PRECISE ISOTOPE SHIFT MEASUREMENTS USING LINE NARROWING INDUCED BY LASER RADIATION*

R. H. Cordover, P. A. Bonczyk, and A. Javan
Physics Department, Massachusetts Institute of Technology, Cambridge, Massachusetts
(Received 27 March 1967)

The influence of laser action on the line shape of spontaneous emission from the upper or lower level of a Doppler-broadened gas-laser transition has been described previously.[1,2] If such a spontaneous emission signal is viewed along the axis of laser propagation, its line shape is changed considerably when the laser is allowed to oscillate. This change occurs over a narrow frequency interval which may be considerably less than the Doppler width. This Letter gives the first application of this effect to ultraprecise measurements of isotope shifts for two optical transitions in neon. The splittings due to the isotope effect are completely resolved. The measured linewidths are also analyzed.

In this experiment, a short Brewster-angle He-Ne gas laser was made to oscillate on a single mode of the 1.15-μ transition ($2p_4 \leftarrow 2s_2$).[3] The spontaneous emission at 6096 Å ($1s_4 \leftarrow 2p_4$),[3] which originates from the lower laser level, was observed through an end mirror. (In the following, we refer to the latter transition as the 0.6-μ transition.) The reflectance of the laser mirrors was designed to be transparent at 0.6 μ. The signals were observed with a high-resolution, pneumatically tuned Fabry-Perot interferometer and a photomultiplier with a pinhole for mode selection. The laser was switched on and off by a square-wave voltage applied to a transducer supporting one of the laser mirrors. Modulation of the transducer at an audio frequency allowed narrow-band phase-sensitive detection of the signal. Before discussing in detail the isotope measurements, let us review the observations for a single isotope.

Suppose the laser is adjusted to oscillate at a frequency close to ω_0, the frequency of the peak of the Doppler-broadened 1.15-μ transition. In this case, the linewidth of the 0.6-μ transition, analyzed in the forward direction, consists of a narrow, nearly Lorentzian response superimposed on a broad, Gaussian, Doppler profile. The center frequency of the Lorentzian response coincides with the center frequency of the Gaussian profile. As the laser frequency is detuned, the Lorentzian response splits symmetrically about its center frequency [see Fig. 1(a)].

For further clarity, we now give a brief explanation of the origin of the above effect. The nearly Lorentzian signals arise from changes

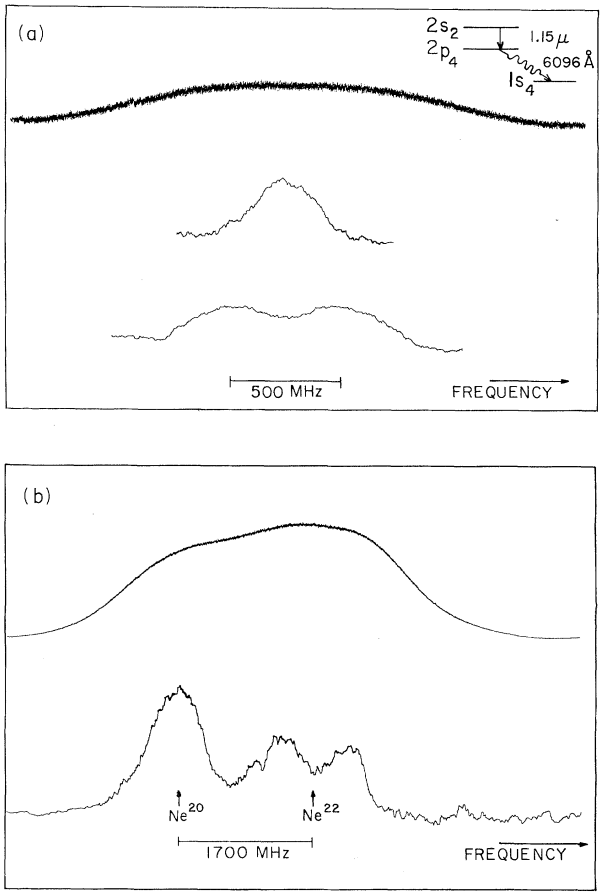

FIG. 1. Spontaneous emission signals from lower laser level observed along laser axis. (a) For pure neon isotope, the upper trace is the direct output of the phototube and represents the normal Doppler-broadened spontaneous emission. The center trace is the output of the phase-sensitive detector when the laser frequency is set to the center of the Doppler-broadened laser transition. In the lower trace, the laser frequency was set away from the center of the 1.15-μ Doppler-broadened line and the signal splits into two components corresponding to both traveling waves. (b) For 3:2 mixture of Ne^{22}-Ne^{20}, the laser frequency was set to the peak of the Ne^{20} Doppler line center. Note the barely resolved structure in the normal spontaneous emission signal in the top tracing.

