reflections, those indexed as 200 were very weak. The average interplanar spacings, as determined from eleven photographs of helium at 1.73°K and 29.01 atm, are

$$d_{110} = 2.911 \pm 0.009\ \text{Å}$$

and

$$d_{200} = 2.053 \pm 0.005\ \text{Å}.$$

Upon assigning equal weight to each set of measurements, one obtains 4.11 Å as the length of the cell edge. This implies a molar volume of 20.91 cc which is in good agreement with the value 20.928 cc obtained from the recent *PVT* measurements of Grilly and Mills.[6]

Seven diffraction photographs were taken of He^4 in the hexagonal phase at the same temperature but at the higher pressure of 29.70 atm. Under these conditions the hexagonal cell axes were found to be $a = 3.650 \pm 0.012$ Å and $c = 5.945 \pm 0.014$ Å. The molar volume is 20.66 cc which is slightly lower than the 20.700 cc computed from the *PVT* measurements.[6] The gross difference in molar volume of the two different phases as determined by x rays is 0.25 cc. After correcting for the difference in pressure by taking 26×10^{-4} cc/atm as the average molar compressibility of the two solids,[6] one obtains 0.21 cc/mole for ΔV_{tr}. Within the experimental error this agrees with the directly measured value[6] of 0.191 cc and the value of 0.187 cc computed from Pauling's equation.[4]

This research will be continued with a cylindrical camera in an effort to obtain reflections at greater Bragg angles.

We wish to express our appreciation to Mr. Robert Chrisman and Mr. Earl McGowen for their excellent work in the construction of the equipment.

[†] Work performed under the auspices of the U. S. Atomic Energy Commission.

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DISSOCIATIVE EXCITATION TRANSFER AND OPTICAL MASER OSCILLATION IN Ne-O_2 AND Ar-O_2 rf DISCHARGES

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This Letter is intended as a summary of excitation studies conducted in low-pressure rf discharges containing noble gases with varying amounts of oxygen impurity. We have found that the dominant mechanisms by which radiative excited states of atomic oxygen are produced under these conditions involve quasi-resonant transfer of energy from noble gas metastables to repulsive, neutral excited states of the oxygen molecule. Application of these results to Ne-O_2 and Ar-O_2 mixtures has permitted continuous optical maser oscillation on the $3\,^3P_2$-$3\,^3S_1$ fine structure transition of atomic oxygen at 8446 Å. The success of these gas maser systems suggests that dissociative excitation transfer represents a more general method for obtaining population inversions in gas discharges than is possible with methods based on inelastic atom-atom collisions.[1] In systems of that type, a close energetic resonance discrepancy (≅ a few kT) is required for efficient transfer. With systems of the present type, there are usually a moderate number of repulsive states of the impurity molecule which encompass an energy range ≅1 or 2 eV at typical internuclear separations and which terminate in the same excited states of the dissociated impurity atoms. Hence, a close coincidence with the energy of the initial metastable carrier is not required and a greater number of possible systems may be expected.

A number of relevant energy levels are shown in Fig. 1. All have been defined in respect to the ground-state oxygen molecule. Only dissociation

