permitting measurement of $\Delta v/v \sim 10^{-7}$, this method should prove superior to the ultrasonic attenuation, to the de Haas–van Alphen, or to the oscillatory magnetoresistance measurements.

The authors are grateful to S. Fischler for growing the bismuth samples, to P. N. Argyres and P. E. Tannenwald for valuable discussions, and to D. F. Kolesar, P. A. Ligor, and C. D. Parker for assistance in various phases of the experiment.

*Operated with support from the U. S. Army, Navy, and Air Force.

†Also at Lincoln Laboratory.

‡Supported by the Air Force Office of Scientific Research.

[1]A. A. Galken and A. P. Koroliuk, J. Exptl. Theoret. Phys. 34, 1025 (1958) [translation: Soviet Phys.–JETP 7, 708 (1958)]; A. G. Beattie, H. B. Silsbee, and E. A. Uehling, Bull. Am. Phys. Soc. 7, 478 (1962).

[2]P. N. Argyres, Lincoln Laboratory Quarterly Progress Report, 1960 (unpublished) p. 37; J. J. Quinn and S. Rodriguez, Phys. Rev. Letters 9, 145 (1962); M. J. Harrison, Phys. Rev. Letters 9, 299 (1962).

[3]J. K. Galt, W. A. Yager, F. R. Merritt, B. B. Cetlin, and A. D. Brailsford, Phys. Rev. 114, 1396 (1959).

[4]R. N. Brown, J. G. Mavroides, and B. Lax (to be published).

[5]M. Cohen and E. I. Blount, Phil. Mag. 5, 115 (1960).

[6]D. H. Reneker, Phys. Rev. Letters 1, 440 (1958).

OPTICAL DOUBLE-PHOTON ABSORPTION IN CESIUM VAPOR*

I. D. Abella
Columbia University, New York, New York
(Received October 29, 1962)

This Letter reports the observation of optical double-photon absorption[1] in an atomic system: excitation of the $6S_{1/2}$-$9D_{3/2}$ transition in Cs by intense light from a ruby maser. For a double-photon transition to occur, the energy of the excited level must be twice that of a single incident photon, and the initial and final states must have the same parity. The $9D_{5/2,3/2}$ levels in Cs, which have term[2] values 28 836.06 and 28 828.90 cm^{-1}, are conveniently at twice the frequency of the 14 400-cm^{-1} (~6940Å) light from a ruby optical maser. Furthermore, they allow a two electric-dipole transition since $\Delta l = 2$, $\Delta J = 1, 2$. The Doppler width of the transition is 0.04 cm^{-1}, and although the actual width may be somewhat greater due to collisions, the necessity for obtaining a coincidence between this sharp absorption and the maser emission frequency imposes a stringent experimental requirement on thermal tuning[3] of the ruby maser. The absorption was detected by observing the fluorescent decay of the $9D_{3/2}$ state to $6P_{3/2}$, yielding quanta at 5847 Å.

An atomic resonant double-quantum transition allows a more straightforward application of theory than have other somewhat similar cases. Two-photon excitation of impurity levels in a crystal has been considered by Kleinman[4] and observed in CaF_2:Eu^{++} by Kaiser and Garrett.[5] In this material the intermediate states are believed to be charge transfer bands of large oscillator strength. Single-quantum absorption is observed over a band as wide as about 5000 cm^{-1}, and the observed forbidden transition may occur because of crystalline field, phonon interaction, or magnetic dipole effects. Single-quantum forbidden transitions have also been observed between levels in alkali metals[6,7] and attributed to quadrupole absorption.[8]

For Cs vapor, the expected transition probability and signal were calculated from second-order electric-dipole matrix elements using the Bates-Damgaard[9] method, and summed over those states with large contributions, i.e., small energy denominators. Our maser produces about 3×10^{18} photons (~1 joule) in 0.5 msec in a beam of 0.25 degree half-angle. If this is focused by a 2.0-cm lens into cesium vapor at 0.1 Torr pressure, we expect to excite about 5×10^{12} atoms into the $9D_{3/2}$ state, assuming that the maser line is about 0.04 cm^{-1} wide. Under these conditions and neglecting possible collision effects, we expect that 5×10^{11} photons will be emitted in the 5847 line. Cesium-cesium collisions and possible maser-line broadening will reduce this number.

Experimentally, the output of a pulsed ruby source, filtered to remove background light, was focused onto a heated glass cell where cesium vapor could be introduced from a reservoir tube. Provisions were made for periodically pumping out accumulated gases due to heating; the residual pressure in the cell with cesium cold was kept below 10^{-6} Torr. The emission from the cell, observed at right angles to the ruby beam, was filtered through a $CuSO_4$ solution to remove scat-

tered red light. It was then incident on a narrow band-pass interference filter centered at 5850 Å and it was finally detected with a photomultiplier. The output of the phototube was displayed on an oscilloscope. The red light leaving the cell in the incident direction was monitored in part by a second photodetector, the remainder being focused onto the slits of a high-resolution grating spectrograph and photographed in the eighth order together with a comparison spectrum. Thus, for each flash, the ruby output, wavelength and intensity, and cesium emission were recorded simultaneously. The output wavelength of the ruby was controlled by varying the firing rate and the nitrogen cooling. It was extremely difficult to adjust the wavelength to the cesium line. A thermocouple was attached to the ruby and used as a rough guide. In practice it was necessary to let the ruby temperature (wavelength) slowly drift through the resonance condition while the maser was periodically fired.

