Adelphi University

A REPORT FROM THE

DEPARTMENT OF

PHYSICS

ADELPHI UNIVERSITY

GARDEN CITY, L. I., N.Y.

STUDIES ON EMISSION PROCESSES

IN

OPTICALLY PUMPED MERCURY VAPOR

By

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1. INTRODUCTION

When optically pumped mercury vapor reemits radiation, the character of the radiation from the 6^3P_1 electron transition to 6^1S_0 (i.e. the 2537A radiation) changes markedly as a function of numbers of atoms per cm^3 in the radiated and pumped mercury gas sample. Because of the importance of this effect on the measurement of lifetimes of excited states in atoms (using the Hanlé method)[1], and the use of the transition as a tool for studying atomic collision cross sections,[2,3,4] a complete investigation of the absorption and emission process was undertaken. Earlier work reported by J. P. Barrat[5] was extended to lower pressure ranges and the agreement between that work and this in regions where the data could be compared, was reasonably good. The investigations reported on here were carried on in the naturally occurring isotopic mixture of mercury, and later with the mercury 198 isotope alone.

The analysis of the data obtained does indicate that the process which occurs in the re-emission is one that can be described in terms of an apparent increase of the excited state lifetime as the pressure is increased. This fact can be determined since data taken at fixed pressures (or what is equivalent, temperatures) as a function of applied magnetic fields can be interpreted in terms of the lifetime of the excited state. At different pressures, a different lifetime

other than the "natural" lifetime occurs. The lifetime data,
however, is single valued at a given pressure (or temperature).
Typically, these experimentally determined lifetimes may vary
from 1.10 (10^{-7}) seconds at pressures of 10^{11} atoms/cm^3 to
several times that value at 10^{12} atoms/cm^3. This behavior
is noted both in the natural mercury isotopic mixture and
in the Hg^{198} sample. The earlier results with natural
mercury have been reported in (6). This report will deal
more extensively with the Hg^{198} data and the results obtained.

2. EXPERIMENTAL TECHNIQUE

The geometry of the experiment is indicated in
Figure I. Light from a low pressure mercury arc (A) passes
through wollaston prism (C). The component of polarized
radiation perpendicular to the plane of the diagram then
falls on the resonance cell (D), an evacuated quartz cell
in which a drop of mercury is contained in a tip which is
in turn immersed in a dewar. The temperature in the dewar
controls the vapor pressure of the mercury and hence the
number of atoms per cm^3 of mercury in the path of the
radiation. The 2537A line of the incident beam is strongly
absorbed by the mercury vapor in the cell, and re-emissions
at right angles to the incident beam are studied by using
the polarizer-analyzer prisms (E & F) and the photomultiplier
(H). The resonance cell is surrounded by two pairs of

Helmholtz coils, one pair annuling the earth field and the other used to apply a field parallel to the direction of observation. Preliminary pump-down and outgassing assured elimination of impurity atoms in the system.

Measurements were made of the polarization P of the emitted radiation, where P is here defined as

$$P = \frac{Iy - Ix}{Iy + Ix} \tag{1}$$

and Iy and Ix are the intensities of emitted radiation perpendicular to and parallel to, respectively, the plane of the diagram of Figure I. The polarization was measured first as a function of the dewar temperature and then as a function of a magnetic field applied parallel to the viewing direction while holding the dewar temperature constant. These measurements were taken at various temperatures ranging from $-40^{\circ}C$ to $+20^{\circ}C$ and in applied magnetic fields from 0.0 to 1.0 gauss.

In the method of analysis used (the Cornu method) the polarization observed is given by

$$P = \cos 2\theta \tag{2}$$

where θ is the angle between the axes of the two Wollaston prisms used as polarizer and analyzer. The appropriate intensity match was performed by use of a photomultiplier. In some of the data to be presented later, plots are made with the angle θ as one of the variables.

As the temperatures were decreased, the number of atoms/cm^3 in the resonance bulb decreased. This, of course, lowered the intensity of the re-emitted signal which is analyzed. The signal size set a limit, using present equipment (a IP21 photomultiplier tube) at -40°C for reasonable intensity readings.

