

Peterson N C, Kurylo M J, Braun W, Bass A M & Keller R A. Enhancement of absorption spectra by dye-laser quenching. *J. Opt. Soc. Amer.* 61:746-50, 1971. [National Bureau of Standards, Washington, DC]

Insertion of weakly absorbing samples into the cavity of a broad-band dye laser enhances photographic detectability by over two orders of magnitude in a 1 μ s laser pulse. Fluorescence is not observed from iodine vapor in a dye laser optical cavity. [The SCI® indicates that this paper has been cited in over 110 publications since 1971.]

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During the academic year 1969-1970, I was on sabbatical leave from the Polytechnic Institute as a visiting scientist at the US National Bureau of Standards. Walter Braun, Mike Kurylo, and I were concerned with atomic reaction rate measurements via resonance fluorescence experiments. I recall an intense conversation with Braun one evening in the fall of 1969. We discussed finding an experimental method for kinetic study of free radical reactions that would have advantages over the photographic technique of Norrish and Porter.^{1,2} I succeeded in convincing Braun that dye-laser quenching might be the ultimate in a multi-pass absorption experiment. The possibility of high sensitivity and short temporal resolution prompted an experimental test of the actual sensitivity. Ed Zalewski and Dick Keller had speculated previously that there should be enhanced absorption in that type of measurement, but they had not done the experiment.

We observed absorption enhancement by May 1, 1970. This was especially gratifying

because a distinguished visitor to the laboratory previously had pontificated that there should be no effect.

I built a dye laser to permit investigation of the effect without further disturbing the activities in Keller's laboratory. It was modeled on an article in the Amateur Scientist column of *Scientific American*. Al Ledford gave considerable help with the mechanical aspects of laser construction, and Keller was very free with advice. The laser functioned on the second try. I enjoyed this work while waiting to see if the administration of the Polytechnic Institute would allow me to have tenure.

For amusement, we coined the acronym DASER to describe the phenomenon, suggesting "Darkness Amplification...." As DASER had already been preempted by an unrelated commercial product, it was dropped.

Why the large frequency of citations? They refer to the report of a new, sensitive spectroscopic method with many applications to real chemical problems. This was the first report of intracavity enhanced absorption using a homogeneously broadened dye laser having a bandwidth of 2-10 nm. That initial work confirmed that the experiment should be useful for detecting low concentrations of short-lived free radicals having absorption spectra overlapping the emission spectrum of the dye laser. Later work demonstrated improved temporal resolution of 100 ns,³ developed a theoretical computer model of the effect,³ demonstrated detection of the NH₂ radical,⁴ and observed reactions of the HCO radical.⁵ Enhancements of 10⁵ and the quenching of I₂¹²⁷ fluorescence by intracavity absorption were soon reported.⁶ An unexpected application was the detection of absorbing species in a flame.⁷ There are now many reports of intracavity laser absorption of combustion gases. The technique has been suggested as a possible molecular beam detector.⁸

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