Investigation of the Spectrum of Resonance Fluorescence Induced by a Monochromatic Field*

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Measurements of the spectrum of resonance fluorescence induced by a monochromatic laser field in an atomic beam of sodium have been made over a wide range of laser intensities for both on- and off-resonance excitation. The spectra are consistent with theoretical calculations first made by Mollow.

Recently there has been considerable theoretical[1,2] and experimental[3,4] interest in the spectrum of resonance fluorescence induced by a monochromatic field. The first experimental data, obtained by Schuda, Stroud, and Hercher,[3] showed that the strong-field spectrum consists of three peaks. Such a spectrum was also observed by Walther.[4] In this Letter we present fluorescence spectra with improved resolution for both on- and off-resonance excitation. In addition, we have observed the spectrum of elastically scattered light under weak-field excitation.

The experimental setup consists of a single-frequency cw dye laser orthogonally exciting an atomic beam of sodium. The laser-induced fluorescence from the $3^2P_{3/2}(F'=3) - 3^2S_{1/2}(F=2)$ transition is analyzed by a Fabry-Perot interferometer along a mutually orthogonal direction in an arrangement similar to that used by Schuda, Stroud, and Hercher.[3] The sodium is heated in an oven at 375°C and effuses through an aperture of 0.5 mm diameter into a high-vacuum chamber. The Na beam is collimated by another 0.5-mm aperture placed 40 cm away and intersects the laser beam 20 cm after the second aperture. The dye laser, which is similar to the one described elsewhere,[5] produces 30 mW of single-frequency power at 5890 Å. The spectral width of the laser beam is less than 250 kHz and the amplitude stability better than 2%. The dye-laser beam, which is linearly polarized parallel to the sodium beam, has a Gaussian-squared intensity distribution with a waist diameter of 2 mm at the region of interaction. Fluorescence from the entire region is monitored on a photomultiplier mounted below the intersecting beams.

The spectrum of the fluorescence emitted orthogonally to both laser and atomic beams is analyzed by a piezoelectrically scanned Fabry-Perot interferometer with a free spectral range of 300 MHz. The light transmitted through the Fabry-Perot interferometer is detected by a cooled photomultiplier, followed by photon-counting electronics. Two apertures, each 1.3 mm in diameter, are located 10 and 40 cm above the interaction region to limit the light entering the Fabry-Perot interferometer to that emitted from the central portion of the interaction region.

Special attention was given to the orthogonality of the atomic beam with respect to both the laser beam and the observation direction of the fluorescence. The laser–atomic-beam alignment was adjusted to yield the narrowest absorption linewidths [shown in Fig. 1(a)] at low laser power. The width of the individual lines is 11.5 MHz (full width at half-maximum), which is slightly broader than the 10 MHz natural width, primarily because of residual Doppler broadening in the

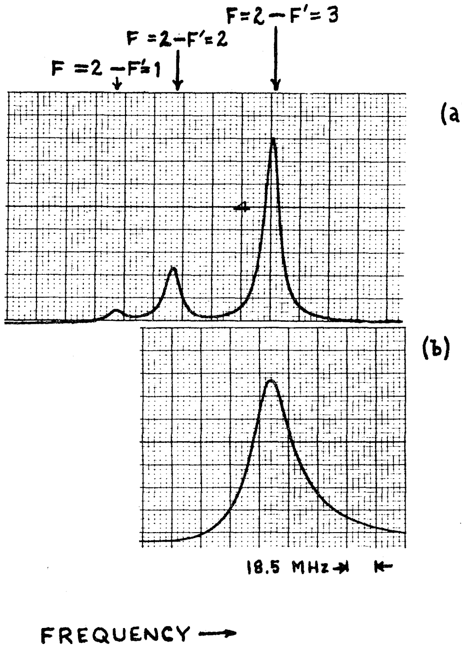

FIG. 1. Na hyperfine absorption spectrum, $3^2S_{1/2}(F=2)-3^2P_{3/2}(F'=1,2,3)$. Scan rate, 20 MHz/sec; $\tau=10$ msec; frequency scale, 18.5 MHz/large division. (a) Peak laser intensity, 0.5 mW/cm^2. (b) Power-broadened spectrum; laser intensity, 900 mW/cm^2.

atomic beam. The alignment of the fluorescence observation direction perpendicular to the atomic beam was determined in a similar manner.

To observe the on-resonance spectra, the dye-laser frequency was held fixed at the peak of the $(F=2)$-$(F'=3)$ transition with a small-bandwidth feedback loop while the Fabry-Perot interferometer was scanned at a rate of 50 MHz/min. The drift of the Fabry-Perot interferometer was less than 1 MHz/min.