in the populations of the laser levels induced by the laser field. For a Doppler-broadened transition, these changes occur for atoms whose velocities v_z lie within a narrow range (where z is the direction of propagation of the laser radiation). This leads to a departure of the distribution of atoms with velocity component v_z from the usual Maxwellian velocity distribution for the population of each laser level. If the laser oscillates close to the peak of its Doppler response, the departure from the Maxwellian distribution occurs around $v_z \simeq 0$. [To first order in the laser field intensity, the functional dependence of this departure on v_z is a Lorentzian with half-width at half maximum intensity given by $\Delta v = c\gamma/\omega_0$,[4] where c is the speed of light and $\gamma = \frac{1}{2}(1/T_1 + 1/T_2)$, with T_1 and T_2 being the radiative widths of the two laser levels.] This change in the Maxwellian distribution leads to the observed behavior of the spontaneous-emission signal viewed in the z direction.[5] Suppose now the laser frequency ω is detuned from ω_0. In this case changes in population occur within two narrow ranges of velocity centered about $v_z{}^0 = \pm c(\omega - \omega_0)/\omega_0$. The + and − signs arise from the standing wave nature of the laser field. This leads to the symmetrical splitting of the signals observed for the 0.6-μ spontaneous emission. It may be seen that, due to the Doppler effect, the splitting at 0.6 μ is given by $2\omega_0' |v_z{}^0|/c = 2|(\omega - \omega_0)| \times \omega_0'/\omega_0$, where ω_0' is the center frequency of the spontaneous emission.

Let us now consider the effect described above as applied to the determination of isotope shifts. The measurements were done using a He-Ne laser tube containing a sample of Ne with a (3:2) Ne^{22}-Ne^{20} mixture. The two isotopic components overlap for the 1.15-μ transition as well as for the 0.6-μ transition. The upper tracing of Fig. 1(b) shows the usual Doppler profile for the spontaneous emission at 0.6 μ. Note that the isotope shift in this case is barely resolvable. The laser-induced signals were observed with the 1.15-μ oscillation set to a frequency close to the peak of the Doppler response of one of the isotopes (for example Ne^{20}). Accordingly, the Ne^{20} transition at 0.6 μ shows a single narrow line. However, because of the isotope shift at 1.15 μ, the response due to Ne^{22} at 0.6 μ is split. Furthermore, the isotope shift for the 0.6-μ transition results in a shift of the center between the two peaks of the Ne^{22} signal away from the center of the signal due to Ne^{20}. This is seen in the lower tracing of Fig. 1(b). Notice that in this case the isotopic splittings are completely resolved. Furthermore, this tracing contains information on isotope shifts for the 1.15-μ transition as well as the 0.6-μ transition. A series of tracings obtained at various frequencies of laser oscillation was analyzed. The measured isotope shift at 0.6 μ is 1706 ± 30 MHz. The shift at 1.15 μ is 257 ± 8 MHz. Ne^{20} lies on the low-frequency side for both transitions. The result of the 1.15-μ measurement is in complete agreement with an earlier precise experiment involving the heterodyning of two lasers.[6] It should be noted, however, that since the 0.6-μ transition is not a laser oscillation, the measurement of the type discussed in Ref. 6 is not applicable to this transition.

We have also studied the line shape of the observed signals as a function of pressure. This line shape, extrapolated to zero pressure, has been analyzed approximately in terms of two Lorentzian responses as discussed in Ref. 5. Using the $2s_2$ and $2p_4$ radiative widths given in Ref. 6, we obtain a radiative lifetime of 1.6×10^{-8} sec for decay of the $1s_4$ level to the ground state. In view of cumulative errors in this measurement, the value given for this lifetime is accurate to within a factor of 2.

We would also like to point out that the above technique is applicable, in principle, to measurement of details of the structure of any transition which involves states populated directly or indirectly via radiative cascade from the laser levels. Furthermore, this technique may be used to define precisely the centers of Doppler-broadened atomic transitions and thus may be applied to problems related to achieving standards of length.

The authors would like to acknowledge the technical assistance of L. W. Ryan, Jr., in the design and construction of the apparatus.

---

*Work supported by U. S. Office of Naval Research and the National Aeronautics and Space Administration.

[1]A. Javan, in Quantum Optics and Electronics; Lectures Delivered at Les Houches During the 1964 Session of the Summer School of Theoretical Physics, University of Grenoble, edited by C. De Witt, A. Blandin, and C. Cohen-Tannoudji (Gordon and Breach Publishers, Inc., New York, 1965), p. 383.

