INFRARED SPECTROSCOPY USING STIMULATED EMISSION TECHNIQUES

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Optical maser oscillation has been obtained using electron excitation in each of the noble gases and on a total of 14 transitions falling in the wavelength range from 1.5 to 2.2 microns. Of these transitions, three have not previously been reported in spontaneous emission.[1] A summary of results is contained in Table I. Previously reported gas discharge masers[2,3] have depended upon less generally applicable excitation methods, and the attainment of maser oscillation at these 14 wavelengths by electron impact suggests that many other similar transitions can be made to oscillate over a wide wavelength range. The brightness and coherence length characteristic of an optical maser beam should permit measurement of term differences with an accuracy unobtainable with fluorescent sources and, further, should permit the establishment of substantially improved wavelength standards in the infrared.

The fluorescent spectrum of the inert gases in the lead sulfide region (up to about 3μ) has been subjected to painstaking study.[4-9] With the use of evacuated spectrometers, cooled detectors, and phase-sensitive electronics, most of the expected term differences in the lower-lying levels have been observed. However, the attainable signal-to-noise ratios have been limited by lack of detectors with sensitivity comparable to those in the visible region and by the natural weakness of spontaneous emission towards the longer wavelengths. It is probable that some lines have been missed entirely because of inadequate signal. More significantly, resolution is often limited by weakness of the signal.[8] However, for transitions on which maser oscillation can be obtained, the signal is increased by many orders of magnitude. Further, by operating the maser on a single cavity mode,[10] the oscillation frequency is automatically set within half the longitudinal mode spacing from the peak of the line (e.g., for the

Table I. Transitions in helium, neon, argon, krypton, and xenon observed under stimulated emission. Identity of the 1.793-micron line in argon has not been established. Relevant maser data are given for the strongest transitions.

Gas	λ_{air} (microns)	Transition	Frequency (cm^{-1}) measured	Frequency (cm^{-1}) calculated[a]	Previous emission data	Optimum pressure (mm Hg)	Gain (% per meter)	Beam power (mW)
Helium	2.0603	$7\,^3D - 4\,^3P$	4852.3	4852.58	---	8	5	3
Neon	2.1019	$4d'[5/2]^0_2 - 4p[3/2]_2$	4756.3	4756.61	---	0.2	3	1
Argon	1.618	$5s[3/2]^0_2 - 4p'[3/2]_2$	6178	6178.80	4	0.05		
	1.694	$3d[3/2]^0_2 - 4p[3/2]_2$	5900	5901.38	4,5	0.035	3	0.5
	1.793	$3d[1/2]^0_1 - 4p[3/2]_2$ or $3d[1/2]^0_0 - 4p[3/2]_1$	5577	5580.49 or 5580.56	4	0.035		
	2.0616	$3d[3/2]^0_2 - 4p'[3/2]_2$	4849.3	4849.23	6,7	0.035	3	1
Krypton	1.690	$4d[1/2]^0_1 - 5p[1/2]_1$	5916	5916.69	4,5	0.07		
	1.694	$4d[5/2]^0_2 - 5p[3/2]_1$	5902	5903.03	4,5	0.05		
	1.784	$4d[1/2]^0_0 - 5p[1/2]_1$	5603	5603.00	4,5	0.07		
	1.819	$4d'[5/2]^0_2 - 5p'[3/2]_2$	5497	5497.49	4,5	0.07		
	1.921	$8s[3/2]^0_1 - 6p[5/2]_2$	5205	5203.86	---	0.035		
	2.116	$4d[3/2]^0_2 - 5p[3/2]_1$	4725	4723.39	6	0.035	3	1
	2.189	$4d[3/2]^0_2 - 5p[3/2]_2$	4566	4564.44	6	0.035	3	1
Xenon	2.0261	$5d[3/2]^0_1 - 6p[3/2]_1$	4934.2	4933.93	5	0.02	10	5

[a]C. E. Moore, Atomic Energy Levels, National Bureau of Standards (U. S. Government Printing Office, Washington, D.C., 1952).

maser used in this work, this mode spacing is about 33 Mc/sec). Also, the increase in signal should aid the investigation of transitions beyond 3μ, where presently available detectors have still less sensitivity than that attainable in the lead sulfide region.