3. RESULTS

a) Naturally Occurring Mercury

Data for the angle Θ (and Polarization) vs. Temperature is presented in Figure 2. The drastic decrease in polarization as a function of dewar temperature is evident. In the range between 1(10^{12})atoms/cm^3 (approximately -20°C) to 1.0(10^{13})atoms/cm^3 (approximately 4°C) the observed polarization decreases from approximately 80% to approximately 50% as shown in the Figure   The steep increase in Θ once a temperature corresponding to -17°C is reached is also evident.

Measurements were taken to determine the lifetimes of the excited state. In the Hanle method for finding the lifetime, a magnetic field is applied along the direction of the line of observation. This magnetic field depolarizes the emitted radiation in a theoretically predicted fashion. The polarization in a given magnetic field is:

$$P = \frac{\sum_r \sum_s N_r \left[\dfrac{A_{rs} - B_{rs}}{1 + \left(\dfrac{eH\tau g_{rs}}{mc}\right)^2}\right]}{\sum_r \sum_s N_r \left[A_{rs} + B_{rs}\right]} = \sum_r \sum_s \frac{(A_{rs} - B_{rs})}{(A_{rs} + B_{rs})} \left(\frac{1}{1 + eH\tau g_{rs}}\right)^2 = \frac{P_0}{\sum_{rs} 1 + \left(\dfrac{eH\tau g_{rs}}{mc}\right)^2} \quad (3)$$

where the subscript r indicated the rth isotope, and s denotes
the sth spectral line. N_r is the fraction of isotope r,
A and B the components of radiation in the y and x direction,
respectively, e the electronic charge, H the magnetic field,
m the electron mass, c the speed of light, and g the Landé
factor for the appropriate isotopic transition.

The typical result of applying a magnetic field to
the natural mercury vapor is shown in Figure 3. The lifetime
of the excited state is determined by comparing theoretically
predicted polarizations (which are a function of the lifetime),
and observed polarizations. The best fit is taken and the
appropriate lifetime is obtained.

b) Mercury 198 Results

Essentially the same data has been taken using the
mercury 198 isotope as sample in the resonance bulb. As is
indicated in Figure 4, the angle Θ and polarization vs.
temperature behavior is essentially of the same form as that
of naturally occurring mercury with the difference that the
low temperature polarization value is approximately 100%.
This is to be expected since the 198 isotope has no nuclear
spin and so there is no splitting of the electron energy
levels due to nuclear spin effects. This means, of course,
that in Equation 3 there is no isotope to be considered other
than Hg^{198} with the resulting simplification of analysis.

In Figure 5 typical polarization vs. resonance cell

temperature is plotted. The zero magnetic field and 0.3 gauss data are shown. The lifetimes determined from data similar to that shown in Figure 5 is presented in Figure 6.

These lifetimes are determined in the following way. The values of polarization as a function of H at various temperatures are plotted. A theoretical plot of polarization as a function of Z where $Z = H\Upsilon$, (Υ being the lifetime of the excited state) is made. Using this definition of Z, equation 1 reduces to (2)

$$P = \frac{Po}{1 + \left(\frac{eq}{mc} Z \right)^2}$$

Typical polarization vs. Z data is shown in Figure 7. Knowing the various values of H, from a least squares fit of observed and theoretical polarization vs. Z data it is possible to determine Υ. This is the procedure used to calculate the lifetime values presented in Figure 6. This plot is an average of the various data.

4. CONCLUSIONS

It appears that there is good agreement between this extended work and the work reported by Barrat. The varied magnetic field values used here, and the temperature ranges involved allowed extensive analysis of data.

The behavior of emitted radiation indicates that a single value of lifetime of the excited state can be assigned