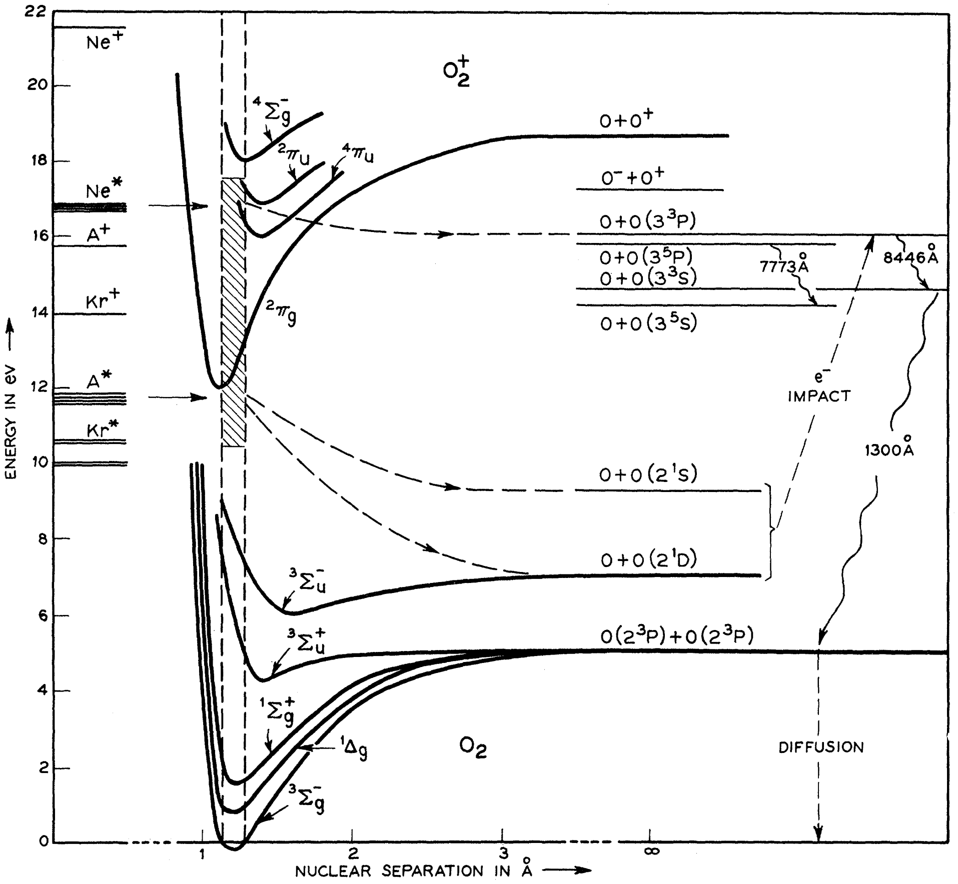

FIG. 1. Relevant energy levels. Oscillation occurs on the 8446 Å line of atomic oxygen. The paths of excitation in the Ne-O_2 and Ar-O_2 masers are shown by the arrows.

limits involving one ground-state oxygen atom are shown, with the exception of the $O^- + O^+$ limit.[2] The $3\,^3P$-$3\,^3S$ and $3\,^3S$-$2\,^3P$ transitions of oxygen are strongly allowed and the radiative lifetimes[3] of the $3\,^3P$ and $3\,^3S$ levels are obviously suitable for obtaining population inversions. As is well known, electron excitation cross sections are roughly proportional to the electric-dipole transition probabilities between a given pair of levels. Consequently, population inversions on the $3\,^3P$-$2\,^3S$ transitions cannot be maintained by electron impact with ground-state oxygen atoms, and selective dissociation into excited states of atomic oxygen is required to produce amplification on the 8446Å transition. The first two excited states ($2\,^1D, 2\,^1S$) of atomic oxygen are metastable and do have large exchange cross sections for electron excitation of the $3\,^3P$ term, however. The $3\,^5S$-$2\,^3P$ transition is forbidden to first order. Consequently, the $3\,^5P$ and $3\,^5S$ populations may not be inverted and absorption of the $3\,^5P$-$3\,^5S$ transition at 7773 Å permits a convenient method for probing the $3\,^5S$ density. The dashed horizontal curves in Fig. 1 represent repulsive curves estimated from the present work and are only intended for qualitative purposes. The dashed vertical lines indicate the region over which the Franck-Condon principle would predict excitation from the ground-state O_2 molecule to be probable. The validity of this principle in the present case is not obvious. However, previous studies[4] of inelastic atom-molecule collisions have shown that the Franck-Condon principle is at least approximately obeyed in transfer processes of the present type. The shaded area in Fig. 1 represents a range over which various (in many cases, unclassified) absorption bands have been observed from the O_2 ground state. Presumably, many of these bands terminate in repulsive potential curves. The four[5] metastable levels ($^3P_2$, $^3P_1$, $^3P_0$, and $^1P_1$, in order of increasing energy) of Ne, Ar, and Kr are denoted by asterisks.

Absorption studies in the afterglow of pulsed discharges containing varying quantities of oxygen demonstrated that the total destruction cross sections for all of the metastable states of Ne, Ar, and Kr in two-body collisions with ground-state O_2 molecules were comparable and in the general order of 2×10^{-15} cm². Comparison of the exponential decay rate [see Fig. 2(a)] of the 8446Å emission line with the decay of neon metastable densities in similar afterglow experiments demonstrated that the reaction,

$$\mathrm{Ne}(^3P_1, {}^3P_0) + \mathrm{O}_2 \rightarrow \mathrm{O}(3\,^3P) + \mathrm{O} + \mathrm{Ne}, \qquad (1)$$

was a dominant source of $\mathrm{O}(3\,^3P)$ excitation for the conditions in the Ne-O_2 maser. A similar reaction contributed to a lesser extent to the formation of the $\mathrm{O}(3\,^5P)$ term, and fluorescent enhancement in the afterglow was not encountered in the only three excited states of atomic oxygen observed with energy levels occurring above the neon metastables. From the time dependence of the maser output and absorption of the 7773Å line in pulsed rf discharges, it was found that the reactions,

$$\mathrm{Ne}(^3P_2) + \mathrm{O}_2 \rightarrow \mathrm{O}(3\,^3S, 3\,^5S) + \mathrm{O} + \mathrm{Ne}, \qquad (2)$$

represented an important source of 3S levels.

