This paper reported the first observation of continuous-wave and pulsed-laser action on vibrational-rotational transitions of molecular species. [The SCI® indicates that this paper has been cited in over 160 publications since 1964.]

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I had just arrived at Stanford University in 1958 to start my graduate studies when Schawlow and Townes published their pioneering paper describing the concept of the laser. My PhD thesis research had no connection with lasers. By the time I came to Bell Labs in 1961 after finishing my graduate work, the ruby laser at 6943 Å and the helium-neon laser at 1.15 µm had already been demonstrated. Some felt that there was nothing more to be done in the discovery of new laser systems while others felt that this was not even the beginning. It was under these circumstances that I joined the laser community.

I had worked on various atomic gas lasers since 1961 and had discovered a host of new atomic laser transitions in noble gases and in oxygen from 8446 Å to 133 µm with a number of my colleagues. I became convinced by mid-1963 that the atomic laser transitions known then could not be candidates for high-power lasers. And among the laser aficionados, gas lasers were relegated to the also-ran status for producing large amounts of coherent optical powers for various applications. The apparent winners were the solid-state lasers. I decided to begin exploring the possibility of laser action on vibrational-rotational transitions of molecules. For a number of reasons (including the nearness of the possible lasing levels to the ground state) it became clear to me that such a laser could be a high-power and high-efficiency system. The first gas I chose to study was CO₂. I tuned the spectrometer to the vicinity of 10.6 µm, and the CO₂ laser worked the first time. My associate Rudy Kerl and I tried it. The field of vibrational-rotational molecular gas lasers was born.

The paper singled out here was my complete description of laser action on vibrational-rotational transitions of a molecule, but the first brief report had appeared in April 1964 in Physical Review Letters. I gave a calculation of gain on vibrational-rotational transition of molecules and explained the observations of the first CO₂ laser. Within a few months, I realized that, to increase the power output, I needed a selective excitation mechanism. Having just read Morgan and Schiff's paper on quenching of vibrationally excited nitrogen by CO₂ and N₂O, I concluded that nitrogen was the right donor molecule. The N₂-CO₂ laser results were published in December 1964 and from that point the power output doubled every couple of weeks as I understood the effects of various added gases. By the middle of 1965, my colleagues and I had achieved more than 100 W of continuous-wave power output. No other laser had achieved this at that time. Different excitation mechanisms such as transverse electrical discharge, chemical transfer pumping, and fluid dynamic pumping followed. The high power easily available in the 10 µm region had made the CO₂ laser the workhorse in infrared nonlinear optics, optical pumping, industrial processing, laser surgery, and so on.

A final comment on my discoveries about laser action is that the ideas were found to be incredibly general. I was associated with the discovery of vibrational-rotational lasers in hydrogen fluoride and deuterium fluoride. The last two systems have found applications in defense. And the use of CO₂ and N₂O lasers as the pump for a variety of far-infrared laser transitions in molecules has made that region very rich with discrete laser transitions. It is easy to see why this paper is a Citation Classic and to gauge the impact of the CO₂ laser. It opened up the field of a completely new class of lasers that have become very important. The CO₂ lasers alone are expected to account for more than $100 million in sales in 1985.