

# NEW PROBLEMS

Charles H. Holbrow, *Editor*

Colgate University, Hamilton, New York 13346

The "New Problems" department presents interesting, novel problems for use in undergraduate physics courses beyond the introductory level. We will publish worked problems that convey the excitement and interest of current developments in physics and that are useful for teaching courses such as Classical Mechanics, Electricity and Magnetism, Statistical Mechanics and Thermodynamics, Modern Physics, or Quantum Mechanics. We challenge physicists everywhere to create problems that show how their various branches of physics use the central, unifying ideas of physics to advance physical understanding. We want these problems to become an important source of ideas and information for students of physics. This project is supported by the Physics Division of the National Science Foundation. Submit materials to Charles H. Holbrow, *Editor*.

## Doppler-free saturated absorption: Laser spectroscopy

Daryl W. Preston

Department of Physics, California State University, Hayward, California 94542

(Received 6 July 1995; accepted 22 July 1996)

### I. SCOPE

These problems illustrate laser-saturated absorption spectroscopy. They show how this technique circumvents Doppler broadening in measurements of optical spectra and so can improve resolution by several orders of magnitude. Solving these problems requires an understanding of the Boltzmann velocity distribution, the Doppler effect, and basic features of induced absorption of laser light by alkali atoms. The problems are suitable for a course in modern physics and provide background for advanced laboratory experiments in both high-resolution spectroscopy and cooling and trapping of atoms.<sup>1-3</sup>

### II. LASER ABSORPTION SPECTROSCOPY

In laser-saturated absorption spectroscopy three beams of light derived from the same laser pass through a cell containing a low-density vapor of atoms. The frequency of the laser light is varied, and resonances are observed by detecting increases in absorption when the frequency matches a transition frequency of atoms in the vapor.

#### A. Rubidium

Rubidium is a convenient atom to study with absorption spectroscopy. Because it has a single electron outside of closed shells, it has a relatively simple hydrogenlike structure of energy levels. The ground and first excited states of the single electron are, respectively,  $5^2S_{1/2}$  and  $5^2P_{1/2,3/2}$ , and, although the  $I=3/2$  nuclear spin of  $^{87}\text{Rb}$  produces the hyperfine splittings shown in Fig. 1, the transitions shown there all have wavelengths around 780 nm and are easily induced with a compact, inexpensive diode laser. Rubidium's high vapor pressure permits it to be used in a vapor cell at room temperature. The feasibility of doing without a complicated oven and also using a diode laser means that the apparatus for laser absorption spectroscopy of rubidium can be quite simple.

#### B. A laser saturated absorption spectrometer

Figure 2 is a diagram of a laser spectrometer based on a tunable diode laser like that described in Ref. 2. The laser frequency can be swept through the atomic absorption lines by varying the voltage across a piezoelectric transducer (PZT) to change the length of the laser's cavity.

The 780-nm wavelength light from the laser is divided into three beams. The most intense beam is called the "pump" beam. The other two beams, which are of considerably lower intensity than the pump beam, are sent through the vapor cell in the direction opposite that of the pump beam. These are called the "probe" beams and each is detected by a photodiode. The probe beam that is arranged to overlap the counterpropagating pump beam inside the cell is called the "overlap" beam; the other probe beam, which does not overlap with the pump beam, is called the "reference beam."

For reasons made clearer below, the signals produced by the two probe beams at the photodiodes are adjusted to be equal and then subtracted from each other. It is the difference between the two signals that is usually recorded.