Figure 2 displays a series of on-resonance spectra taken at various laser intensities (all laser-intensity measurements are ±10%). Figure 2(a) shows a 6-MHz-wide line obtained with a peak intensity of 0.8 mW/cm^2. Figures 2(b)-2(d) are spectra taken at successively higher intensities, showing contributions from both elastic and inelastic scattering. Figure 2(e) shows a symmetric three-peaked spectrum almost entirely due to inelastic scattering at a peak intensity of 920 mW/cm^2. The central peak has a width of 17.5 MHz and a height 3.2 times that of the sidebands. The sidebands are located ±42 MHz from the central peak and have widths of about 30 MHz.

In order to obtain the spectra in Fig. 2, the laser beam was centered below the observation apertures, thus ensuring the strongest and most uniform intensity distribution in the region under observation. This alignment (accurate to within 0.25 mm) was identified by the narrowest and most widely separated sidebands in the strong-field spectrum.[6] Under these conditions we estimate the effective intensity to be between 0.4 and 0.7 times the peak value.

As first pointed out by Weisskopf,[7] the spectrum of resonance fluorescence excited by a weak monochromatic field should be identical with the spectrum of the exciting field, i.e., the scattering is elastic. Thus in the low-field case of Fig. 2(a) the spectral width of the fluorescence should be that of the laser. The measured width of 6 MHz is certainly narrower than the 10 MHz natural width[8] but is greater than the laser linewidth because of the 2-MHz instrument width of the Fabry-Perot interferometer and Doppler broadening of the observed fluorescence.[9]

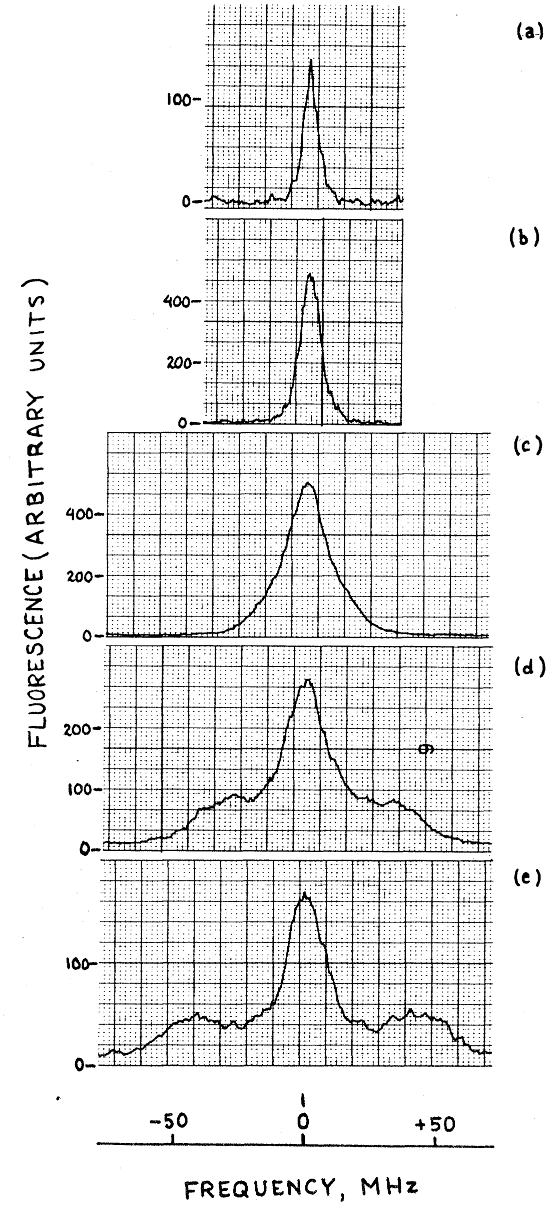

FIG. 2. On-resonance fluorescence spectra at various peak laser intensities. Scan rate, 50 MHz/min; frequency scale, 10 MHz/large division. (a) 0.8 mW/cm^2, $\tau=0.4$ sec. (b) 8 mW/cm^2, $\tau=0.4$ sec. (c) 85 mW/cm^2, $\tau=3$ sec. (d) 490 mW/cm^2, $\tau=3$ sec. (e) 920 mW/cm^2, $\tau=3$ sec.

