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INFRARED RESONANCE OF OH WITH THE H_2O LASER: A GALACTIC MASER PUMP?

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The 79-μm electric dipole spectrum of OH ($^2\Pi_{3/2}, J=\frac{3}{2} \leftrightarrow {}^2\Pi_{1/2}, J=\frac{1}{2}$) has been measured by a magnetic-resonance absorption method, with a water-vapor laser as the source oscillator. Corresponding magnetic dipole spectra are calculated from the data. A near overlap of the water line on one magnetic-dipole transition suggests a possible pumping mechanism for the 18-cm maser emission of stellar OH.

In discussions of galactic maser models[1] one optical pumping process, well known in the laboratory,[2] appears to have been overlooked. This is the pumping caused by the partial overlap of an emission line of one molecule on an absorption line of another. When the absorption line has resolved structure a variety of pumping situations, sensitive to small Doppler shifts, can occur. Line pumping is in fact so critically dependent on the structure and degree of overlap of the two lines that astronomical speculation, even when velocity information is available, is difficult unless based on precise laboratory measurements of the line frequencies. This paper describes such a measurement of an accidental overlap in the far-infrared spectra of OH and water vapor; it was prompted by the growing evidence of an association of certain galactic OH masers and water emission sources with infrared stars.[3,4]

Two accidental line coincidences, more or less exact, are evident when a diagram of low-lying OH energy levels[5] is compared with a table of radiations emitted by the water-vapor laser.[6] The 79.1-μm laser line coincides with the $^2\Pi_{3/2}$, $J=\frac{3}{2} \leftrightarrow {}^2\Pi_{1/2}$, $J=\frac{1}{2}$ transition of OH, and the 34.6-μm laser line coincides with the $^2\Pi_{3/2}$, $J=\frac{3}{2}$ $\leftrightarrow {}^2\Pi_{1/2}$, $J=\frac{5}{2}$ transition of OH. We compare against the laser spectrum of water rather than its entire infrared spectrum for two reasons, one of which is the large energy requirement of an astronomical maser and the other of which is a matter of experimental convenience. The experimental reason is that the laser, used as an absorption source, makes easy what would otherwise be a difficult experiment, that of measuring just where the water line is relative to the several fine-structure and hyperfine-structure components of the OH transition. The laser line is tunable to only a limited degree, but a larger tuning range, enough to scan the entire OH spectrum past the laser-line frequency, is provided by the Zeeman effect of OH. This technique, called laser electron paramagnetic resonance, already has been tested on the oxygen[7,8] molecule, using the 337-μm hydrogen cyanide laser, and we have used much the same methods to measure the 79.1-μm transition of OH. The water laser was 4 m long and of the Michelson interferometer type,[9] with OH at a few Torr pressure contained in one side arm. The OH was made by mixing atomic hydrogen with nitrogen dioxide.

The observed 79-μm magnetic resonance spectrum of OH is shown by Fig. 1. Orientations of the magnetic field both perpendicular and parallel to the linear polarization of the laser are represented. These are electric dipole transitions,

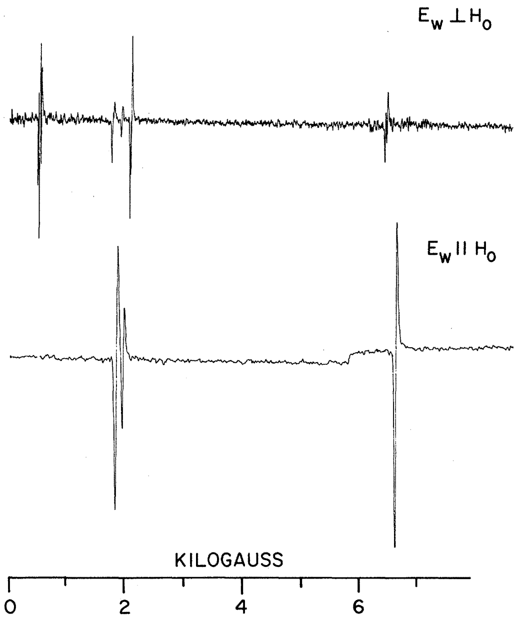

FIG. 1. Infrared magnetic resonance absorption spectrum of OH. Source: 79-μm H_2O laser.

