in the Pacific and other data, the scientist ventured the opinion that the eastern United States would experience a mild winter, a guess subsequently and fortunately confirmed by events. The incident underscores both the limits to our understanding of climatic processes and the role which weather patterns can play in national policy. It also illustrates the growing conviction of many scientists that it is the oceans and not the atmosphere which hold the best clues to climatic change.

Warmer weather, heavier rains, and similar changes of climate are experienced largely as shifts in atmospheric weather patterns. The oceans, however, also undergo shifts in circulation patterns and temperature which influence human commerce. The reappearance 2 years ago of el Niño, the temporary cessation of the upwelling of nutrient-rich waters off the Peruvian coast, led to the collapse of anchovy fishery in that region and a subsequent boom in soybean futures as that crop became a substitute source of protein. Recently oceanographers have found that el Niño is only a coastal manifestation of major, nonseasonal changes in currents, wind patterns, and rainfall throughout the equatorial Pacific. Smaller but similar variations in sea-surface temperature occur further north in the Pacific, and it is these variations which appear to be linked with weather patterns over the North Pacific, the United States, and possibly more remote areas.

The temperature variations appear in the form of patches of water 1° to $2^{\circ}C$ above or below normal, 100 meters or more in depth, and covering as much as 1 million square kilometers. These patches typically persist throughout a Pacific winter and may recur in successive years in the same region. As reservoirs of an enormous amount of thermal energy, these patches of anomalously warm or cool water are thought to be evidence of air-sea heat exchange or other phenomena on a scale that is large enough to influence weather patterns, although the specific mechanisms involved are not understood. Indeed, whether the hot and cold patches or other oceanic phenomena can initiate shifts in climate or are merely passive responses to atmospheric changes is still uncertain. In a series of experiments and theoretical studies that constitute the North Pacific Experiment (NORPAX), meteorologists and oceanographers are attempting to find out how the patches are formed and what their role is in the exchange of heat with the atmosphere. In the long run, NORPAX investigators hope to be able to monitor ocean temperature variations by satellite and subsurface sensors and then to predict some features of weather patterns and climatic shifts months and perhaps years in advance--predictions that, if accurate, would be of inestimable value to farmers and national policy-makers alike.

The possibility of long-range weather forecasting arises from the fact that changes in the ocean occur more slowly than those in the atmosphere, and from the development of statistical (as opposed to mechanistic) forecasting techniques in the NORPAX program. Daily weather forecasts in the United States are now based primarily on mechanistic models of atmospheric processes, a preference anecdotally attributed by meteorologists to the outcome of a debate between mathematicians John von Neumann and Norbert Wiener during the years after World War II, when computers were first being applied to weather prediction. According to the story, von Neumann was much more persuasive in his advocacy of mechanistic forecasting than Wiener, who

favored statistical methods. But despite the undoubted successes of mechanistic or "dynamic" forecasting, studies by E. Lorenz, of the Massachusetts Institute of Technology, and others indicate that the method is not likely to be extendible in detail beyond a week or two even with greatly improved data. The limitation arises because small errors in the computed forecast grow with time (a property of the nonlinear equations on which the models are based) and eventually produce an essentially random weather pattern. Nor is the difficulty purely an artifact of the models used-the predictability of turbulent flows such as those in the atmosphere is not established, even in principle. But if atmospheric motions cannot be forecast weeks or months ahead in detail, some statistical properties may nonetheless be predictable. Indeed, precipitation and other weather phenomena that are too small to show up on existing computational grids are now forecast statistically, so the methods are not completely distinct.

Reliable long-range prediction is still many years away, but statistical analysis of more than 25 years of data on sea-surface temperatures by J. Namias of Scripps Institution of Oceanography and other NORPAX scientists has turned up some interesting features. Namias finds that variations from the long-term temperature averages tend to occur as large, spatially coherent patches 1000 km or more across. Definite patterns emerge from the data-a tendency, for example, to have an anomalously cold patch in mid-Pacific and an anomalously warm patch off the California coast, or the reverse-although anomalies occur in all parts of the Pacific. Namias also finds correlations between oceanic and atmospheric data in which wind patterns are statistically associated with sea-surface temperature patterns. This association of air and sea phenomena is particularly likely in certain regions such as that from north of Hawaii to the Aleutians, which Namias speculates may be due in part to cold polar air sweeping