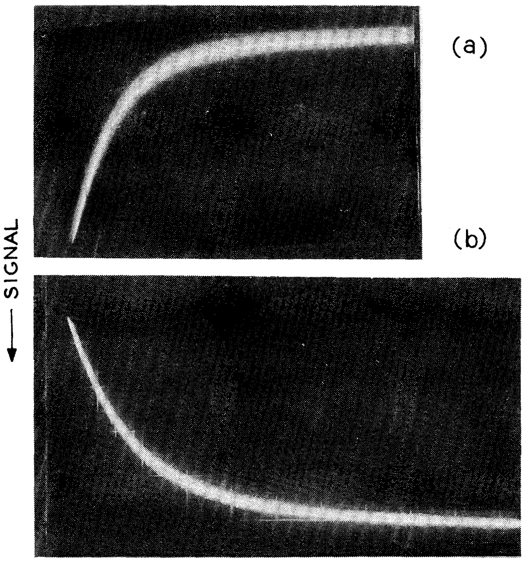

FIG. 2. (a) Fluorescent decay of the 8446Å line in Ne-O_2 mixtures superimposed on the pure exponential decay generated by an RC network ($RC = 37\,\mu$sec); (b) buildup of the 8446Å line in Ar-O_2 mixtures superimposed on the response of an RC network to a step function ($RC = 180\,\mu$sec).

The total destructive frequency $[(0.6\pm0.3)\times10^{-10}$ cm³/sec at 300°K] for $\mathrm{Ne}(^3P_2)$ - O_2 collisions is less than that $[(2.0\pm0.5)\times10^{-10}$ cm³/sec at 300°K] for $\mathrm{Ne}(^3P_1, {}^3P_0)$ - O_2 collisions. Hence, the combined effects of reactions (1) and (2) lead to a sharply rising spike in the maser output when the rf is turned on and a slower decrease to the equilibrium output. A monotonic, rapid decrease occurs when the rf is turned off.

Time-dependent studies of the 8446Å emission line in Ar-O_2 and Kr-O_2 mixtures demonstrated that the excitation arose almost entirely through the two-step process:

$$\left.\begin{matrix}\mathrm{Ar}^*\\ \mathrm{Kr}^*\end{matrix}\right\} + \mathrm{O}_2 \rightarrow \mathrm{O}^* + \mathrm{O} + \left\{\begin{matrix}\mathrm{Ar}\\ \mathrm{Kr}\end{matrix}\right. \qquad (3)$$

and

$$(\text{energy}) + e + \mathrm{O}^* \rightarrow \mathrm{O}(3\,^3P) + e, \qquad (4)$$

where O^* represents a metastable level of atomic oxygen. The identity of these reactions was established from the slow buildup time [characteristic of diffusion, Fig. 2(b)] and extremely fast decay (characteristic of electron thermalization) found in the emission line and Ar-O_2 maser output under pulsed rf excitation. Relative intensity measurements made on the 7773 and 8446Å lines showed an order-of-magnitude increase in 8446Å emission in going from Kr-O_2 to Ar-O_2 mixtures. The latter clearly implies a resonance effect in the production of oxygen metastables and may be explained either through increased production of $2\,^1D$ metastables (e.g., by dissociation into two $2\,^1D$ atoms) or by increased production of $2\,^1S$ metastables.

Oscillation in the present masers has been observed using two-meter, confocal Fabry-Perot interferometers[6] with external mirrors and windows placed at the Brewster angle. Measurements obtained with a 7-mm i.d. quartz discharge tube gave optimum pressures of 0.014 mm Hg of O_2 and 0.35 mm Hg of Ne in the Ne-O_2 case and 0.036 mm Hg of O_2 and 1.3 mm Hg of Ar in the Ar-O_2 case. Output powers of $\cong 2$ mW are obtained in each case, corresponding to an optical gain $\cong 3\,\%$ per meter. Measurements made with 10- and 7-mm i.d. discharge tubes show the gain at optimum conditions varies roughly as the reciprocal of the tube diameter. The higher pressures required for the Ar-O_2 case are consistent with the different excitation methods discussed above. Fluorescent intensity measurements made with a high resolution (1:272 000) grating spectrometer under these conditions show that the $3\,^3P$ populations are distributed according to their statistical weights and that the fluorescent widths ($\cong 3$ kMc/sec) in both masers are primarily determined by pressure broadening. With both masers, an anomalous shift has been observed in the oscillation frequency towards shorter wavelengths than the fluorescent peak of the $^3P_2$-$^3S_1$ component. This anomaly suggests the presence of an unidentified absorption line in close proximity to 8446 Å.[7]

