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IMPRISONMENT OF RESONANCE RADIATION IN ATOMIC ABSORPTION SPECTROSCOPY *

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ABSTRACT

Imprisonment of resonance radiation by atomic absorption of Mn and Zn at 4030.76 Å and 2138.56 Å respectively, for a wide range of concentrations between 1 and $5x10^3$ ppm in an air-acetylene flame has been investigated. The optical depths τ_0, the mean probability values of photon escape θ,, and photon capture λ are determined. It is shown that there is certain state of equilibrium between photon escape and capture, where $\tau_0 = 1$, and $\theta = \lambda = 0.5$ which were relevant to 50% of total absorption. The dependence of both τ_0 and λ on the total number of atoms in ground state N cm^{-3} has been found significant. It is found that imprisonment of resonance radiation is negligible at low absorber opacity ($\tau_0 < 1$), while is remarkable at high absorber opacities ($\tau_0 > 1$), and is independent on N at $\tau_0 \gg 1$. The significance of imprisonment concept is carefully assessed.

INTRODUCTION

Imprisonment of resonance radiation in radiative plasmas was first developed by Biberman[1] and, independently, by Holstein[2,3] considering repeated reabsorption in the case of optical excitation. Some authors[4-6] were interested in studying the experimental verification of the imprisonment radiation phenomena and connected it with changes in the atomic life times, transition probabilities and excitation cross-sections. Definitely, the problem might be treated by solving the Holstein's integro-differential equations using simple analytical approximations which lead to certain expression denoted by the escape factor.[7,8] The escape factor represented the mean probability of photon escape. It could be expressed in terms of either photon escape[9-12] or photon capture.[4,13,14] Since the photon escape and photon capture are complementary processes, the escape factor has a dual character. Therefore, the same expression for the escape factor was obtained considering both concepts.[15]

Emphasis in this paper is on the evaluation and interpretation of imprisonment of resonance radiation from an atomic absorption point of view. The mean optical depths τ_0 for wide range of concentrations from 1 to $5x10^3$ ppm of Mn and Zn at the resonance lines 4030.76 Å and 2138.56 Å respectively, were determined. The escape factor θ, and accordingly, the mean probability of photon capture λ were calculated. The dependence of both τ_0 and λ on the total number of atoms in ground state N was studied extensively.

THEORY

A. Imprisonment of atomic resonance radiation:

On the basis of photon escape concept, the escape factor

of a spectral line obeying a Gaussian profile in a uniformly excited source having a plane-parallel slab geometry, may be given by[16]

$$\theta = \frac{2 - \exp(-10^{-3}\tau_0)}{1 + \tau_0} \quad , \tag{1}$$

where $\tau_0 = k_0 L$ is the optical depth, k_0 is the absorption coefficient and L is the length of source.

But on the basis of photon capture concept, and assuming that the source is isolated and only line photons within the source contribute to photo-excitation, the mean probability of photon capture (or imprisonment) may be expressed using the model predicted by Zanstra,[9] Anderson,[17] Osterbrock[10] and Hearn[11] (ZAOH) as[15]

$$\psi = 2 \int_0^{\nu_1} \phi(\nu) d\nu \quad , \tag{2}$$

where ν_1 is the frequency at the line center, $\phi(\nu)$ is the line profile (absorption or emission) which obeys the normalization condition $\int_{-\infty}^{+\infty} \phi(\nu) d\nu = 1$. The mean Biberman-Holstein coefficient Λ(a notation given by Drawin and Emard[18,19]), or the escape factor, may be given by

$$\begin{aligned} \Lambda &= 1 - \psi \\ &= 1 - 2 \int_0^{\nu_1} \phi(\nu) d\nu \\ &= 2 \int_{\nu_1}^{\infty} \phi(\nu) d\nu \\ &= T^{as}(\tau_0) \quad , \end{aligned} \tag{3}$$

where $T^{as}(\tau_0)$ is the asymptotic form of Holstein's transmission factor. The escape factor Λ may be identified in a more straightforward way with the escape factor $\theta = T^{as}(\tau_0)$, thus

$$\theta = 1 - \psi \quad . \tag{4}$$

From Eq.(4), ψ can be calculated once θ was calculated.