The data shown in Table I were taken with a medium resolution ($\cong$1:5000) grating instrument and are intended primarily to illustrate the validity of the method. Although the accuracy of these measurements is not adequate to improve the known term values, it is sufficient to establish identification of the transitions in all cases but one. The maser used for the present measurements consisted of a 225-cm length, 7-mm i.d. quartz discharge tube equipped with Brewster angle windows[11] and external, confocal mirrors. A more detailed description of the apparatus will be given elsewhere.[12]

With the exception of helium, the electron configurations in the noble gases are similar. The ground states in Ne, Ar, Kr, and Xe consist of closed-shell, $(np)^6$ configurations where $n=2$, 3, 4, and 5, respectively. In each case, strongly allowed vacuum-ultraviolet transitions connect the ground state with levels in the $(np)^5ms$ and $(np)^5md$ configurations ($m>n$). As is well known, these same excited levels will have the largest electron excitation cross sections and large cross sections are also to be expected for neighboring levels within the same configuration from electron exchange. For the pressures used, photons emitted in the strong vacuum-ultraviolet transitions are completely trapped. Hence, all four levels in the $(np)^5(n+1)s$ configuration become metastable and electrons excited to the higher s and d shells decay primarily by radiation through transitions to terms in the lower p shells. The latter decay with strong transitions in the visible and near-visible range to the metastable states. The relative transition probabilities and excitation cross sections are such, therefore, as to permit obtaining population inversions in low-pressure discharges on many of the $s\rightarrow p$ and $d\rightarrow p$ transitions of Ne, Ar, Kr, and Xe in the infrared.

The mere production of a population inversion is, of course, not adequate for the present purposes; the excited-state densities must be sufficient to produce oscillation in a cavity of practical dimensions. The initial research on the He-Ne maser[2] indicated that both excitation of the lower neon maser levels by electron impact on neon metastables and resonance trapping by neon metastables would prevent obtaining the required inversions in a pure discharge. Although such processes involving the metastable states limit the inversion in the present systems, the effects are not as serious as initially thought. In the case of pure neon, optical gain[13] and maser oscillation[12] have been obtained previously on the $4s'[1/2](J=1)$ to $3p'[3/2](J=2)$ transition at 11 523 Å. Careful attempts to obtain oscillation on the analogous transitions of Ar, Kr, and Xe have yielded negative results so far. As is evident from Table I, oscillation in the heavier noble gases has been obtained almost entirely on transitions between d and p configurations. The latter is not too surprising since the d shells become more and more depressed in respect to the appropriate s shells in going from Ne to Xe and require increasingly less excitation energy. In fact, it is only in the case of neon that the s shells appropriate for maser action fall below the d shells. Of the transitions observed, the strongest oscillation was obtained in the case of xenon. Here, the addition of a moderate pressure of helium or neon improves the maser action—presumably by increasing the density of high-energy electrons.

In helium the case is somewhat different. Oscillation was obtained on the $7\,^3D\rightarrow 4\,^3P$ transition at 20 608 Å. Masking by the strong, neighboring $2\,^1P-2\,^1S$ transition at 20 580 Å may have prevented previous observation of this line in the fluorescent spectrum. Direct excitation by electron impact with ground-state helium atoms and $2\,^1S$ metastables goes almost entirely to the $n\,^1P$ levels. A number of indirect excitation mechanisms lead to an efficient production of the $2\,^3S$ metastable level, and electron collisions with this metastable can result in a large production of $n\,^3P$ states at high discharge intensities. It seems apparent, however, that no direct electron excitation process can lead to a high yield of $7\,^3D$ atoms. The most probable source of $7\,^3D$ excitation in the present case consists of excitation transfer from the $7\,^1P$ through collisions with ground-state helium atoms in which the Wigner spin-conservation rule is violated. Processes of this type have been reported earlier by Maurer and Wolf[14] for initial states as low as the $3\,^1P$. More recent studies,[15] however, indicate that such processes are only important in the higher-lying states of helium and that they may occur largely through intermediate steps involving the F levels. In any case, the high pressures and discharge con-