### III. DOPPLER BROADENED ABSORPTION SPECTRA

In conventional laser spectroscopy the closely spaced spectral lines arising from atomic fine structure or hyperfine structure are often not resolved because of Doppler broadening. When the overlap probe beam is blocked so that it does not reach the photodiode, the absorption signal then comes only from the reference probe beam and displays Doppler broadened absorption lines. Figure 3(a) shows such a spectrum. Here, the stronger line is due to  $^{85}\text{Rb}$  (72% natural abundance) and the weaker line is due to  $^{87}\text{Rb}$  (28% natural abundance). The  $^{87}\text{Rb}$  line arises from the three  $5^2S_{1/2}(F=2) \rightarrow 5^2P_{3/2}(F'=1,2,3)$  transitions (see Fig. 1), but these appear together as one line in the data of Fig. 3(a) because of Doppler broadening.

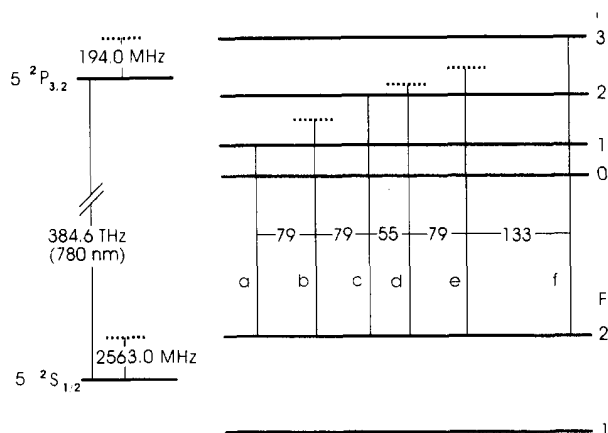


Fig. 1. Hyperfine levels and  $F=2 \rightarrow F'=1,2,3$  transitions for  $^{87}\text{Rb}$ . The numbers 79, 79, etc., are accepted values of line separations in MHz. The horizontal dashed lines correspond to crossover frequencies as explained in the text. The diagram of the overall transition shown at the left is not linearly scaled.

This well-known effect arises because the atoms have a thermal (Maxwellian) distribution of velocities. If a vapor of atoms is irradiated by laser light, thermal motion will cause most of the atoms to receive the incident light at frequencies Doppler shifted away from the laser frequency. Thus, for example, if the laser is tuned exactly to the atoms' resonant frequency  $\nu_1$ , only those atoms that have no component of velocity along the line of the incident light will be resonant with it. Those atoms which have a component of velocity  $v_z$  along the axis of the laser beam will become resonant only when the laser frequency is tuned to

$$\nu = \nu_1 \left( 1 + \frac{v_z}{c} \right), \quad (1)$$

where  $c$  is the speed of light. Equation (1) is just the equation for the Doppler shift of light when  $v_z \ll c$  as is the case here. Put another way, when the laser light is tuned to frequency  $\nu$ , the light will be resonant with just those atoms that have velocity along the laser beam of

$$v_z = \left( \frac{\nu - \nu_1}{\nu_1} \right) c. \quad (2)$$

When  $v_z > 0$ , the atoms are moving in the direction of the laser beam; when  $v_z < 0$ , they are moving opposite to the light.

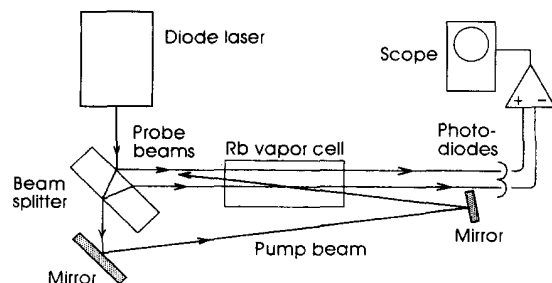


Fig. 2. A laser-diode, saturated absorption spectrometer.