The dependence of ψ on the total number of atoms in ground state N results from the relation between τ_0 and N according to the following expression[20]

$$\tau_0 = \frac{2(\pi \ln 2)^{\frac{1}{2}} e^2}{mc \, \delta\nu_D} f N L \quad , \tag{5}$$

where $\delta\nu_D = 7.16 \times 10^{-7} \nu_0 (T/M)^{\frac{1}{2}}$ is the Doppler width, e is the electronic charge, m is the mass of electron, c is the velocity of light, f is the oscillator strength of the transition dealt with, M is the atomic mass and T is the absolute temperature of the source.

B. Calculation of the total number of atoms in ground state N:[21]

The total number of atoms in ground state N can be calculated in terms of the total absorption A_T which is defined by .

$$\begin{aligned} A_T &= \frac{I_{o\nu} - I_\nu}{I_{o\nu}} \\ &= 2\pi \int_0^{\infty} \left\{ 1 - \exp\left[- \int_0^L k(\nu) ds \right] \right\} d\nu \quad , \\ &= 2\pi \frac{\delta\nu_D}{(\ln 2)^{\frac{1}{2}}} \int_0^{\infty} \left\{ 1 - \exp\left[- \int_0^L k(y) ds \right] \right\} dy \quad , \end{aligned}$$

where $y = 2(\nu - \nu_0)(\ln 2)^{\frac{1}{2}} / \delta\nu_D$, and ν_0 is the frequency at the line center. For a homogeneous source (,e.g., a flame) we may write

$$\int_0^L k(y) ds = k(y) L .$$

Therefore,

$$\frac{A_T}{2b} = \int_0^{\infty} \left\{ 1 - \exp\left[- k(y) L \right] \right\} dy . \tag{6}$$

From Eq.(6), the double logarithmic plot of A_T as a function of NfL represents a theoretic curve of growth. Using the computed

tables by Van Trigt et al,[21] for values of $A_T/2b$ at certain value of the damping constant a, relevant values of NfL/b could be estimated, where $b = \pi\delta\nu_D/(\ln 2)^{\frac{1}{2}}$.

EXPERIMENTAL

The measurements were carried out using an atomic absorption spectrophotometer (Carl Zeiss). It is fitted with a long rectangular (slot) burner which produces a wedge-shaped flame with 5 cm length, and fairly flat sides which can be viewed edge-on. The measurements were done on absorbance scale which can be increased by 1, 2, ...etc, to fit the samples of very high concentrations. Several aqueous salt solutions of a wide range of concentrations ranging between 1 and 5×10^3 ppm were prepared for Mn and Zn. The absorbances were measured for each concentration at the resonance lines 4030.76 Å and 2138.56 Å for Mn and Zn respectively, by spraying the salt solution into the flame. The air-acetylene flow rates which gave the highest value of $\log(I_o/I)$ for test samples were 75-60 divisions respectively. The necessary background corrections due to flame constituents and water molecules were carefully done. The hollow cathode lamps (H.C.L.) used were of the Westinghouse type, and the working current was 15 m.A . The flame temperature was measured using the line-reversal method and was found to be $T = 2400 \pm 20^{o}K$.

RESULTS AND DISCUSSIONS

Before applying Eq.(1), the following approximations have to be taken into consideration:

(a) The resonance lines emitted from the H.C.L. have Gaussian profiles, and the absorbed resonance line profiles, at maximum intensity, will be considerd still obey Gaussian distribution.,
(b) The absorbed resonance lines into the flame are mainly broadened by adiabatic collisions (besides Doppler), and hence, they will have Voigt profiles. For this reason, the corrections made by Blickensderfer et al[6] were used to obtain values of optical depths $\tau_o = k_oL$ of the Gaussian profiles from the measured optical depths as: for Mn, the damping constant[23] $a = 0.5$; $k_{max.}L = 0.615k_oL$, and for Zn, the damping constant $a = 0.4$; $k_{max.}L = 0.68\ k_oL$(obtained by interpolation)., and
(c) The rectangular flame used will be considered as a source of uniform excitation and has an infinite plane-parallel slab geometry.