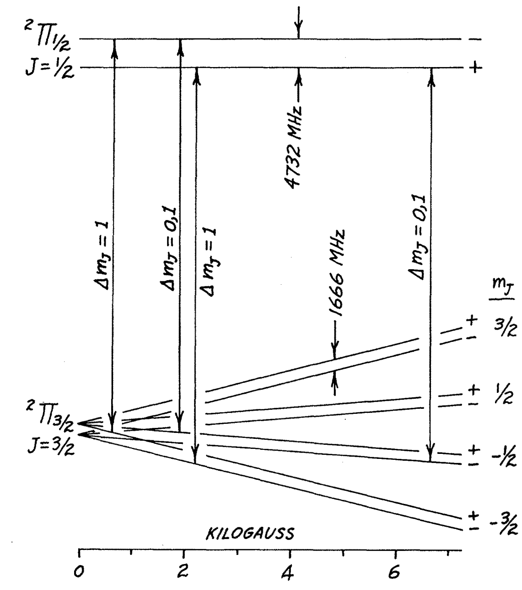

FIG. 2. Identification of transitions in Fig. 1. Hyperfine structure, and small Zeeman splittings of the upper energy levels, are omitted. Drawn to scale, except for infrared energies.

normally forbidden between members of a spin multiplet but allowed in OH because of large rotational-distortion effects. The splitting into groups of lines at four different field strengths is due to Λ-type doubling and the Zeeman effect of the $^2\Pi_{3/2}$ level, as is demonstrated by the energy diagram of Fig. 2. Hyperfine structure and the small Zeeman effect of the $^2\Pi_{1/2}$ level, omitted from Fig. 2 for simplicity, account for the small splittings within each group of lines. All of these effects have been measured before in microwave absorption experiments on the two levels separately.[10] The only adjustable parameter left to be fitted to the spectrum is the net frequency difference between the OH and H_2O infrared transitions. A numerical fit, using magnetic resonance data in Table I, yields the OH transition frequencies, for zero-field conditions, given in Table II. Both electric and magnetic dipole transitions are listed; the latter were too weak to be detected in the magnetic resonance spectrum, but their frequencies are determined uniquely by the electric dipole spectra and the known Λ-type doubling frequencies.

The laser frequency can be pulled slightly by varying the tuning controls, and this is probably the main source of uncertainty in the frequencies of Table II. The 79-μm laser line has not been measured accurately yet, but the 78-μm line has[11] and it is found that a resetting error of ±3 MHz is possible. We double this to arrive at

Table I. Measured line positions, relative intensities, and polarizations in the infrared magnetic resonance absorption spectrum of OH. Source: 79-μm H_2O laser.

H (G)	I (relative)	Polarization
	Lines 6 G wide[a]	
620	7	⊥
641	15	
665	7	
2181	2	⊥
2193	11	
2208	12	
	Lines 18 G wide[b]	
680[c]	0.1	∥
1870	8	∥
1890	14	
2004	8	
1877	3	⊥
1903	0.7	
2041	1.5	
6501	5	⊥
6533	5	
6563	22	∥

[a] Uncertainty in location of line centers: ±2 G.
[b] Uncertainty in location of line centers: ±4 G except for the 680-G line, which is ±6 G.
[c] Unidentified, but probably a magnetic dipole line.

Table II. Derived zero-field frequencies of the 79-μm absorption spectrum of OH, relative to the 79-μm laser line of H_2O.

Transition type	Parity	$F'' \to F'$	$\nu_{OH}-\nu_{water}$ (MHz)[a]
Electric dipole	$+\to-$	$2\to1$	-1261
		$1\to0$	-1296
		$1\to1$	-1205
	$-\to+$	$2\to1$	-4344
		$1\to0$	-4306
		$1\to1$	-4291
Magnetic dipole	$+\to+$	$2\to1$	-6011
		$1\to0$	-5971
		$1\to1$	-5956
	$-\to-$	$2\to1$	407
		$1\to0$	370
		$1\to1$	460

[a]Experimental uncertainties are ±6 MHz.

what we believe is a conservative error estimate of ±6 MHz for the OH frequencies, relative to the "true" 79-μm laser frequency. The OH linewidths are 12 MHz (6 G for the $|m_J| = \frac{3}{2} \to \frac{1}{2}$ lines and 18 G for the $|m_J| = \frac{1}{2} \to \frac{1}{2}$ lines) which is just that expected from Doppler broadening. Errors in locating line centers and measuring the corresponding magnetic field strengths do not exceed ±3 MHz in equivalent frequency units, and so are relatively unimportant.

Further work is being done to extend the laser resonance technique to other transitions of OH and other magnetic molecules. The 119.0-μm OH transition $^2\Pi_{3/2}$, $J=\frac{3}{2} \to J=\frac{5}{2}$ has been observed at high field strengths, excited by the 118.6-μm H_2O laser line, and several spectra of NO and NO_2 have been measured.

What this experiment says about the possibility of line pumping of OH is summarized in Fig. 3. A relative Doppler velocity of about 30 km/sec will put the water line amid the hyperfine structure of the magnetic dipole transition that connects the lower Λ-type doubling level of the $^2\Pi_{3/2}$, $J=\frac{3}{2}$ ground state with the upper $^2\Pi_{1/2}$, $J=\frac{1}{2}$ level. Spontaneous electric dipole transitions connect this level with the upper Λ-type doubling level of the ground state, and the net result is a transfer of molecules from the lower (negative-parity) levels of the ground state to the upper (positive-parity) levels of the ground state. The resulting maser emission will be concentrated in the main hyperfine lines ($\Delta F=0$) at 1665 and 1667 MHz, with weaker emission possible in the 1720-MHz satellite line ($F=2\to1$). Although the