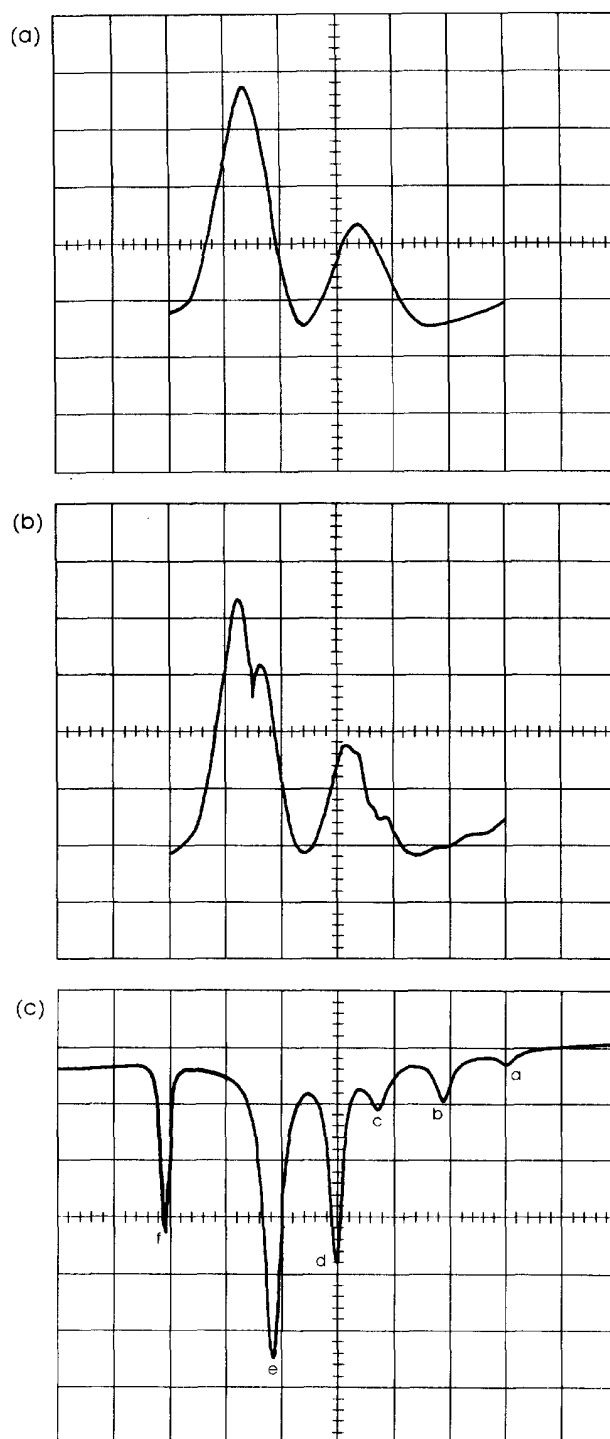


Fig. 3. Experimentally observed spectral lines of  $^{87}\text{Rb}$ , where frequency is increasing to the left: (a) Doppler broadened spectral lines; (b) Doppler broadened lines with dips; (c) saturated absorption lines, where the labels on the lines correspond to the labels in Fig. 1. In (a) and (b) one horizontal division = 534 MHz; in (c) one horizontal division = 69 MHz.

This means that as the frequency  $\nu$  of the laser is varied, the light interacts with different parts of the thermal distribution of the velocities of the atoms. A graph of absorption as a function of laser frequency  $\nu$  will have the shape of the

$v_z$  velocity distribution of the atoms, which for atoms in thermal equilibrium is just the Maxwellian distribution of the  $z$  component of the velocities:

$$e^{-Mv_z^2/2k_B T}, \quad (3)$$

where  $k_B$  is the Boltzmann constant ( $1.38 \times 10^{-23}$  J/K),  $M$  is the mass of an atom, and  $T$  is the temperature of the vapor.