We have observed that with no cesium vapor in the cell, no emission was detected over a wide range of ruby wavelengths. With 0.1 Torr of Cs vapor present in the cell, no fluorescence was detected except when the incident light was centered at $\lambda = 6935.5$ Å ± 0.05 Å, as measured on the spectrograph plates. Cooling the cesium reservoir and reheating caused the disappearance and subsequent reappearance of the emission. Figure 1 shows some of these results. There is no level in cesium to which the above wavelength could excite atoms from the ground state with a single quantum. However, two photons of $\lambda = 6935.5$ Å have total energy 28 828 cm^{-1} which corresponds to the $9D_{3/2}$ term value. The dependence of the emission at 5847 Å on the presence of Cs vapor and on the incident wavelength is evidence that the two-photon absorption has occurred. Detection of stray background radiation at 5847 Å coming from the ruby source would not exhibit any ruby wavelength dependence. Various other possible competing mechanisms for production of the 5847Å radiation also seem to be ruled out. The cooling arrangement used did not permit similar observations on the $6S_{1/2}$-$9D_{5/2}$ transition, which requires ruby photons of 4 cm^{-1} higher energy.

We have detected about 4×10^4 photons at the phototube and, allowing for geometry and $CuSO_4$ filter absorption, calculate that 5×10^9 photons were emitted, or a factor of 10^2 below the number expected. The maser radiation might be as much as a factor of two wider than the Cs resonance, accounting for a small part of this discrepancy. The largest part is probably accounted for by interatomic collisions which quench the $9D_{3/2}$ state, allowing radiationless transitions to other levels. Similar effects in cesium vapor have already been observed.[10] An approximation to the rate of such quenching is given by assuming that it occurs whenever the interatomic electric dipole-dipole interaction is as great as the energy separating atomic levels. Under conditions of the experiment, this calculation would predict quenching by about a factor of 100, which is the discrepancy observed.

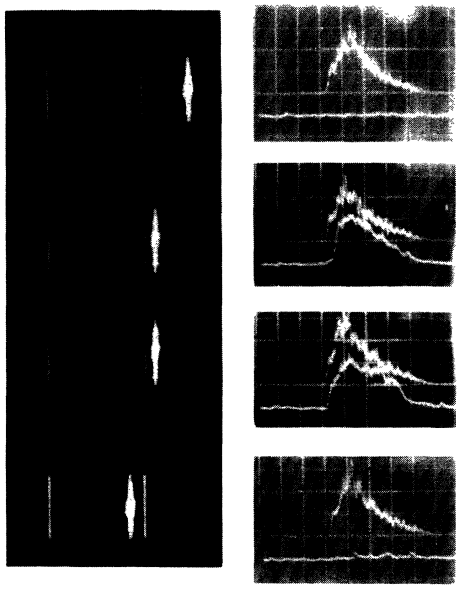

FIG. 1. (a) Each of the spectra shows two cadmium comparison lines (sixteenth order), 3466.20 and 3467.65 Å, and a single flash of the thermally tuned ruby line $\lambda \sim 6935$ Å (eighth order). Wavelength increases to the right. The ruby temperatures were about -160°C. (b) The oscilloscope records corresponding to the pulses in 1(a) show the cesium emission (lower beam) and the ruby output (upper beam). The time scale is 0.2 msec/cm. The fluorescence is observed when $\lambda_R = 6935.5 \pm 0.05$ Å. The cesium temperature was 210°C (~0.1 Torr).

Grateful acknowledgement is made to Professor C. H. Townes for valuable contributions to the progress of this work, and to Professor R. Novick for useful discussions.

*Work supported in part by the Air Force Office of Scientific Research, and in part jointly by the U. S. Army Signal Corps, the Office of Naval Research, and the Air Force Office of Scientific Research.

[1]M. Goppert-Mayer, Ann. Physik 9, 273 (1931).

[2]C. E. Moore, National Bureau of Standards Circular

467, Vol. III, p. 125.
[3]I. D. Abella and H. Z. Cummins, J. Appl. Phys. 32, 1177 (1961).
[4]D. Kleinman, Phys. Rev. 125, 87 (1962).
[5]W. Kaiser and C. G. B. Garrett, Phys. Rev. Letters 7, 229 (1961).
[6]A. L. M. Sowerby and S. Barratt, Proc. Roy. Soc. (London) 110A, 190 (1926).
[7]Ny Tsi-Ze and Weng Wen-Po, Compt. rend. 203, 860 (1936).
[8]A. F. Stevenson, Proc. Roy. Soc. (London) 128A, 591 (1930).
[9]D. R. Bates and A. Damgaard, Phil. Trans. Roy. Soc. London 242, 101 (1949).
[10]H. Z. Cummins, I. D. Abella, O. S. Heavens, N. Knable, and C. H. Townes, Advances in Quantum Electronics, edited by J. Singer (Cambridge University Press, New York, 1961), 2nd ed., p. 12.