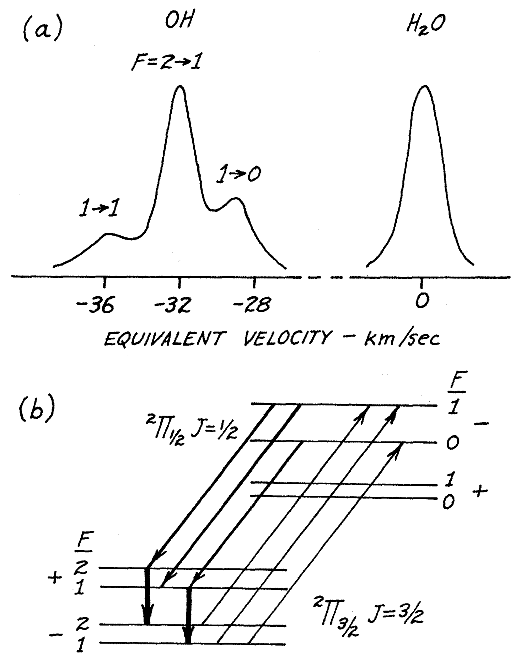

FIG. 3. (a) Sketch of the 79 μm $-\to-$ magnetic dipole line of OH, at zero field, relative to the 79-μm H_2O laser line. Frequency differences have been converted to equivalent Doppler velocities, and a linewidth of 2 km/sec has been assumed. (b) Line pumping transitions in OH. Upward arrows represent magnetic-dipole absorption. Downward arrows represent spontaneous electric-dipole emission. The principle maser emission lines at 1665 MHz ($F=1\to1$) and 1667 MHz ($F=2\to2$) are indicated.

characteristic OH emission of infrared stars is in the 1612-MHz satellite line ($F=1\to2$), many stars also show some main-line emission and in certain ones it is both strong and displaced in velocity from the satellite-line emission. Quite possibly this range of OH emission properties in infrared stars can be accounted for by a mixture in varying proportions of continuum pumping and line pumping, the former giving satellite-line emission[1] and the latter main-line emission. In another large class of OH masers (type I), main-line emission is dominant, and it is perhaps significant that some masers in this class also radiate from excited rotational levels. The 79.1-μm resonance can populate and invert only its own excited level, but the 34.6-μm resonance can, by radiative cascade, populate and invert several others as well. Since in all presently known sources the excited emission comes from two or more levels at once, and in the same range of velocity, it may be that the 34.6-μm resonance is the better maser pump. Unfortunately, we were unable to measure this resonance with our present cw water laser.

The inversion process in laboratory water lasers is not known, but it no doubt involves selective excitation by collision. Possibly the same process operates in shock fronts or other locally dense and disturbed regions of interstellar gas, thereby powering type-I masers, but there is also a simpler laser inversion process that can account reasonably well for the main-line OH radiation from infrared stars. This is a radiative inversion, drawing its energy from the 2.8-μm blackbody radiation of the star and operating through the special properties of perturbed vibration-rotation energy levels in water vapor. The upper level of the 79.1-μm laser transition, because it is perturbed, has a much larger probability of excitation from the ground state by 2.8-μm radiation than does the lower level. The same holds for several other pairs of laser levels. Hence population inversions can be maintained in water vapor near an infrared star, and amplification of the star's radiation at 79.1 μm and other discrete wavelengths will result. Under saturated laser conditions, one 79.1-μm photon will be emitted for each 2.8-μm photon absorbed in lines that populate the upper laser level. Used in turn to pump the OH maser, each 79.1-μm photon will be converted to an 18-cm photon. With good geometry, correct velocities, and sufficient optical depth in the infrared absorptions, the overall efficienty of this process can be as much as 100 times greater than that of Litvak's near-infrared continuum pumping process.[1] The condition most difficult to satisfy may be that of sufficient optical depth, since the absorption coefficient of the magnetic-dipole pumping transition is 300 times smaller than that of the weakest pumping transitions in Litvak's scheme. For an OH region of given size and density the orders of magnitude cancel, and it is probably fair to say that the two pumping processes can have about equal efficiency.

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MONOCHROMATIC RADIATION FROM A COHERENT MODULATED BEAM OF CHARGED PARTICLES

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It is suggested that intense laser fields may be used to modulate the wave functions of electrons. A classical model is used to demonstrate the greatly altered radiation fields of these electrons after they emerge from the laser beam. The observation by Schwarz and Hora of visible radiation from a nonluminescent target appears to result from these fields.

With the advent of laser techniques, it has become possible to modulate a particle beam at laser frequencies by a method similar to that employed in the conventional klystron. This rapid modulation introduces the possibility of observing effects associated with the longitudinal coherence length of individual particles. [This length is a measure of the size of a particle's wave packet in the direction of the beam.] If the pulse length in the modulated beam is long compared with the coherence length, the only possible effect of the modulation is the physical bunching of the particles in the beam; any subsequent radiation from them may be treated in the convention-