## IV. DOPPLER PROBLEMS

### A. Doppler broadened absorption line shape

By substituting Eq. (2) into Eq. (3) show that the relative number of atoms in a vapor that are resonant with light in a range of frequencies around  $\nu_1$  is given by the Gaussian function

$$e^{-Mc^2(\nu-\nu_1)^2/2k_B T \nu_1^2}. \quad (4)$$

### B. Calculated Doppler linewidth

Show that this distribution has a full width at half maximum of

$$\Delta \nu_{1/2} = \sqrt{8k_B \ln 2} \frac{\nu_1}{c} \sqrt{\frac{T}{M}} = 2.92 \times 10^{-20} \nu_1 \sqrt{\frac{T}{M}} \text{ Hz}, \quad (5)$$

where  $\nu_1$  is in hertz,  $M$  is in kilograms, and  $T$  is in Kelvins. This quantity  $\Delta \nu_{1/2}$  is the linewidth due to Doppler broadening.

### C. Observed Doppler linewidth

In Fig. 3(a) the calibration of the horizontal axis is 534 MHz/div (where “div” means major division on the oscilloscope screen). Use this fact to determine the  $^{87}\text{Rb}$  absorption linewidth from the data. Compare this result with what you would expect from Eq. (5).

### D. Resolving power

From your values of the Doppler broadened linewidths, estimate the resolving power of this kind of laser absorption spectroscopy.

## V. DOPPLER ANSWERS

### A. Doppler broadened line shape

To find the linewidth, note that you want the value of  $\Delta \nu_{1/2} \equiv 2(\nu - \nu_1)$  for which Eq. (4) equals 1/2; i.e.,

$$\frac{1}{2} = e^{-Mc^2 \Delta \nu_{1/2}^2 / 4k_B T \nu_1^2}.$$

Take the internal log of both sides, and solve for  $\Delta \nu_{1/2}$ .

### B. Experimentally observed linewidth

Direct measurement from Fig. 3(a) shows the full width at half maximum of the curve is about 1.05 divisions on the oscilloscope screen. From the calibration given above, the width must be  $534 \times 1.05 = 561$  MHz.

To obtain the Doppler linewidth from Eq. (5) substitute in the mass of the absorbing atom,  $M = 87 \times 1.67 \times 10^{-27}$  kg, the temperature,  $T \approx 300$  K, and the resonance frequency of an atom in the rest frame of the laser,  $\nu_1 = (3 \times 10^8) / (780 \times 10^{-9}) = 3.85 \times 10^{14}$  Hz. From Eq. (5) the calculated linewidth for  $^{87}\text{Rb}$  at room temperature is 513 MHz.

## C. Resolving power

The frequency separation of the well-resolved Doppler broadened lines shown in Fig. 3(a) is approximately 534 MHz/div  $\times 2$  div = 1.1 GHz. If the spacing were about 1 div or 534 MHz, then the lines would be barely resolved. Hence, the resolving power is  $\nu_1 / \Delta \nu = 7 \times 10^5$ .

## VI. LASER SATURATED ABSORPTION SPECTROSCOPY

In the early 1970s Schawlow and Hänsch<sup>4</sup> developed a practical way to use nonlinear interactions of laser light with atoms to produce spectra without Doppler broadening. Their technique, known as laser-saturated absorption spectroscopy, grew out of fundamental work on nonlinear optics done by them and other physicists, e.g., Bloembergen, Lamb, and Javan.<sup>5</sup>

In this technique, two counterpropagating, overlapping laser beams of exactly the same frequency interact with atoms in a vapor. When the laser frequency is different from the resonant frequency of the atoms,  $\nu_1$ , one beam interacts with a set of atoms with some velocity  $v_z$  and the other beam interacts with an entirely different set of atoms, those with velocity  $-v_z$ . However, when the frequency is tuned to  $\nu_1$ , the two beams interact with the same group of atoms, those with velocity component parallel to the beams  $\approx 0$ . Under these circumstances the stronger beam, the pump beam, reduces the absorption experienced by the weaker overlap probe beam but only over a very narrow range of frequencies that under proper conditions can approach the natural width determined by the lifetime of the atomic transition.