Considering photon escape concept, values of the escape factor θ using Eq.(1) were calculated at the different values of τ_o for both Mn and Zn. According to the model predicted by ZAOH, the mean probability values of photon capture (or imprisonment) γ were calculated by Eq.(4) through values of θ. Values of τ_o, and γ are reported in Table I. Fig.1,(a) and (b) shows a plot of both θ (dashed lines) and γ (solid lines) against τ_o for Mn and Zn respectively. It could be noticed that both θ and γ obeyed the familiar exponential behaviour represented by Eq.(1), which is in agreement with similar plots of other relations predicted by several authors.[7,8,15] But in atomic absorption spectroscopy, it might be significant to study the behaviour of γ with τ_o which looks, apparently, to Beer's law. At higher values of τ_o (,i.e., > 3), γ tended towards constancy which means that

absorption approached saturation. It is interesting to find that at the intersection point of the two plots, $\theta = \psi = 0.5$ which is relevant to $\tau_0 = 1$. At this point, both probabilities of photon escape and capture are equal. From a graphical representation of the percentage total absorption A_T and τ_0 for both Mn and Zn, we found out that $A_T = 50\%$ at $\tau_0 = 1$. Therefore, the intersection point, where $\tau_0 = 1$, may represent the equilibrium or midpoint between photon escape and capture (or between transmission and absorption) of the resonance radiation.

The total number of atoms in ground state N were estimated using the determined values of A_T for each concentration of Mn and Zn. The calculated values of $\delta\nu_D$ were $3.524 \times 10^9 s^{-1}$ and $6.086 \times 10^9 s^{-1}$, and hence, values of b were $13.30 \times 10^9 s^{-1}$ and $22.97 \times 10^9 s^{-1}$ for Mn and Zn respectively. By means of the computed Tables of Van Trigt et al,[21] values of NfL/b were obtained for values of $A_T/2b$, at the relevant values of a. Substituting by $f = 0.057$ for 4030.76 Å Mn, and $f = 1.3$ for 2138.56 Å Zn, and by $L = 5.0$ cm, values of N cm^{-3} could be estimated as given in Table I. Figs. (2) and (3) show a plot of τ_0 against N cm^{-3} for Mn and Zn respectively. It is clear that τ_0 is directly proportional to N and follow a straight line up to the value of $\tau_0 = 1$, over which it deviates from the straight line behaviour till it tends to be independent on N at larger values of $\tau_0 > 6$ for Mn and $\tau_0 > 1.5$ for Zn. Returning again to the equilibrium point (where $\theta = \psi = 0.5$, and $\tau_0 = 1$), it could be pointed out that when $\tau_0 > 1$, imprisonment of resonance radiation become remarkable and effective, to which, reabsorption might be attributed. At larger values of $\tau_0 \gg 1$,

or at high absorber opacities, it could be concluded that imprisonment of resonance radiation becomes greater, and hence, causes greater reabsorption which may lead to saturated absorption where τ_0 is independent on N. While, at low absorber opacities, i.e., $0 < \tau_0 < 1$, imprisonment of resonance radiation might be negligible.

Figs. (4) and (5) are plots of ψ against N cm^{-3} for Mn and Zn respectively. It was found that the intersection point of the two asymptotes (at lower and higher values of $N^{\frac{1}{2}}$) falled at $\psi = 0.5$ and at $N = 1.6 \times 10^{12}$ cm^{-3} and at $N = 0.62 \times 10^{11}$ cm^{-3} for Mn and Zn respectively. These values were approximately equal to those for N at $\tau_0 = 1$ in Figs. (2) and (3). This result supports the significance of the equilibrium point referred to before. However, the general behaviour of ψ clarified in Figs. (1), (4), and (5), foretells that the expression given by Eq.(4) based on ZAOH model might be favorable in representing the idea of imprisonment of resonance radiation in atomic absorption spectroscopy.

CONCLUSIONS

(a) There is certain state of equilibrium between photon escape and capture, or between transmission and absorption, where $\tau_0 = 1$, and $\theta = \psi = 0.5$ and are relevant to 50% of the total absorption.

(b) Imprisonment of resonance radiation is negligible at low absorber opacities, i.e., $\tau_0 < 1$, while it is remarkable and effective at high absorber opacities, i.e, $\tau_0 > 1$, and is independent on N at $\tau_0 \gg 1$.