ditions required for optimum output on the $7\,^3D$-$4\,^3P$ transition are compatible with an indirect process of this type. As with the other noble gases, the metastable densities may interfere with the inversion through resonance trapping of the $4\,^3P$-$2\,^3S$ transition and electron excitation of the $4\,^3P$ term. Indirect support of this conclusion has been obtained by observing that a slight trace of nearly any impurity will enhance the oscillation, presumably through ionizing collisions with the $2\,^3S$.

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[2]A. Javan, W. R. Bennett, Jr., and D. R. Herriott, Phys. Rev. Letters 6, 106 (1961).

[3]W. R. Bennett, Jr., W. L. Faust, R. A. McFarlane, and C. K. N. Patel, Phys. Rev. Letters 8, 470 (1962).

[4]C. J. Humphreys and H. J. Kostkowsky, J. Research Natl. Bur. Standards 49, 73 (1952).

[5]W. R. Sittner and E. R. Peck, J. Opt. Soc. Am. 39, 474 (1949).

[6]E. Paul, Jr. and C. J. Humphreys, J. Opt. Soc. Am. 49, 1186 (1959).

[7]G. Hepner, Compt. rend. 248, 1142 (1959).

[8]G. Hepner, Ann. Phys. 6, 735 (1961).

[9]C. J. Humphreys, E. Paul, Jr., and K. B. Adams, NAVWEPS Report 7205, 1961, U. S. Naval Ordnance Laboratory, Corona, California (unpublished).

[10]See W. R. Bennett, Jr., Phys. Rev. 126, 580 (1962).

[11]W. W. Rigrod, H. Kogelnik, D. J. Brangaccio, and D. R. Herriott, J. Appl. Phys. 33, 743 (1962).

[12]C. K. N. Patel (to be published).

[13]W. R. Bennett, Jr., Bull. Am. Phys. Soc. 7, 15 (1962).

[14]W. Maurer and R. Wolf, Z. Physik 92, 100 (1934); 115, 410 (1940).

[15]C. C. Lin and R. G. Fowler, Ann. Phys. (New York) 15, 461 (1961).

POLARIZATION IN PROTON-He^4 SCATTERING AT 38 MeV*

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With the polarized proton source[1] which was developed for the Minnesota linear accelerator, the polarization of the protons scattered by He^4 at 38.4 MeV was measured.

The incident 39.9-MeV polarized proton beam was focussed to a spot $\frac{3}{32}$ in. wide and $\frac{1}{4}$ in. high by a pair of quadrupole magnets on the liquid helium target. The experimental setup and procedures used here and other experiments which have been done with solid targets will be reported in more detail in the near future. The liquid helium target was contained in a 1.7-cm diameter by 4-cm high cyclinder made of 1-mil Mylar, which was attached to a 2.3-liter reservoir. The whole assembly was surrounded by a liquid nitrogen heat shield, which made possible 14 hours of experimental observation for each liquid helium filling. The energy loss in the liquid helium target was 3.2 MeV so that the mean energy was 38.3 ± 0.5 MeV. Two identical NaI crystal counters, each subtending a fixed solid angle to the target, were used as detectors. The angular resolution was $\pm 1.5°$ to $\pm 2.3°$, the variation depending on the finite thickness of the target and the angle of scattering. The beam intensity was about 1.5×10^6 protons per second with a maximum polarization of 38%.

The orientation of the beam polarization can be readily changed to up or down by reversing the direction of the spin-orienting magnetic field in the ionizer of the polarized source. This means that the asymmetry measurements can consist of a pair of runs with the beam polarization up and down instead of a left-right asymmetry measurement with fixed polarization direction. This makes it possible either to set both counters at different angles on the same side of the incident beam or to set them at the same angle on the opposite side of the beam. Measurements made with the two configurations for the same scattering angle agree with each other within statistical errors. Beam polariza-