Narrow Hyperfine Absorption Lines of Cs¹³³ in Various Buffer Gases

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Line widths as narrow as 40 cps have been obtained for the hyperfine transition at 9192 Mc/sec in cesium vapor. The Doppler width was greatly reduced by collisions with an inert buffer gas. An optical detection method was used to observe the microwave absorption lines. Shifts in the observed frequency due to collisions with the buffer gas were measured as a function of the temperature and pressure of the gas. It was found possible to reduce greatly the shift in frequency with temperature by using the proper mixtures of inert gases. The contribution to line width due to collisions with the buffer gas was also determined.

R ECENTLY collisional narrowing¹ has been used to reduce the Doppler width of hyperfine transitions observed in hydrogen² and in alkali vapor absorption cells.³⁻⁵ For the ground-state hyperfine transition in Cs¹³³ at 9192 Mc/sec we have obtained line widths of 40 cycles/sec with good signal-to-noise ratios. The method used was an extension of the optical pumping procedures introduced by Kastler,⁶ Dicke,⁷ and Dehmelt.⁸

A schematic diagram of the energy levels in Cs is given in Fig. 1. The excited state hyperfine splittings are much smaller than the ground state splitting and will be ignored. The 8521.4 A line from an argon discharge underwent Zeeman splitting in a field of about 5000 gauss and focused on an absorption cell containing Cs vapor and an inert buffer gas. One component then overlapped the 8521.2 A component of the Cs $6S_* \rightarrow 6P_*$ absorption line and produced transitions from the F=4 sublevel of the ground state to the excited state. Subsequent spontaneous emission to both ground state sublevels left a net difference in population between them. Since light could be absorbed by only those atoms in the F=4 sublevel, the amount of scattered light provided a convenient measure of the population difference. When microwaves of the right frequency to produce the relatively field-independent $(3,0) \rightarrow (4,0)$ transition were applied, the population difference decreased and this was seen as an increase in the scattered light intensity. The frequency source, supplied by the NRL Radio Techniques Branch, has been described elsewhere.9 It was stable to 1 part in 10¹⁰ over periods of from several seconds to many hours. Figure 2 shows the type of line obtained with this apparatus by changing the frequency in 4-cps steps.

 ² J. P. Wittke and R. H. Dicke, Phys. Rev. 103, 620 (1956).
 ³ M. Arditi and T. R. Carver, Phys. Rev. 109, 1012 (1958).
 ⁴ W. E. Bell and A. L. Bloom, Phys. Rev. 109, 219 (1958).
 ⁵ M. Arditi and T. R. Carver, Phys. Rev. 112, 449 (1958), preceding paper.

- ⁶ A. Kastler, J. phys. radium 11, 255 (1950).
 ⁷ W. B. Hawkins and R. H. Dicke, Phys. Rev. 91, 1008 (1953).
 ⁸ H. G. Dehmelt, Phys. Rev. 105, 1487 (1957).

⁹ Bender, Beaty, and Chi, Proceedings of the Twelfth Annual Frequency Control Symposium, Asbury Park, New Jersey, May 6, 7, 8, 1958 (unpublished).

The resonance frequency was found to be proportional to buffer gas pressure, the shifts being +1050, +890, +580, -190 and -1300 cps/mm Hg for He, N₂, Ne, A, and Kr. These results are similar to those of Arditi and Carver reported in the preceding paper.⁵ A mixture of 75% A-25% Ne gave no pressure shift at 23°C. At higher temperatures the shift was linear and negative, and at lower temperatures it was linear and positive. An extrapolation to zero buffer gas pressure gave a frequency about 30 cycles/sec below the Cs beam reference frequency.⁹ The absorption cells used were thoroughly outgassed 500-ml Pyrex flasks containing distilled Cs and spectroscopically pure buffer gases. The shifts in frequency with bulb temperature were found to be about +15, +1, -7 and -5 cps/(degcm Hg) for He, Ne, A, and the A-Ne mixture. A small amount of A in Ne is expected to make the resonance frequency independent of temperature for a fixed density.



FIG. 1. Energy level diagram for Cs133.

¹ R. H. Dicke, Phys. Rev. 89, 472 (1953).



FIG. 2. Optically detected cesium hyperfine transition for an absorption cell containing neon at 10-cm pressure as the buffer gas. The frequency of the transition is shifted by about 59 kc from the frequency that would be obtained with no buffer gas present.

Line-width measurements gave considerably narrower lines for Ne and He than for A. With Ne the width at half-power was about 40 cps from 1 cm to 10 cm, and about twice this width at 3 mm and 50 cm with much weaker signals. With He, line widths of 40 cps were found at 1 and 3 cm. For Ne, the calculated reduced Doppler width at 3 mm was about 50 cps, while the extra width at 50 cm was consistent with a contribution from buffer gas collisions of 1 to 2 cps/cm. The line width for A, however, was about 120 cps at 1 and 4 cm,

and somewhat more at 15 cm. Light intensity, magnetic field inhomogeneity, and Cs-Cs collisions do not appear to be responsible for the residual line widths, but short-term fluctuations in the applied frequency could contribute. The lines observed were Lorentz-shaped.

Measurements were also made of line widths for the Zeeman transitions¹⁰ ($\Delta F = 0$) in Rb and Cs vapor by transmission monitoring.^{8,11} At high buffer gas pressures the line widths in Cs were comparable with those for the hyperfine transition, i.e., about 4 cps/cm for A and roughly one third as much for Ne. The value of T_2 derived from the line width for Cs in A was about a factor 5 smaller than the relaxation time measured by Dehmelt's method.⁸ A reasonable explanation is that the transitions for which the change in M is small are the most important ones in the relaxation process. Line widths as low as 15 cps were observed in the earth's field,¹² permitting complete resolution of the 46-cps second-order splittings in Rb⁸⁷ and observation of multiple quantum transitions.

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¹⁰ The Zeeman transition measurements were carried out at the Fredericksburg Magnetic Observatory of the U.S. Coast and Geodetic Survey

 ¹⁰ W. E. Bell and A. L. Bloom, Phys. Rev. 107, 1559 (1957).
 ¹² T. L. Skillman and P. L. Bender, J. Geophys. Research (to be published).