(c) The expression given for ψ by Eq.(4) and based on ZACH model may be favorable to represent the significance of imprisonment aspect in atomic absorption spectroscopy.

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Table I

Conc.n ppm	Mn(4030.76Å)			Zn(2138.56Å)		
	τ_0	ν	$N \times 10^{12}$ cm^{-3}	τ_0	ν	$N \times 10^{11}$ cm^{-3}
1				0.4397	0.305	0.276
2				0.7653	0.433	0.485
5	0.0937	0.086	0.134	1.7269	0.633	1.012
10	0.1834	0.155	0.247	2.8444	0.739	1.452
20	0.3294	0.248	0.456	3.8941	0.795	1.680
30				4.2327	0.808	1.730
50	0.8237	0.451	1.236	4.7068	0.824	1.776
80				5.0793	0.835	1.813
100	1.6024	0.615	2.660	5.2485	0.839	1.830
200	3.0701	0.754	4.912	5.7565	0.851	1.860
300	4.4180	0.815	6.180	6.0274	0.857	1.874
500	6.7393	0.870	6.926	6.5015	0.866	1.890
700	7.7502	0.885	7.038	6.8400	0.872	1.903
1×10^3	8.4615	0.893	7.083	7.1787	0.879	1.911
2×10^3	8.8733	0.898	7.101	7.5512	0.882	1.920
3×10^3	8.9857	0.899	7.105	7.9237	0.887	1.923
5×10^3	9.3976	0.903	7.118	8.1268	0.890	1.925