at various pressures of mercury vapor. The value so assigned increases with pressure as shown in Figure 6. The spread of values does appear to increase as more elevated pressures are studied, but this can be explained as principally due to the effects of walls and wall collisions on the character of the emitted radiation. The fact that a general flattening of results below temperatures of -18°C occurs, indicates that there the "natural" lifetime is the appropriate one. Photons emitted by excited atoms at pressures lower than 10^{12} atoms/cm^3 are not reabsorbed by other atoms before they escape from the resonance bulb. At pressures higher than this photons are absorbed, but the subsequently remitted radiation is apparently coherently absorbed and can be described by the increased lifetime.
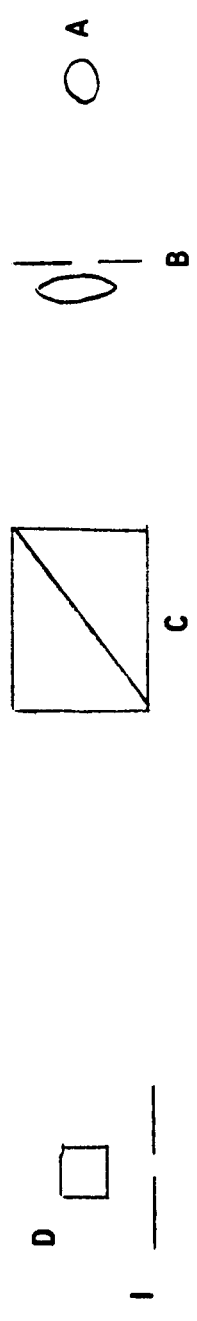
A Mercury Arc