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AZBEL'-KANER RESONANCE IN TIN WITH MAGNETIC FIELD PERPENDICULAR TO THE SURFACE*

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We are reporting the observation of Azbel'-Kaner resonance in white tin with the external magnetic field at large angles, including 90°, relative to the crystal surface. In all cases, large angle resonances were observed only when the magnetic field was directed along or within roughly 10° of the [100] axis. For the 90° case, cyclotron resonance signals were found to depend on the direction of the linearly polarized rf currents relative to crystal axes in the surface.

The theory of cyclotron resonance in metals as proposed by Azbel' and Kaner[1] predicts a resonant behavior of the surface impedance periodic in $1/H$, when the field is aligned parallel to the surface. Azbel' and Kaner predict that even a very small angle of inclination of H to the surface would seriously diminish the amplitude of such oscillations. Experiments on a large number of metals have born out these predictions. Tipping the field out of the surface to 1 or 2 degrees in most instances destroys the resonance entirely.

Contrary to expectation, we have found cyclotron resonance signals in Sn with the characteristic subharmonic structure clearly resolved, when the magnetic field is perpendicular to the surface and along the [100] axis of the crystal. This resonance series, as observed at 35 kMc/sec and with rf current directed along the [100] axis in the surface, is shown in Fig. 1. We have also plotted the $1/H$ dependence of the absorption derivative maxima indicating an $m^* = 0.57\, m_e$. Both fundamental and first subharmonic show some distortion whose cause we do not understand as yet, and the points chosen are such as to fit well with the remaining higher subharmonics. Signals observed persist for about 10° in the vi-

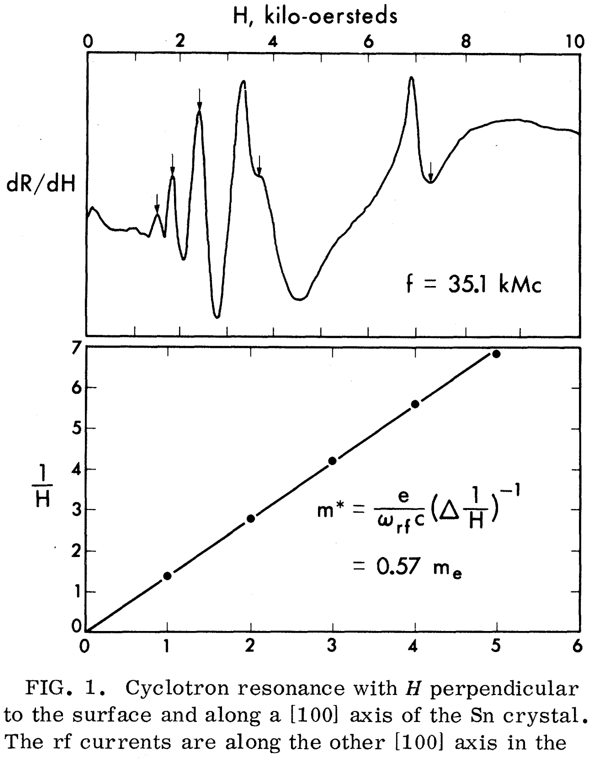
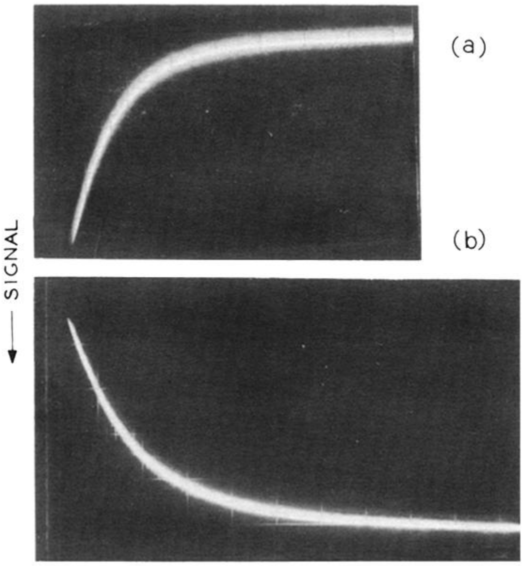


FIG. 1. Cyclotron resonance with H perpendicular to the surface and along a [100] axis of the Sn crystal. The rf currents are along the other [100] axis in the surface. Absorption derivative maxima show the characteristic $1/H$ dependence of cyclotron resonance in metals.



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