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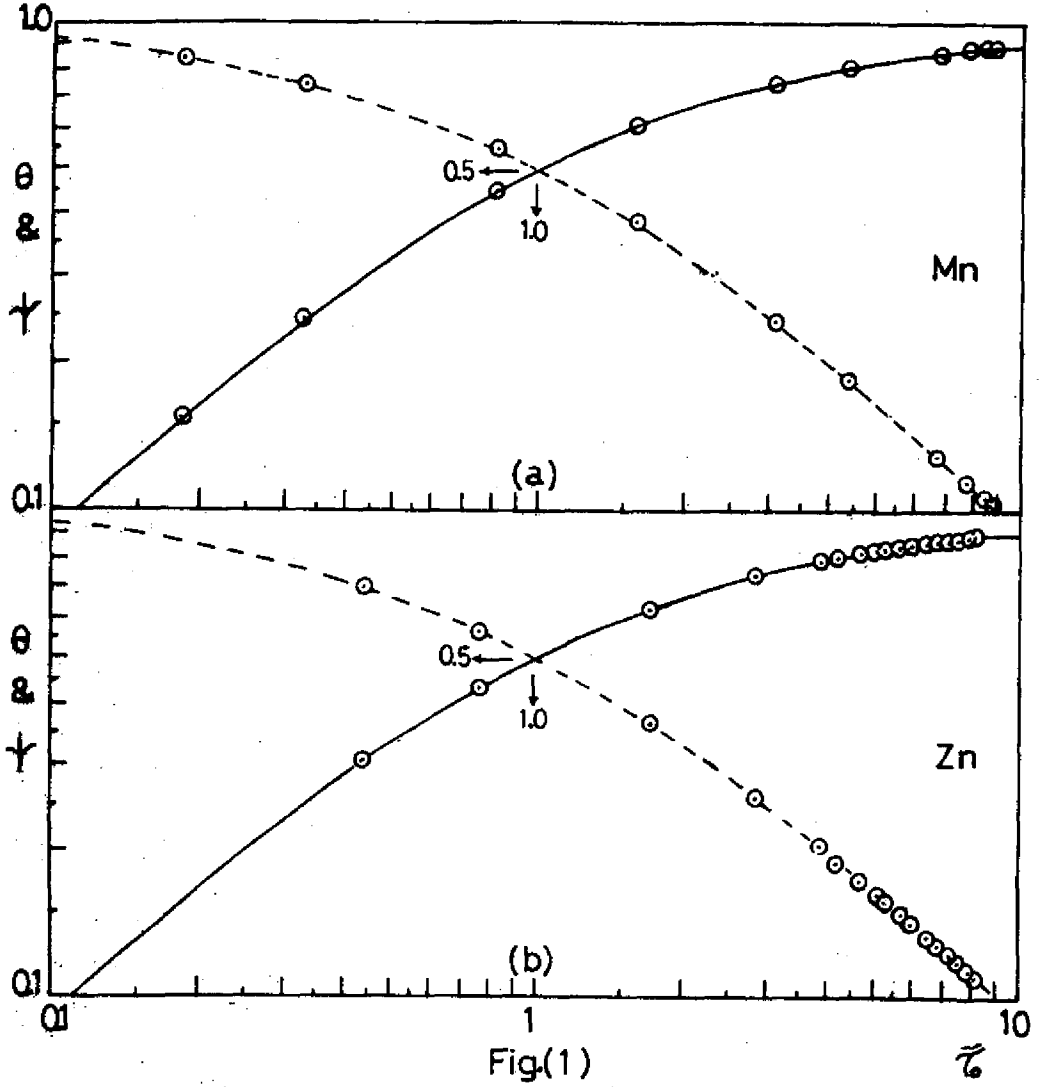
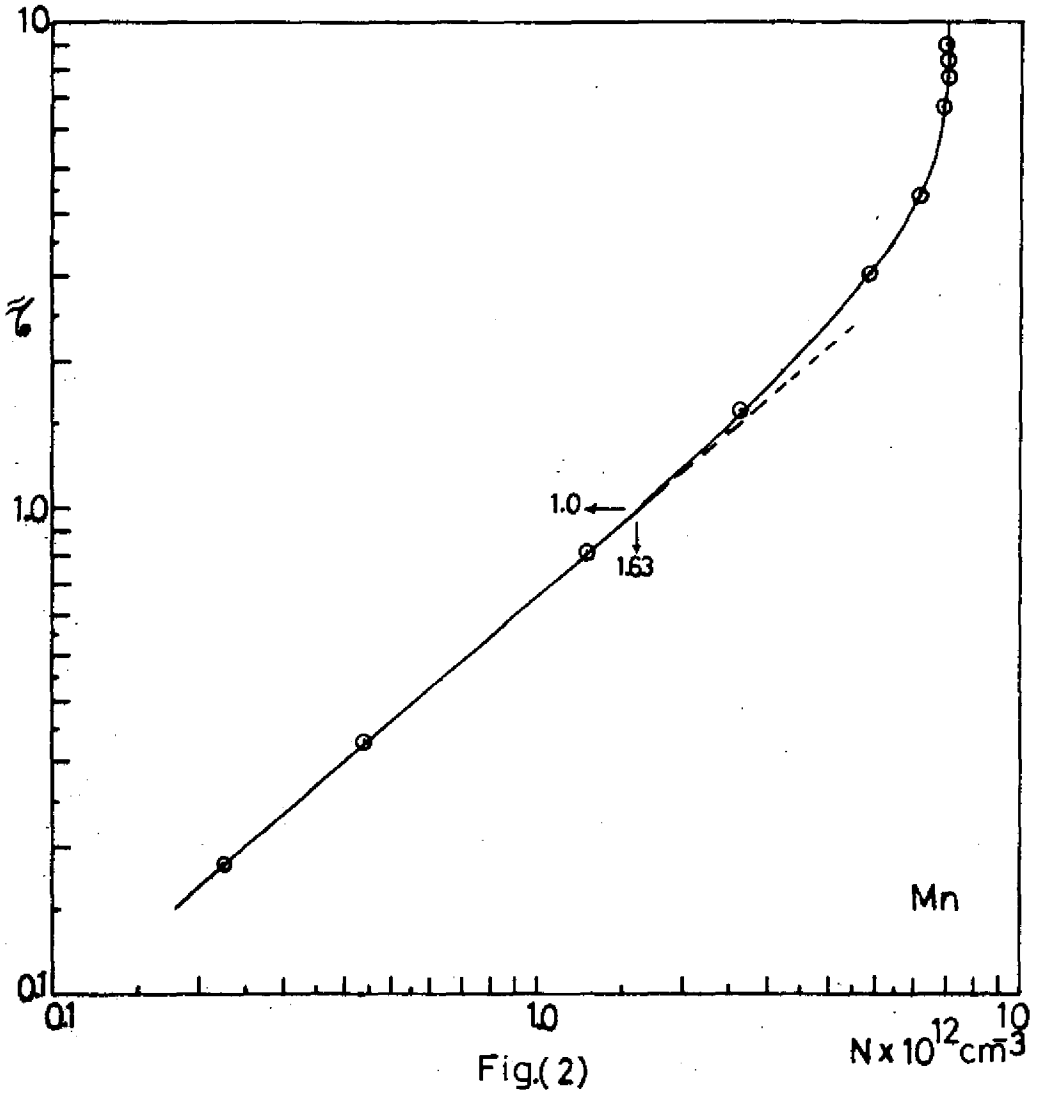