B Lens - slit System

C Wollaston Prism

D Resonance Bulb

E Wollaston Prism

F Wollaston Prism

G Lens

H Intensity Measurer

I Rectangular Slit

FIGURE 1

EXPERIMENTAL SET - UP
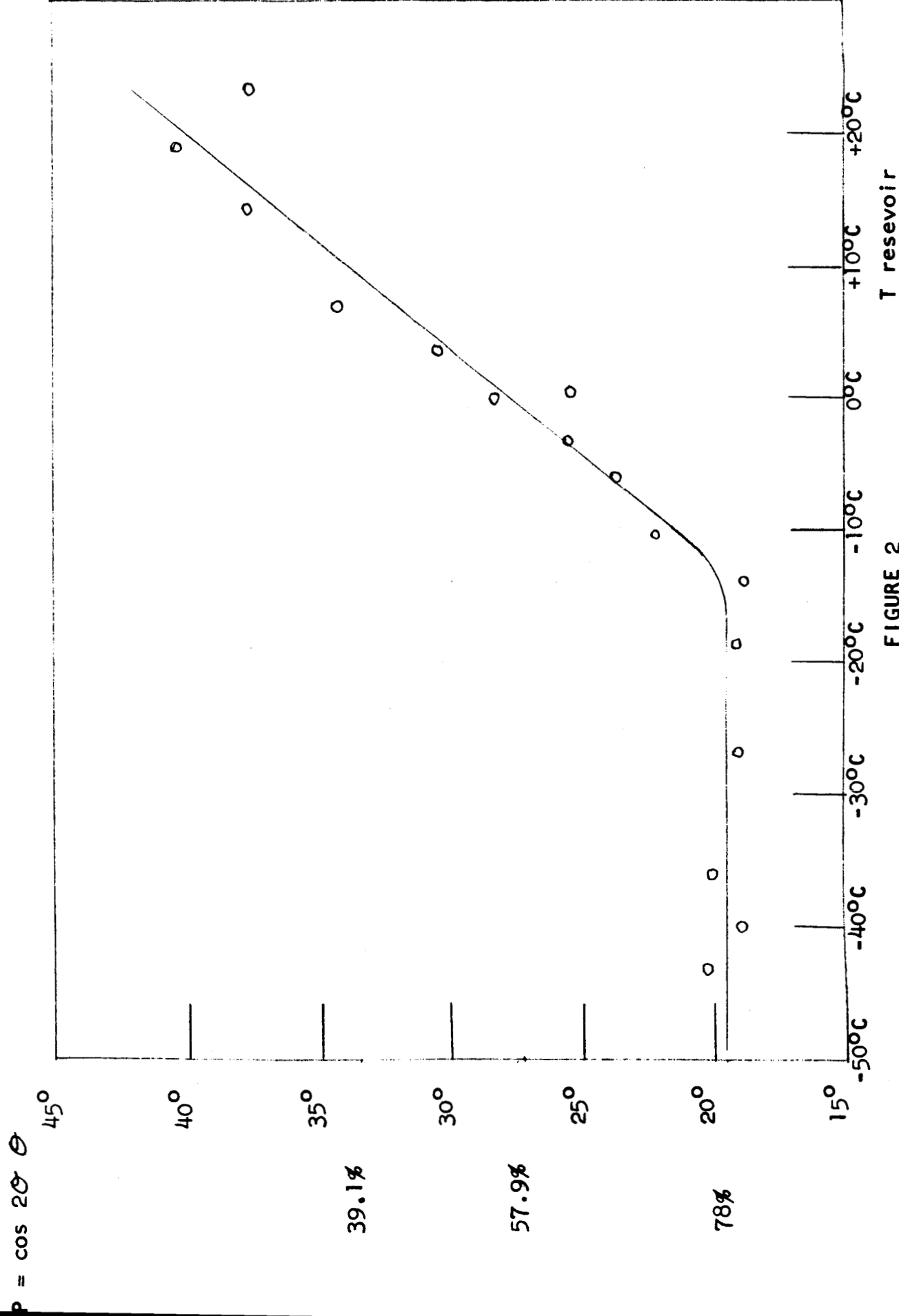
FIGURE 2

θ vs T(H=O) For Naturally Occurring Hg
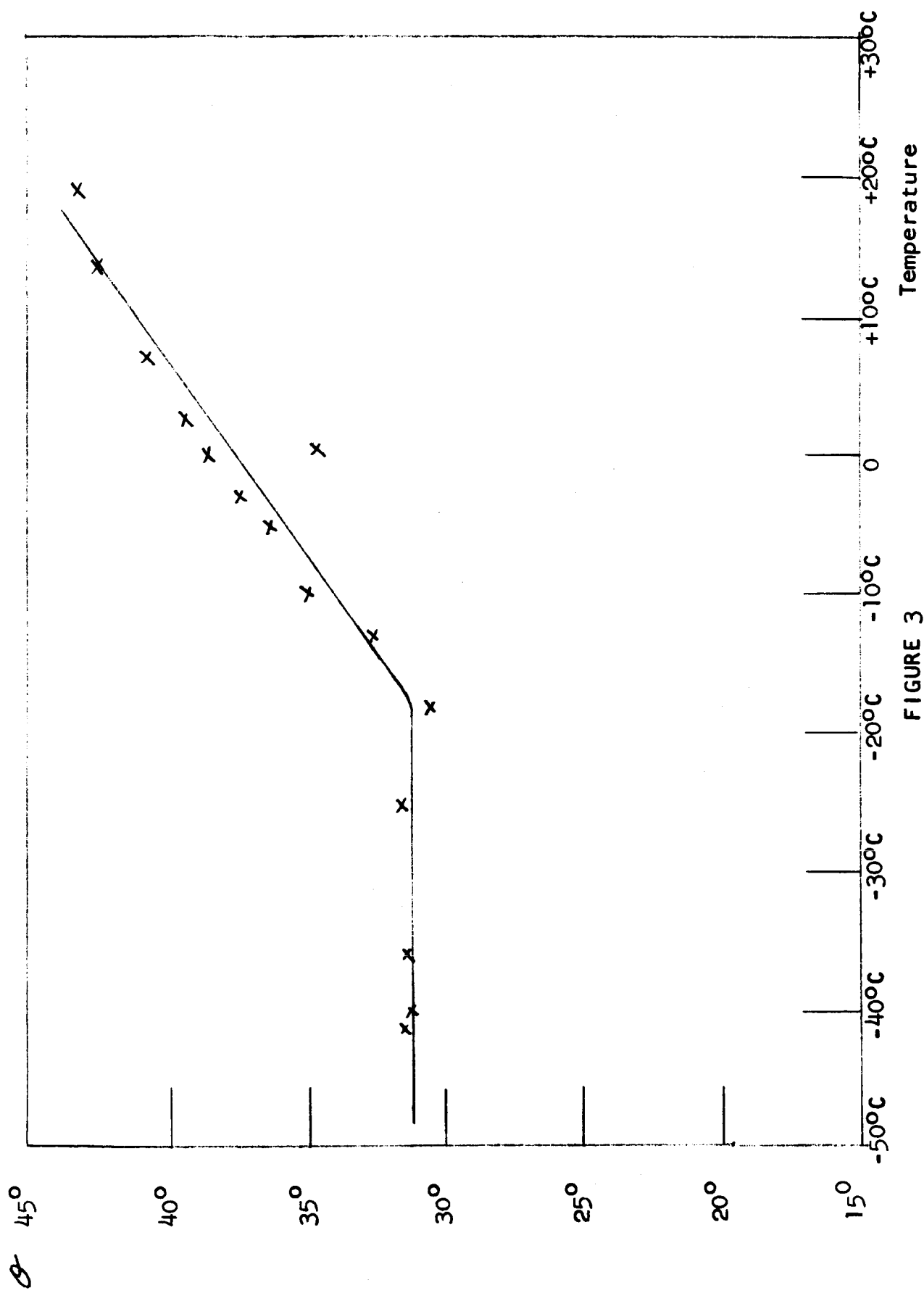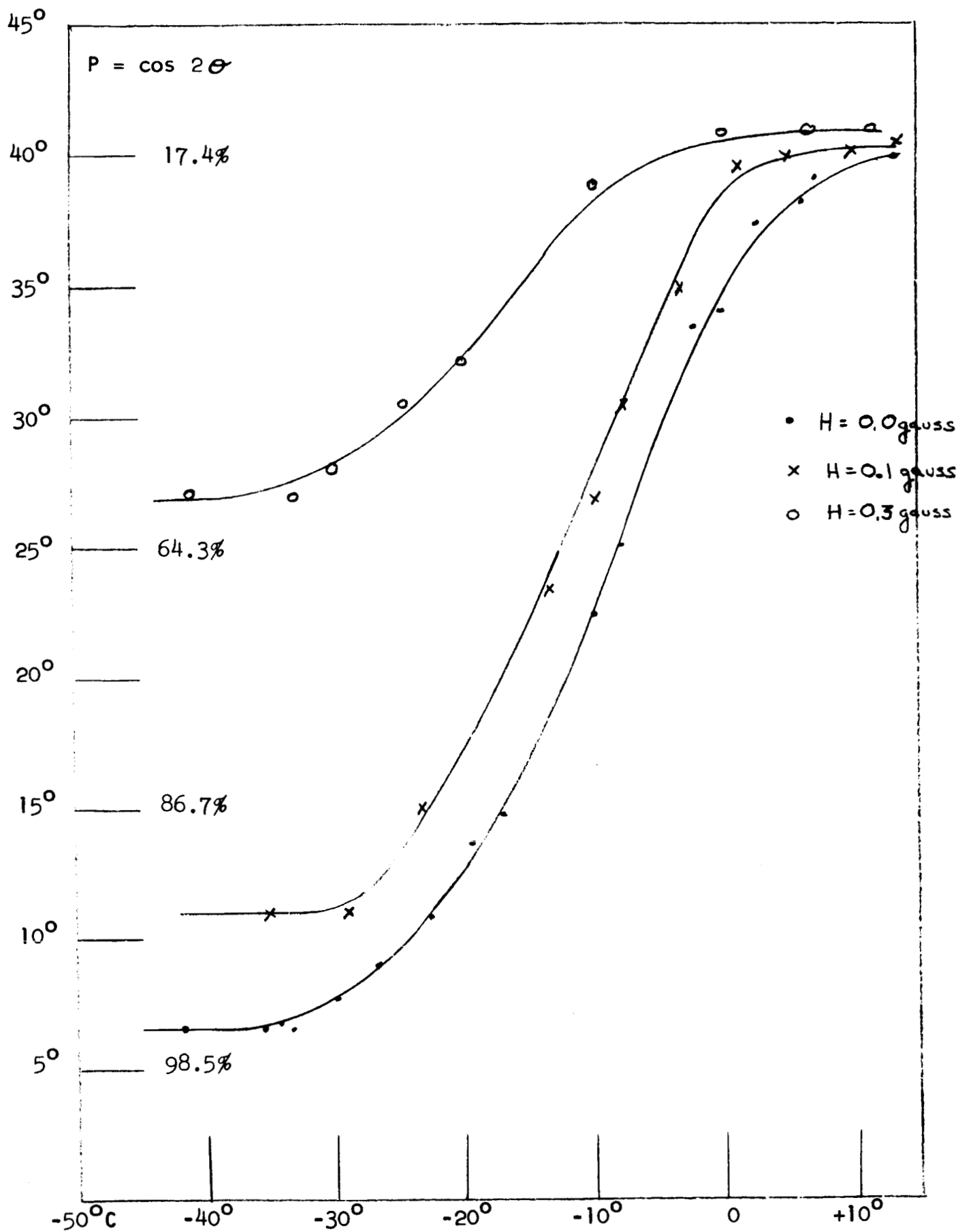
$P = \cos 2\theta$

