Heater coils were of platinum and gold. The yield, Φ_f, is given as

$$\Phi_f = \left(\frac{\bar{1}}{\lambda}\right) \frac{\int(dQ/d\lambda)d\lambda}{hc\int(dQ/d\lambda)d\ln\lambda} \cdot \left(1 - \frac{E_Q}{E_K}\right)$$

where $(dQ/d\lambda)$ = quanta s^{-1} nm^{-1} emitted at λ. After correction for self-absorption, the result is $\Phi_f = 0.561 \pm 0.039$.

The quantum yield of fluorescence of azulene (in degassed cyclohexane) irradiated at 283 nm (100 Å band pass) was determined by comparison with quinine bisulphate. Our result is $\Phi_f = 0.024$ for $S_2 \leftarrow S_o$ emission. Similarly, our measured value of Φ_f for 9,10-diphenylanthracene (in degassed cyclohexane) irradiated at 365 nm, is 0.84 at 25 °C.

$\eta 2$

Two-Photon Doppler Free Spectroscopy of Gas Phase Molecules

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Diffuseness in the gas phase electronic spectra of complex molecules caused by the Doppler effect has been a significant limitation to investigation of the excited state. This paper describes application of the technique of counter-propagating beam two-photon absorption [1] to molecular electronic spectroscopy. Doppler free spectra have been obtained for the nitric oxide $(A)^2\Sigma^+(v = o) \leftarrow (X)^2\Pi_{3/2}$ $(v = o)$ transition. The measured linewidth of 250 MHz. ($8 \times 10^{-3} cm^{-1}$) at 3 Torr was instrument limited and represented about one-tenth the Doppler width.

The ungerade vibrational levels of the benzene $^1B_{2u}$ state are being investigated at ultra-high resolution to evaluate the possibility of direct spectral manifestations of the interactions responsible for non-radiative decay in the isolated molecule. The data will also serve to evaluate the selectivity of the method for photoexcitation of single rovibronic levels in a complex spectrum. It will establish the ability to detect and identify large molecules in the gas phase and to characterize the state of vibrational and rotational excitation.

Experiments are being conducted which will test the effects of excitation coherence on radiative and radiationless decay of excited states. They rely on the ability to excite isolated molecule compound states with light of bandwidth comparable to the spectral width of the states, and to measure absorption spectra with a resolution commensurate with the product state decay time.

These experiments have been accomplished using an excitation source that combines a dye laser oscillator of the Hansch configuration [2] with an external Fabry-Perot interferometer that is piezoelectrically scanned. Output of this system is amplified and then passed through focus in a sample cell and on to a spherical mirror which returns the beam to focus in coincidence with the incoming beam. Absorption is detected by measurement of ultra-violet luminescence using a solar blind photomultiplier.

1 L. S. Vaslenko, V. P. Chebotaev, A. V. Shishaev, JETP Letters, 12 (1970) 113.
2 T. W. Hansch, Appl. Opt., 11 (1972) 898.