In the strong-field case the most widely supported theory, first proposed by Mollow[1] and later substantiated by others,[2,10] predicts a symmetric three-peaked spectrum centered on the excitation frequency. The central peak has the natural width Γ and a height 3 times that of the sidebands; the sidebands have a width $3\Gamma/2$ and are displaced from the central peak by the Rabi frequency $\Omega=\mu E/h$. We have compared the spectrum in Fig. 2(e) with these predictions. From the estimate of the effective intensity mentioned eariler we expect a Rabi frequency between 40 and 53 MHz, which is consistent with the observed spectrum. The measured Rabi frequency was also in agreement with a calculated value of 38 MHz based on power-broadening measurements.

After allowance for the broadening mechanisms mentioned above, the measured height ratio and the width of the central peak are in apparent agreement with the predicted spectrum. The sidebands suffer additional broadening due to the nonuniform laser-intensity distribution and also due to the unequal dipole moments of the various $\Delta m = 0$ transitions,[11] making it difficult to compare their widths with the predicted value.

The off-resonance spectrum was observed with a peak laser intensity of 900 mW/cm^2. Figure 3 shows the spectrum for different values of detuning $\Delta\nu$. The frequency scale in this figure was derived from the power-broadened absorption line shape shown in Fig. 1(b). The laser frequency was held fixed at each value of detuning by a dc feedback loop. As the laser is detuned toward high frequencies the contribution to the spectrum from elastic scattering increases, and the sidebands separate as $[(\Delta\nu)^2 + (\mu E/h)^2]^{1/2}$. Detuning towards low frequencies shows a similar increase in sideband separation; however, the relative contributions of elastic and inelastic scattering are different from those obtained for detuning toward high frequencies. Such asymmetry with detuning was also observed by Schuda, Stroud, and Hercher,[3] and may be attributed to the presence of the neighboring hyperfine levels $F' = 2$ and $F' = 1$ in the $3^2P_{3/2}$ state.

We also checked that the off-resonance spectrum is centered on the excitation frequency by scattering some laser light into the Fabry-Perot interferometer. Figure 4(a) shows the spectrum for a detuning of -15 MHz and Fig. 4(b) shows

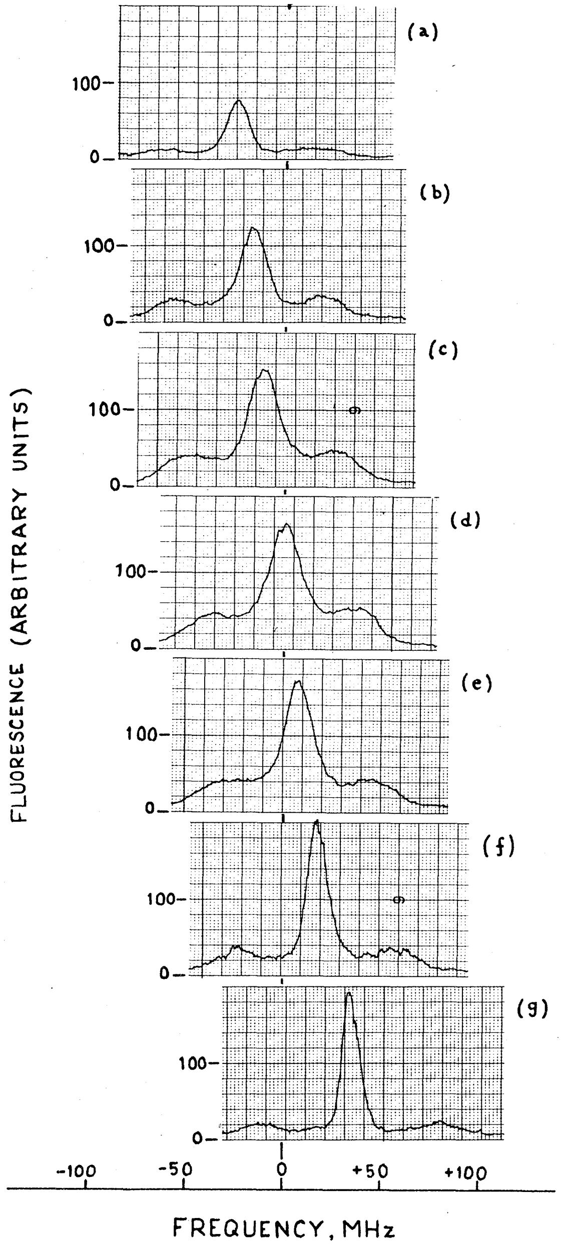

FIG. 3. Off-resonance fluorescence spectra for various detunings. Peak laser intensity, 900 mW/cm^2. Scan rate, 50 MHz/min; frequency scale, 10 MHz/large division. (a) -26 MHz, $\tau = 3$ sec. (b) -17 MHz, $\tau = 3$ sec. (c) -11 MHz, $\tau = 3$ sec. (d) near resonance, $\tau = 3$ sec. (e) $+7$ MHz, $\tau = 3$ sec. (f) $+17$ MHz, $\tau = 1$ sec. (g) $+34$ MHz, $\tau = 1$ sec.