FIGURE 3

$\mathcal{B}$ vs Temperature (T) for H = 0.27 gauss

FIG. 4 Angle (Θ) vs. Temperature (T) for Hg^{198} °C
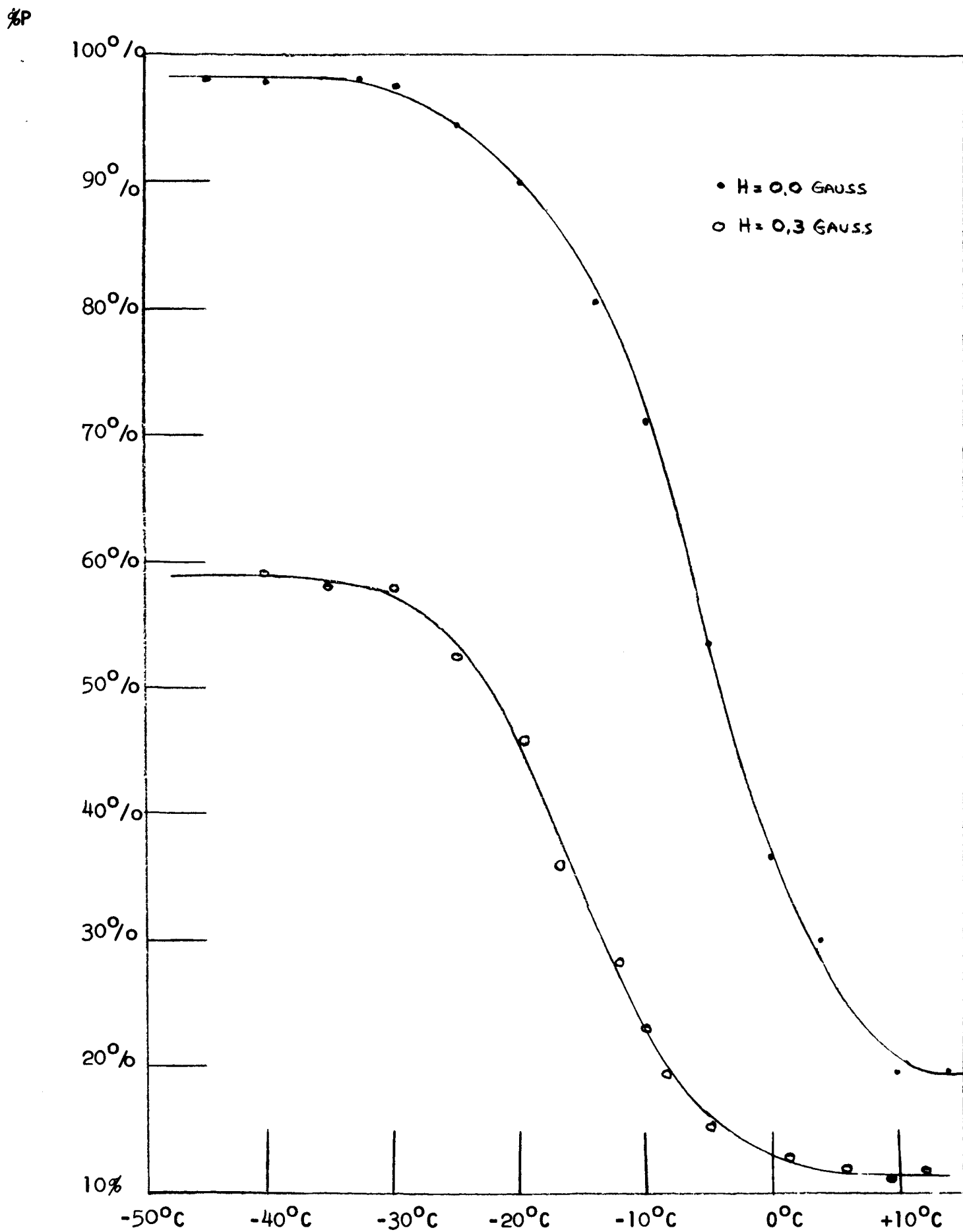
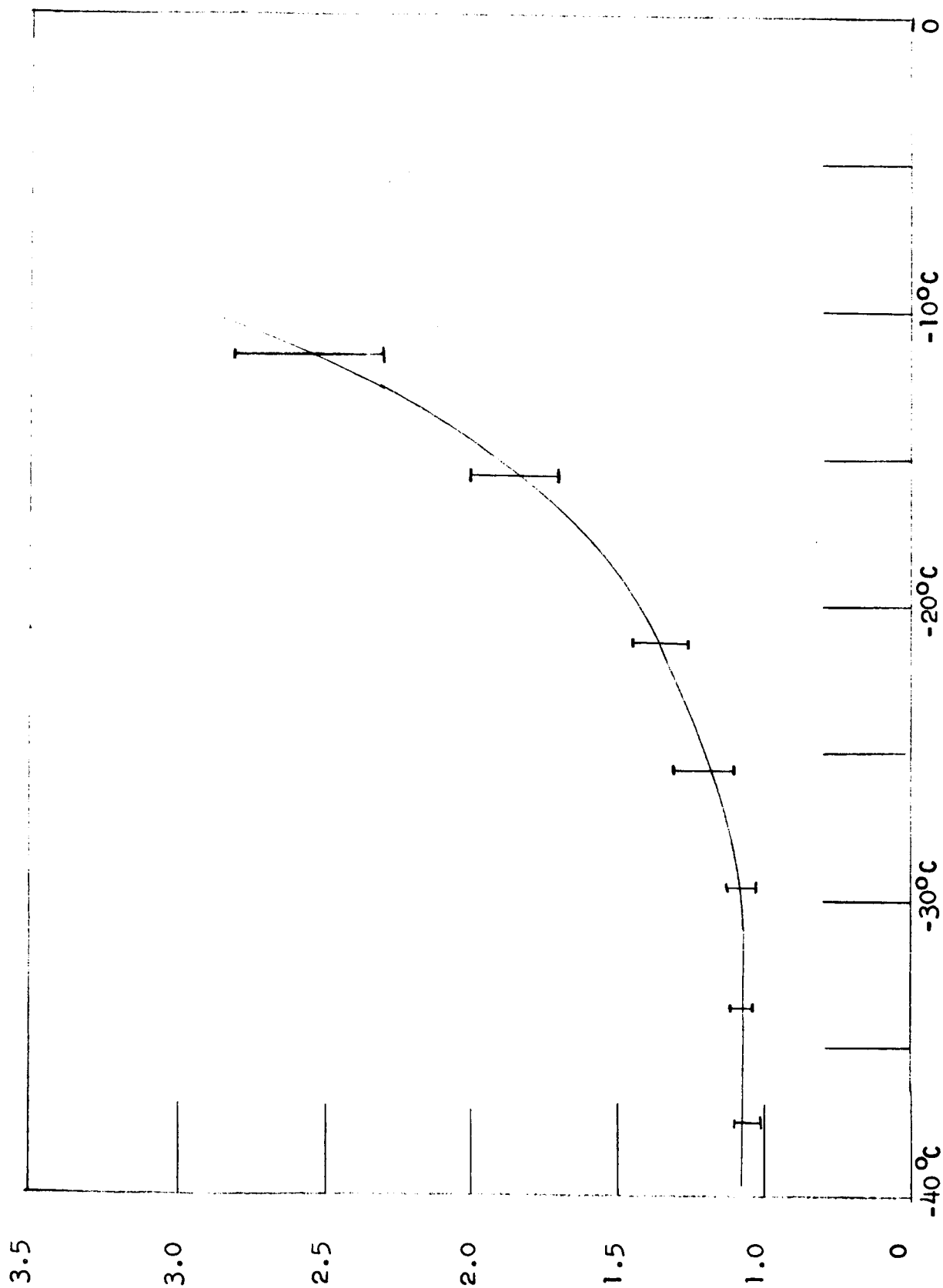FIG. 5 Polarization (P) vs. Temperature (T) for Hg^{198}