### A. Laser saturated absorption spectrum

To see how the narrow absorption dip arises, consider what happens when the reference probe beam is blocked and only the overlap probe beam is allowed to strike its photodiode. Figure 3(b) shows that there appear “dips” in the spectral lines. To understand these it helps to look at Fig. 4, which shows the number of atoms in the ground state,  $N_{gs}(v_z)$  as a function of atomic velocity  $v_z$  (where positive  $v_z$  is parallel to the probe beams). In Fig. 4(a) the laser frequency  $\nu$  is less than  $\nu_1$ , the frequency of a transition from a ground-state  $F$  level to an excited  $F'$  level. The notch to the left corresponds to atoms moving in the negative direction with exactly the correct velocity  $-v_{z1}$  to see the overlap probe beam blueshifted to  $\nu_1$ ; as a result, the number of atoms in the ground state is reduced as shown in Fig. 4(a). A different group of atoms with the positive velocity  $v_{z1}$  will see the pump beam also blueshifted to  $\nu_1$ , and the number of ground-state atoms with this velocity will be reduced as shown by the notch on the right. For  $\nu \neq \nu_1$  the two beams interact with these two different groups of atoms, and as  $\nu$  is increased, the notch due to the pump beam moves left and that due to the probe beam moves right.

When  $\nu = \nu_1$  as in Fig. 4(b), the two beams interact with the same group of atoms. Then the pump beam depletes the number of atoms in the ground-state  $F$  level, and the probe beam passes through the vapor cell with reduced absorption, so that “dips” appear in the Doppler broadened absorption line, as shown in Fig. 3(b).

The two photodiodes shown in Fig. 2 are wired so that their signals subtract. If neither probe beam is blocked, the signals shown in Fig. 3(a) and Fig. 3(b) are subtracted, and the resulting signal is like that in Fig. 3(c).

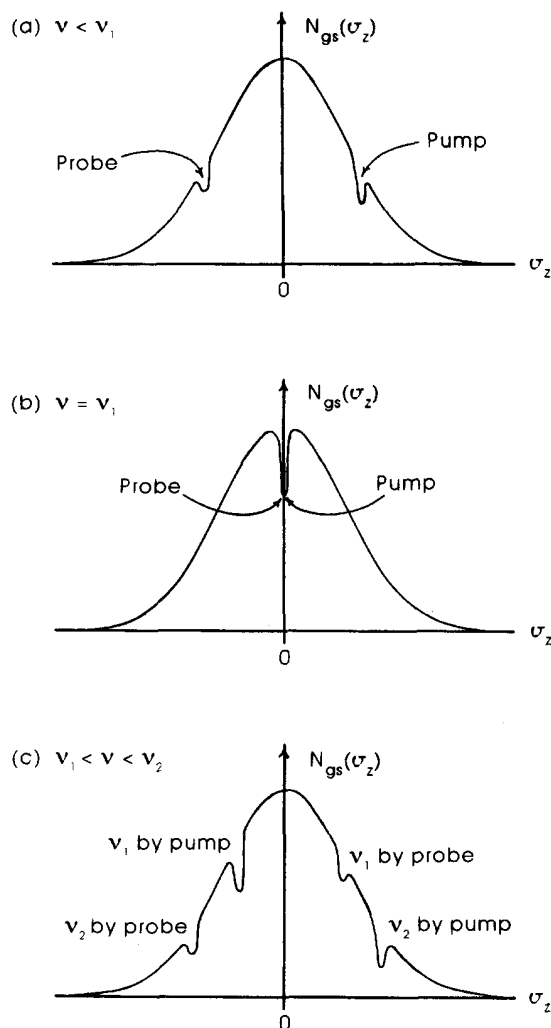


Fig. 4. Absorption of pump and probe beams by ground-state atoms depends on the Doppler shift of the laser frequency  $\nu$  to the transition frequency  $\nu_1$  by the atoms' thermal velocity: (a)  $\nu < \nu_1$ ; (b)  $\nu = \nu_1$ ; and (c) the crossover situation when  $\nu_1 < \nu < \nu_2$  and the two transitions  $\nu_1$  and  $\nu_2$  are within a Doppler width of each other.