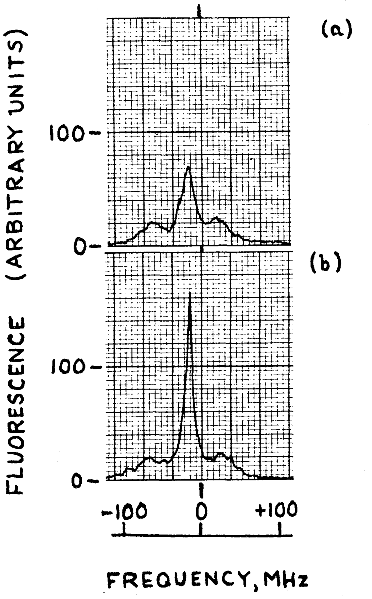

FIG. 4. (a) Fluorescence spectrum, -15 MHz detuning. Peak laser intensity, 900 mW/cm^2. Scan rate, 100 MHz/min; frequency scale, 37 MHz/large division; $\tau = 1$ sec. (b) Same as (a) but including scattered laser light.

the simultaneous recording of this spectrum and that of the scattered laser light. The deviation of the center of the spectrum from the laser frequency is within the Fabry-Perot-interferometer width of 2 MHz.

Although none of the theories predicts any asymmetry in the heights of the side peaks, we have observed a slight, but reproducible, asymmetry.[12] The low-frequency peak was consistently smaller than the high-frequency peak by about 5-10%. A similar, but more severe, asymmetry has been observed by Schuda, Stroud, and Hercher[3] and Walther.[4] The effect of neighboring hyperfine levels may be eliminated by a scheme suggested by Abate[13] for the preparation of atomic sodium as a two-level system.

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[6] This alignment is not the one that gives the most fluorescence from the observed region. We calculated and verified that optical pumping to the $F=1$ ground state depletes the $F=2$ population in the maximum-field region by a factor of about 2.5 (for 920 mW/cm^2).

[7] V. Weisskopf, Ann. Phys. (Leipzig) 9, 23 (1931).

[8] H. M. Gibbs and T. N. C. Venkatesan, post-deadline paper presented at the Conference on Laser Engineering and Applications, Washington, D. C., 28-30 May 1975 (unpublished).

[9] In the weak-field case the spatial distribution of the fluorescence is the same as that of the laser intensity. Therefore the Doppler broadening is less than that determined by aperture geometry.

[10] G. Oliver, E. Ressayre, and A. Tallet, Lett. Nuovo Cimento 2, 777 (1971); E. V. Baklanov, Zh. Eksp. Teor. Fiz. 65, 2203 (1973) [Sov. Phys. JETP 38, 1100 (1974)]; H. J. Carmichael and D. F. Walls, J. Phys. B: At. Mol. Phys. 8, L77 (1975); S. S. Hassan and R. K. Bullough, J. Phys. B: At. Mol. Phys. 8, L147 (1975).

[11] Broadening from this effect is not as severe as might be expected, since optical pumping transfers most of the atoms into the $m_F=0, \pm 1$ sublevels, which have approximately equal matrix elements for $\Delta m=0$ transitions.

[12] This cannot be explained by fluorescence from the transition $F'=2$ to $F=1$ which we found negligible except for large negative detuning.

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Vibrational Excitation in Benzene by Electron Impact via Resonances: Selection Rules*

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The vibrational excitation in the energy ranges of the lowest resonances in benzene has been measured and it is found that only few fundamental modes become excited. This selectivity of the excitation process can be understood in terms of selection rules which are based on symmetry considerations. The angular distribution of scattered electrons in the range of the lowest shape resonance (~1.2 eV) is consistent with an f_{δ} wave as the dominant partial wave.

In this Letter we present experimental measurements on the vibrational excitation in benzene (C_6H_6 and C_6D_6) by electron impact at low energies. We consider the experimental observations presented here as prototypes for similar processes in the larger molecules. In particular, we study the vibrational modes excited by electron impact in the regions of the lowest shape resonances at energies of about 1.2 and 4.8 eV. Despite the large number of fundamental vibrational modes in which the benzene molecule can vibrate, *only very few modes are observed.* In order to explain this selective excitation, we develop a set of *selection rules* for vibrational excitation via resonances. These selections rules are based on the symmetry of the resonances and a few additional, physically reasonable postulates.

We find that the angular distribution of elec-