Physics with Lasers: High Resolution Coming of Age

Physicists have long been interested in using lasers for very precise measurements, but various difficulties have hindered the full utilization of the narrowness of the frequency range of light emitted by a laser. Now a new class of techniques that might be called saturation spectroscopy (no name is widely accepted yet) is making high resolution measurements of atomic and molecular structure widely applicable. In another development, extremely stable lasers may make possible the accurate determination of the speed of light and the detection of minute strains in the earth's crust.

An example that illustrates the improvement of saturation spectroscopy over past techniques is the first direct measurement of the Lamb shift in hydrogen at visible frequencies. Precise measurements by indirect methods of the Lamb shift-the separation of two closely spaced energy levels in an atom with one electron-provide the most convincing evidence for the correctness of the theory of quantum electrodynamics. The techniques of saturation spectroscopy make possible measurements of visible light frequencies with resolution of 1 part in 108, a value almost two orders of magnitude better than was previously possible. The same physical effect that was the progenitor of saturation spectroscopy has been applied to the building of a helium-neon laser that emits infrared radiation with a frequency so constant that it can be used as a reference for precision measurements in many new applications.

The basic physical effect that must be nearly eliminated to achieve the high resolution of saturation spectroscopy is the Doppler broadening of the frequency of an atomic transition. In the process of absorption of a photon by an atom, the frequency necessary to excite an atom to a particular energy level depends on the velocity of the atom. Since the atoms of a gas move in random direction at various speeds, some finite spread is observed in the frequencies that cause absorption-a Doppler effect. For many spectroscopic measurements, the Doppler spread is the limiting factor. In particular, the separation of the hyperfine structure of atoms (the splitting of atomic levels caused by

the spin of the nucleus) is generally smaller than the Doppler width.

In effect, the techniques of saturation spectroscopy make possible the tagging of each atom with its velocity information. Absorption is detected only for atoms moving in one direction, rather than in all directions, thus nearly eliminating the Doppler width. Many variations of saturation spectroscopy have been developed. One example is the technique of Ted Hänsch and Arthur Schawlow, of Stanford University. Two beams traveling in opposite directions pass through a glass cell containing a gas, such as hydrogen in the case of the Lamb shift experiment. The two beams have the same frequency because they originate in the same laser. If the laser is exactly tuned to one of the absorption frequencies of the gas, the first beam (called the saturating beam) will excite almost all the atoms of the gas that are moving neither forward nor backward in the beam directionthose that have no Doppler shift-and the second beam (called the probe beam) will pass through relatively undiminished in intensity.

At any other frequency, however, the two beams will excite two different populations of atoms having different velocities, and the probe beam as well as the saturating beam will be diminished. If careful monitoring techniques are used, it is possible to ascertain whether changes of intensity of the probe beam are indeed due to the saturating beam. Even if the degree of saturation is quite small, it is possible to accurately map the frequencies at which the probe beam is most intense, namely the atomic transition frequencies. In the measurement of the Lamb shift (1) the width of the narrowest transition observed was about 300 megahertz, a value reduced by a factor of 20 from the Doppler width in hydrogen (about 6000 megahertz).

Two significant improvements over earlier techniques now make the techniques of saturation spectroscopy very versatile. The absorption cell can now be moved outside the laser cavity and the sort of precision laser needed can now be tuned over a wide range of frequencies. V. I. Letokhov of the Institute of Spectroscopy at the Academy of Sciences of the U.S.S.R. in Moscow, among other people, studied absorption with a cell moved outside the laser cavity. Peter Toschek, of the Institute of Applied Physics at Heidelberg, Germany, and Hänsch studied saturation effects with two beams tuned separately inside a laser cavity. Using a CO₂ laser, Christian Bordé of the University of Paris has developed a technique similar to that of Hänsch. The contribution of the group at Stanford is that they have extended the technique to the broad spectral range of a tunable dye laser. Earlier measurements with lasers were limited to studies of gases with transitions that happened to be coincident with the lines available in lasers with very limited tunability. For example, Ken Baird, of the Canadian National Research Council in Ottawa, measured the hyperfine structure of an iodine line coincident with a visible line in a standard, helium-neon gas laser only tunable over a very narrow range. Richard Brewer and associates at the International Business Machines Laboratory at San Jose, California, have utilized saturation spectroscopy at infrared wavelengths to resolve transitions between the vibrational states of methane.

For the purposes of saturation spectroscopy, a tunable laser several orders of magnitude more monochromatic than previous devices was necessary. To achieve this, Hänsch started with a dye laser that has a tunable range of about 200 Å near the visible wavelength of 6000 Å. Dye lasers basically consist of a cell containing a dye dissolved in a solvent. When the dye solution (for instance, rhodamine in ethanol) is illuminated with another laser, called the pumping laser (a fixed-frequency nitrogen laser operating at 80 pulses per second with a power output of 100 kilowatts), the dye emits amplified light tunable to different frequencies by diffraction gratings or other optical means. The output of the basic dye laser that Hänsch uses has a bandwidth of about 500 Mhz (or 1/100 Å) which is not narrow enough for saturation spectroscopy. By the use of additional passive devices, Hänsch has been able to reduce the bandwidth to 7 Mhz (or 8×10^{-5} A), unusually narrow for tunable devices. Though many physicists will be

likely to develop laser saturation experiments of similar versatility, it seems that no one else has yet developed a tunable laser that is quite so monochromatic.