The three dips labeled a, c, and f in Fig. 3(c) correspond to the atomic transitions labeled a, c, and f in Fig. 1. The three dips labeled b, d, and e in Fig. 3 are known as “crossover” dips and are peculiar to saturated absorption spectroscopy.

## B. Crossover transitions

A crossover dip can occur when two transitions share a common ground state and differ in frequency by less than the Doppler linewidth. In this situation there are laser frequencies at which the pump beam interacts with two different groups of atoms at the same time. An example is shown in Fig. 4(c), where  $\nu_1 < \nu < \nu_2$  so that the atoms with velocity  $-v_{z1}$  see  $\nu$  redshifted to  $\nu_1$ , while at the same time those with velocity  $+v_{z2}$  see  $\nu$  blueshifted to  $\nu_2$ . The probe beam interacts in a similar way with atoms at  $+v_{z1}$  and  $-v_{z2}$ . As the laser frequency is increased, the notches corresponding to the pump beam move left, and those of the probe beam move right. Clearly, there will be some laser frequency  $\nu_c$  such that the same group of atoms will be resonant at  $\nu_1$  with the

pump and at  $\nu_2$  with the probe (and another group at  $\nu_1$  with the probe and  $\nu_2$  with the pump). In both cases the pump beam reduces the population in the  $F=2$  level, and there is increased transmission of the overlapping probe beam giving rise to the extra dips, b, d, and e in Fig. 3.

## VII. SOME SATURATION SPECTROSCOPY PROBLEMS

### A. What is the crossover frequency?

Show that the crossover frequency  $\nu_c$  is

$$\nu_c = \frac{\nu_1 + \nu_2}{2}.$$

### B. What levels produce the crossovers?

The horizontal dashed lines on the right-hand level diagram in Fig. 1 correspond to crossover frequencies giving rise to the dips labeled b, d, and e in Fig. 3(c). Identify what two atomic transitions are involved in each case and verify that they differ in frequency by less than the Doppler width.

### C. Saturation spectroscopy linewidths

What are the widths of lines e and f in Fig. 3(c)? Given that the lifetime of these atomic states is about 28 ns, calculate the natural linewidth predicted by the Heisenberg uncertainty principle and compare it with your measured linewidth.

### D. Resolving power

From your values of the widths of the lines in Fig. 3(c) estimate the resolving power exhibited in these saturation laser spectroscopy data.

## VIII. ANSWERS TO SATURATION SPECTROSCOPY PROBLEMS

### A. What is the crossover frequency?

The crossover occurs when the laser frequency  $\nu_c$  is such that atoms with velocity  $-v_{z1}$  see the pump beam Doppler redshifted to the transition frequency  $\nu_1$  and at the same time see the probe beam blueshifted to  $\nu_2$ . In other words,

$$\nu_1 = \nu_c - \frac{v_{z1}}{c} \nu_c$$

and

$$\nu_2 = \nu_c + \frac{v_{z2}}{c} \nu_c.$$

Adding these two equations and solving for  $\nu_c$  gives

$$\nu_c = \frac{\nu_1 + \nu_2}{2}$$

exactly halfway between the two transition frequencies.

### B. Crossover frequencies in the Rb hyperfine spectrum

The crossover peaks marked b, d, and e arise, respectively, from the pairs of transitions  $F=2 \rightarrow F'=1$  and  $F=2 \rightarrow F'=2$ ;  $F=2 \rightarrow F'=1$  and  $F=2 \rightarrow F'=3$ ; and  $F=2 \rightarrow F'=2$  and  $F=2 \rightarrow F'=3$ .