1.0
0.1
θ
&
ϒ
0.5
1.0
Mn
(a)
θ
&
ϒ
0.5
1.0
Zn
(b)
0.1
1
10
τ

Fig.(1)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1.63
Mn
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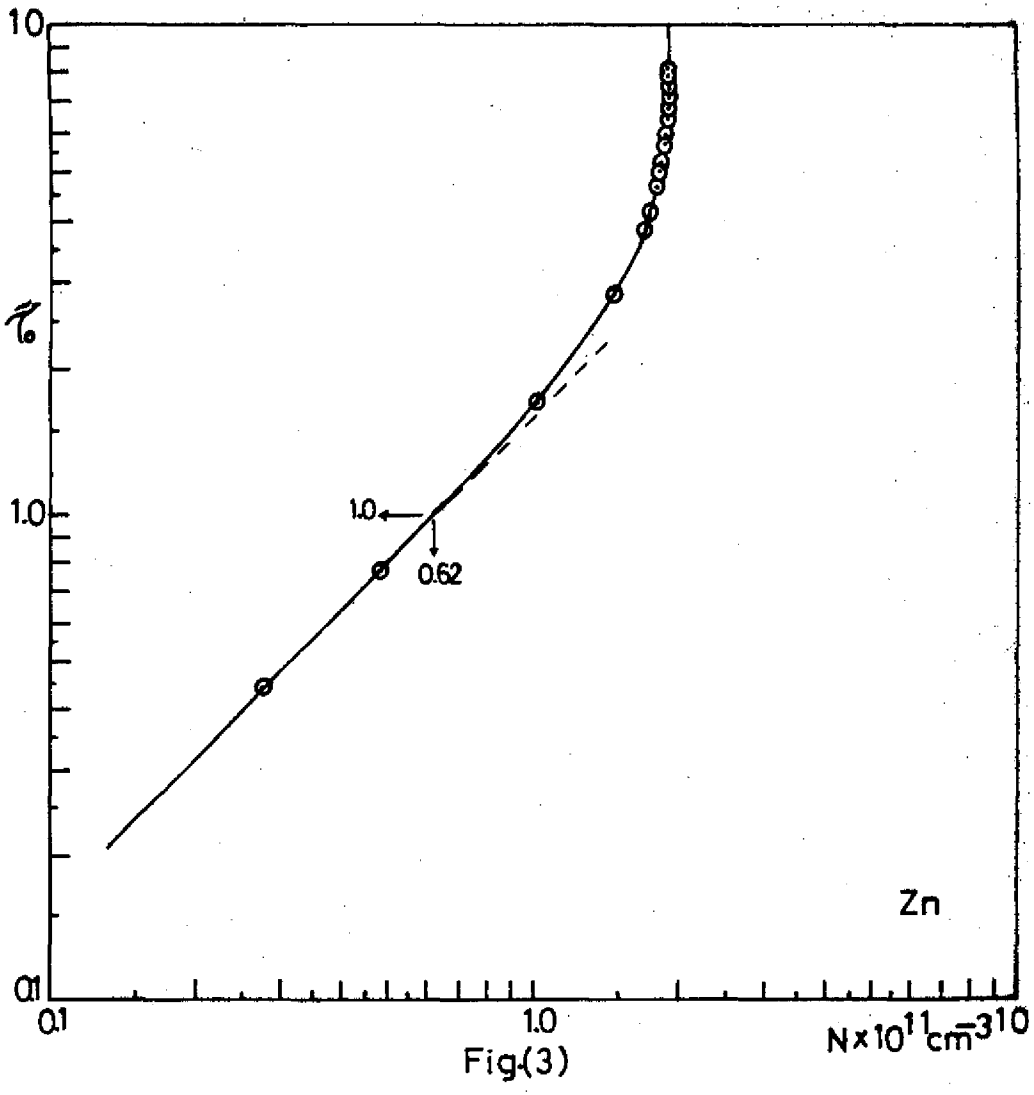
1.0
10
N x 10^12 cm^-3