[2]J. A. White, J. Opt. Soc. Am. 55, 1436 (1965).

[3]Paschen notation. The configuration for $2s_2$ is $2p^5(^2P_{1/2}{}^\circ)4s(J=1)$, for $2p_4$ it is $2p^5(^2P_{1/2}{}^\circ)3p(J=2)$, and

for $1s_4$ it is $2p^5(^2P_{1/2}^\circ)3s(J=1)$. For notation, see C. E. Moore, Atomic Energy Levels, National Bureau of Standards Circular No. 467 (U.S. Government Printing Office, Washington, D. C., 1949), Vol. 1.

[4]See Eq. (19) in A. Szöke and A. Javan, Phys. Rev. 145, 1137 (1966).

[5]The line shape resulting from the change in population due to the laser field is a linear superposition of two Lorentzians both centered at ω_0', the center frequency of the 0.6-μ transition. Their corresponding half-widths at half maximum intensity (in angular-frequency units) are given by $\gamma_\pm = \gamma' \pm \alpha\gamma$, where $\alpha = \omega_0'/\omega_0$ and γ' is similar to γ, except referred to the two levels of the 0.6-μ transition. The intensities of the two Lorentzians are proportional to their respective half-widths at half maximum intensity. This result applies only when the usual Doppler width is considerably larger than $\gamma_\pm$.

[6]A. Szöke and A. Javan, Phys. Rev. Letters 10, 521 (1963).

OBSERVATION OF TUNABLE OPTICAL PARAMETRIC FLUORESCENCE*

S. E. Harris, M. K. Oshman, and R. L. Byer
Department of Electrical Engineering, Stanford University, Stanford, California
(Received 24 March 1967)

We report the observation of an optical fluorescence which is thermally tunable over a significant portion of the visible and near-infrared spectrum. Using a crystal of $LiNbO_3$ and a 4880-Å argon laser as a pump, we have observed tuning from 5400 to 6600 Å. Fluorescent powers were estimated to be about 10^{-12} W and to have linewidths which were measured to be less than 75 Å, and are probably less than 1 Å.

We believe that the observed effect is that of spontaneous emission or fluorescence from a phase-matched parametric system. This effect has been predicted by a number of authors[1-3] and is a result of the quantum-mechanical possibility that a driving pump photon may spontaneously split into a signal and an idler photon such that $\nu_S + \nu_i = \nu_p$. Alternatively the effect can be considered to arise from zero-point fluctuations of the signal and idler modes. From another point of view the observed fluorescence can be considered as a direct observation of the quantum noise that is associated with any linear amplifier.[4]

A schematic of the experimental arrangement is shown in Fig. 1. A cw argon laser with an output power of about 300 mW at 4880 Å was used as the pumping source. The $LiNbO_3$ crystal was 1 cm long and was oriented with its optic axis in its face and parallel to the polarization of the incoming laser beam. The crystal was mounted in an oven capable of varying its temperature from room temperature to about 375°C. By changing the crystal temperature and thereby varying the refractive indices, the momentum-matching condition $\vec{k}_S + \vec{k}_i = \vec{k}_p$ for an extraordinary pump wave and ordinary signal and idler waves could be satisfied and tuned over a wide range of wavelengths.[5,6] The output analyzer was oriented at 90° to the input polarizer and together with two blue stop filters provided about 120 dB of discrimination against the laser pumping light. A Leiss prism monochromator with a resolution which we estimated at about 75 Å followed the filter.

With the crystal temperature at about 125°C, a dull fluorescent emission in the red was observed visually. As the crystal temperature was increased, this emission moved progressively toward the green. Results obtained using the Leiss monochromator are shown in Fig. 2. For temperatures below 100°C the emission moved continuously into the far red, and at about 75°C it was no longer detectable. As a result of loss of visual sensitivity, wavelengths in this region were not measured. The short-wavelength end of the data was limited by the inability of our oven to reach hotter temperatures. If the crystal was rotated 90° around its longitudinal axis, so as to make the pump an ordinary wave, fluorescence was no longer observed. We expect that associated with the visual-range fluorescence described above, there was also a near-infrared idler fluores-

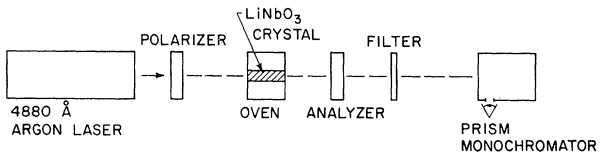

FIG. 1. Schematic of experimental arrangement.