These three crossover frequencies are shown in the diagram of Fig. 1 by the vertical lines labeled b, d, and e connecting dashed horizontal lines to the  $F=2$  groundstate level. The frequency separations between transitions are labeled horizontally across the middle of the diagram, 79, 79, etc. They show that all three of the observed hyperfine transitions fall within about 400 MHz of each other, well within the observed Doppler width of 561 MHz.

### C. Linewidths

In Fig. 3(c) the horizontal axis was calibrated to be 69 MHz/div. Consequently, the linewidths of the saturated absorption dips e and f shown there are respectively approximately  $69 \times 0.40 = 28$  MHz and  $69 \times 0.18 = 13$  MHz.

The natural linewidth predicted by the Heisenberg uncertainty principle for a state with lifetime  $\tau = 28$  ns is  $\Delta\nu = 1/(2\pi\tau) = 6$  MHz, which is 1/2 to 1/5 of what is measured.

Spectral linewidths depend on the rate at which the atoms are undergoing collisions, on the particular gas species present in the cell, and on the rate at which the laser light stimulates the observed transitions. They also depend on the spread of frequencies in the laser light itself, but, since this is no more than a few MHz for the laser used here, the discrepancy between the observed value of  $\Delta\nu$  and the value predicted by the uncertainty principle is probably due to a combination of the other effects.

### D. Resolving power

The frequency separation of spectral lines labeled e and f is  $69 \text{ MHz/div} \times 1.9 \text{ div} = 131 \text{ MHz}$ . If the separation was about 0.3 div or 21 MHz, the peaks would be barely resolved. Hence, the resolving power is about  $18 \times 10^6$ , a more than 20-fold improvement over the resolving power of ordinary linear spectroscopy.

### ACKNOWLEDGMENT

The author is indebted to Carl Wieman, University of Colorado, for providing Fig. 3(c).

<sup>1</sup>Drayl W. Preston and Carl E. Wieman, "Doppler-free saturated absorption spectroscopy: laser spectroscopy," an unpublished advanced laboratory writeup available from Daryl W. Preston, e-mail: dpreston@csuhayward.edu

<sup>2</sup>K. D. MacAdam, A. Steinbach, and C. Wieman, "A narrow-band tunable diode laser system with grating feedback, and a saturated absorption spectrometer for Cs and Rb," *Am. J. Phys.* **60**, 1098–1111 (1992).

<sup>3</sup>Carl Wieman, Gwenn Flowers, and Sarah Gilbert, "Inexpensive laser cooling and trapping experiment for undergraduate laboratories," *Am. J. Phys.* **63**, 317–330 (1995).

<sup>4</sup>Arthur L. Schawlow, "Spectroscopy in a new light," *Rev. Mod. Phys.* **54**, 697–707 (1982) (1981 Nobel lecture).

<sup>5</sup>Nicolaas Bloembergen, "Nonlinear optics and spectroscopy," *Rev. Mod. Phys.* **54**, 685–695 (1982) (1981 Nobel lecture).

### ASSEMBLY LANGUAGE PROGRAMMING

What you can't see in the picture is that this machine was programmed entirely in assembly language. Over 4000 statements on punch cards were eventually written for the analysis codes. Also, due to the limited hardware of the system (16K memory, no disk drive), I could not use the available operating systems provided by the manufacturer, which would have been too large and slow for the purposes of this application anyway. Hence the task of programming the computer also involved writing my own custom drivers for the peripherals, servicing the interrupts, and the like. I do recall a certain sense of pride that every bit set and every action carried out by that machine was explicitly controlled by code that I had written. I also recall saying when it was done that while it had been quite a valuable and interesting experience to understand and program a computer so extensively at such a fundamental level, once in a lifetime for this type of experience was enough!

Russell A. Hulse (Nobel Lecture, December 8, 1993) "The Discovery of the Binary Pulsar," *Reviews of Modern Physics*, **66** (3), 699–710 (1994).