Lifetime τ (in units of 10^{-7} seconds) vs Cell Temperature T (average) for Hg^{198}
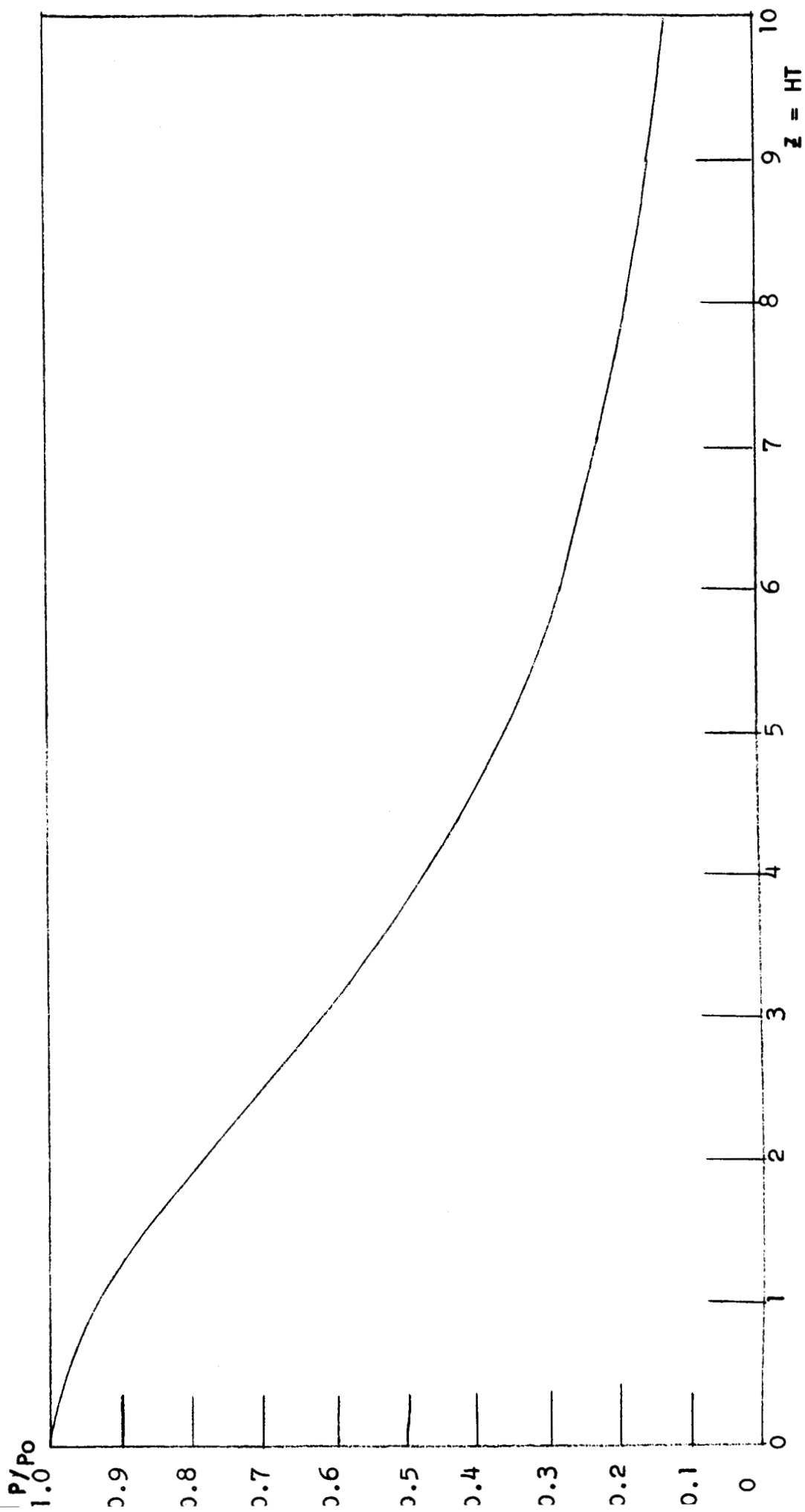
FIGURE 6

FIGURE 7

P/P_O vs z

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