STIMULATED RAMAN SCATTERING FROM ORGANIC LIQUIDS*

Gisela Eckhardt, R. W. Hellwarth, F. J. McClung, S. E. Schwarz, and D. Weiner
Hughes Research Laboratories, Malibu, California

and

E. J. Woodbury
Hughes Aircraft Company, Culver City, California
(Received October 1, 1962)

The existence of stimulated emission near 7670 Å accompanying normal emission from a ruby laser operating in the pulsed reflector mode (sometimes called giant-pulse, or Q-spoiling, mode[1,2]) has been reported by one of us.[3] We have found that this "satellite" emission originated in the nitrobenzene of the Q-spoiling Kerr shutter, that the same mechanism that produced this light can be employed to produce other laser lines from nitrobenzene and from a number of other organic liquids, and that the mechanism is almost certainly stimulated Raman scattering.

To operate a liquid laser, it has been convenient to place the liquid inside the ruby-laser cavity where the 6943Å ruby light which pumps it is strong and where the same cavity can be employed for laser action from the liquid. To avoid complications, the Kerr material employed in the reflectivity shutter was changed from nitrobenzene to solid potassium dihydrogen phosphate (KDP) which appears not to produce any spurious light. The ruby-pumped liquid laser apparatus then consisted of a linear array of the following components: (1) a partially transmitting multilayer dielectric-coated reflecting end plate; (2) a 3-in. $\times\frac{3}{8}$-in. polished pink ruby rod (surrounded by an H22A helical flash lamp); (3) a polarizing Wollaston quartz prism; (4) an absorption cell (2.5, 5, or 10 cm long) in which the laser liquid is placed; (5) a KDP Kerr cell; and (6) the second multilayer-coated reflecting end plate. Unless stated otherwise, the end-plate characteristics were such that the loss factor (minus one half the log of the product of the reflectivities) was about 0.17 at 6943 Å, remained flat to 7400 Å, then rose to 0.45 at 7500 Å and 0.55 at 7660 Å, and rose even faster beyond. This loss factor plus other presumably constant losses must be (at least) equaled by the gain per cm in the liquid times the length of the liquid column for laser action to occur. The output pulse of ruby R-line laser light was typically between 0.2 and 2 MW peak, of 20- to 70-nsec duration, and between 50 and 200 millijoules in energy depending on flash-lamp excitation, shutter timing, and some uncontrollable factors such as flash-lamp age, and end-plate and Kerr-cell deterioration. The output light was monitored from both ends: on one end by a photomultiplier with filtering and attenuation so as to monitor the 6943Å light; on the other end by a Bausch and Lomb dual-grating spectrograph, employing either photographic plates or a sliding photomultiplier, for recording the light output from the liquid.

The wave numbers (as read from photographic plates) of the outputs which resulted when each of several liquids was in the apparatus during a "giant" ruby-laser pulse are listed in Table I. Three observations indicate that what occurs at these frequencies is stimulated or induced emission which manifests itself as a material gain per cm that overcomes cavity losses (i.e., laser action). First, the degree of beam collimation at the listed frequencies appeared to be (within a factor of two from experimental uncertainty) the same as that of the ruby-laser light (~one milliradian). Secondly, spectral narrowing was observed as the liquid was pumped harder; the lines in some instances were as narrow as 0.3 Å and

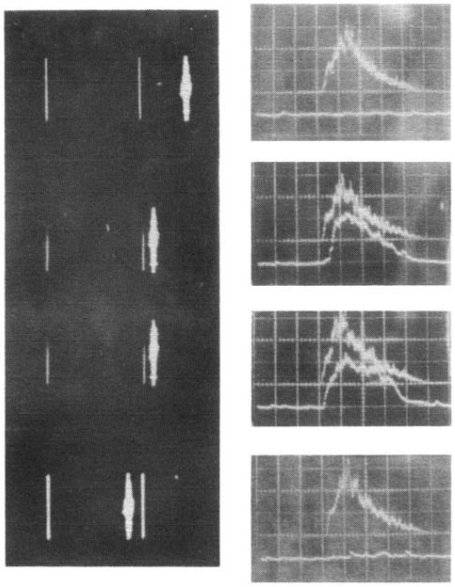

FIG. 1. (a) Each of the spectra shows two cadmium comparison lines (sixteenth order), 3466.20 and 3467.65 Å, and a single flash of the thermally tuned ruby line $\lambda \sim 6935$ Å (eighth order). Wavelength increases to the right. The ruby temperatures were about -160°C. (b) The oscilloscope records corresponding to the pulses in 1(a) show the cesium emission (lower beam) and the ruby output (upper beam). The time scale is 0.2 msec/cm. The fluorescence is observed when $\lambda_R = 6935.5 \pm 0.05$ Å. The cesium temperature was 210°C (~0.1 Torr).