With continuing advances in saturation spectroscopy, it may soon be feasible to measure molecular structure at infrared and optical wavelengths with an accuracy that was previously only possible with microwave techniques. In general, it appears that fine structure, hyperfine structure, and other small perturbations of atomic levels such as isotope shifts and the Lamb shift are now accessible to measurement with visible light. In addition, by delaying the probe beam with respect to the saturating beam, it is possible to study the rate at which equilibrium is reestablished in a gas after saturation. Such studies may be useful for learning about the collision properties of gases and detailed properties of gas discharges.

The physical principle that makes possible such high resolution has been known for many years. However, previously a high resolution effect had been observed only within an amplifying medium, such as inside the laser cavity. The internal effect is also caused by a reduction of the Doppler line width. Like the external interaction of two opposing beams traversing the same path, a saturation effect occurs in the amplifying medium of a laser as light is reflected back and forth between the two end mirrors of the cavity. This effect was predicted by Willis Lamb, of Yale University.

The precision of the Lamb dip is very useful for defining the frequency of a laser. Although lasers are extremely monochromatic, the frequency of the emissions will vary somewhat if anything changes the optical pathlength. (Changes of temperature or movements of the end mirrors will do it.)

Extremely Stable Lasers

Ali Javan and others at the Massachusetts Institute of Technology pioneered in stabilizing laser frequencies with techniques utilizing the Lamb dip, and developing methods of measuring the frequencies of such lasers. John Hall at the National Bureau of Standards in Boulder, Colorado, has built several extremely stable helium-neon lasers. These can be made as monochromatic as 1 khz, but the central frequency may have any value within the broader spectral range of about 300 Mhz. Thus, the output of such a laser is far more monochromatic than it is

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reproducible. If a quartz cell containing methane is inserted into the laser, however, a resonance (the Lamb dip) much narrower (50 to 100 khz) than the broad spectral range can be achieved. By highly refining a feedback system keyed on the methane Lamb dip, Hall has built a laser (emitting light with a wavelength of 3.39 μ m in the infrared) that is stable to 1/2 part in 10¹¹. Furthermore, the Lamb dip occurs at a frequency that is universally reproducible because it is determined by the molecular structure of methane.

With the use of lasers stabilized with the Lamb dip technique, extremely precise measurements of fundamental constants can be made. Using the heliumneon laser built by Hall and Dick Barger, physicists of the National Bureau of Standards at Boulder expect to measure the speed of light to an accuracy of 1 part in 108. The speed of light is now known to an accuracy of 3 parts in 107. Ken Evenson has measured the frequency of the 3.39- μm line of the helium-neon laser to an accuracy of about 1 part in 10^8 (2). Barger is measuring the wavelength of the same line. The product of frequency and wavelength is the speed of light. Since the accuracy with which the center of the absorption line can be measured is 1/2part in 1011, Evenson foresees the possibility of eventually measuring the absolute frequency of this line to the same accuracy. However, the accuracy of the present standard of length is only about 1 part in 10⁸, so it may soon become the limiting factor in the measurement of the speed of light by this technique. Many scientists think that eventually laser lines may replace the present standards of both length and frequency.

In addition to measurements of basic physics and fundamental constants, the accuracy made possible with the Lamb dip is being used to improve measurements of geophysical effects. Judah Levine, of the National Bureau of Standards in Boulder, has developed a laser strain-gauge capable of measuring extremely small changes in distances between two points in the earth. There are two helium-neon lasers in his experiment. One laser is always tuned to resonate with an integral number of wavelengths in the 30-meter path between two reference points. Since the wavelength of the first laser must change as the earth moves, the frequency also changes. By comparison of the frequency changes with the fixed frequency of a second laser (built by Hall) stabilized with the Lamb dip, the

relative motions of the two reference points can be charted. At some frequencies the background from random vibrations in the earth limits the sensitivity to 1/2 part in 10^{10} . At other frequencies it is possible to achieve sensitivities approaching 1 part in 10¹³. Jon Berger and R. H. Lovberg, of the University of California, San Diego, have constructed a laser strain gauge that is different from Levine's in several respects, including the use as a frequency reference of a quartz cavity kept in an environment with constant temperature. Because it is less sensitive to temperature changes and vibrations, the laser strain gauge stabilized with Hall's Lambdip technique appears to be a significant improvement; but earth strain measurements were previously limited to an accuracy far less than that afforded by either laser device, so that both the experiments of Berger and Levine are making accessible new observations of a wide range of geophysical phenomena.

As more improvements in laser capabilities are made, the laser saturation technique may become a more powerful tool. For several reasons some physicists think that other types of tunable lasers that emit light at infrared frequencies, such as spin-flip Raman lasers or diode lasers, may ultimately prove to be even more useful than the visible dye laser at Stanford. Both these types are significantly different from dye lasers. In the spin-flip laser a spin resonance in a semiconductor-indium antimonide, for example-is used to achieve stimulated emission that can be tuned over a limited frequency range by changing an external magnetic field. The diode laser, a semiconductor also, can be tuned over a limited frequency range by simply changing the amount of current through it. Unlike dye or spin-flip lasers, diode lasers don't need to be pumped by ordinary, nontunable lasers. Furthermore, the frequency range of a diode laser may be selected by controlling the process of fabrication of the semiconductor, and manufacture may eventually be very cheap. However, even before the next generation of lasers arrives, the resolution now attainable with saturation techniques appears to be sufficiently improved over other techniques to have a significant impact on many disciplines.

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