Fig.(2)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τ_0
1.0
0.62
Zn
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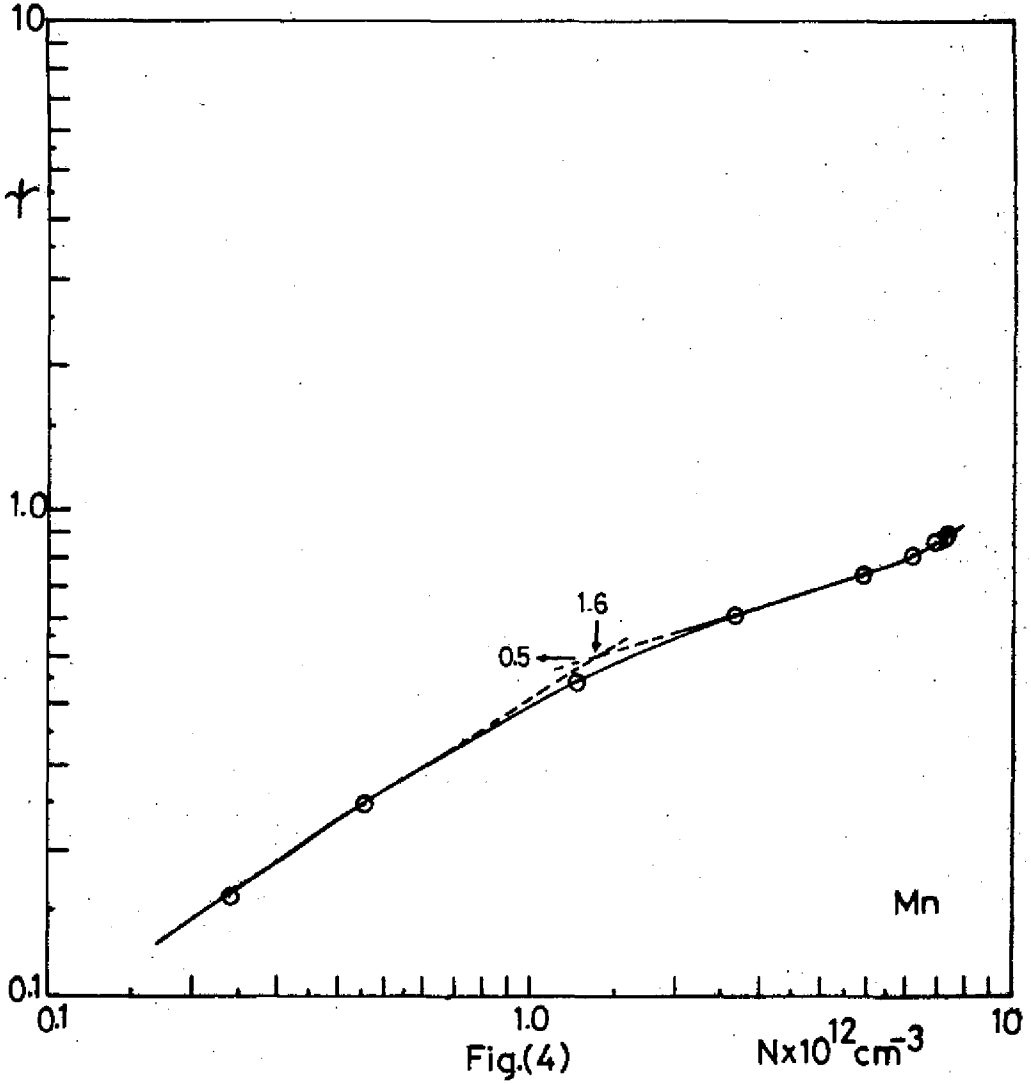
1.0
10
N x 10^11 cm^-3

Fig.(3)


10
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τ
1.6
0.5
Mn
0.1
1.0
10
N x 10^12 cm^-3

Fig.(4)

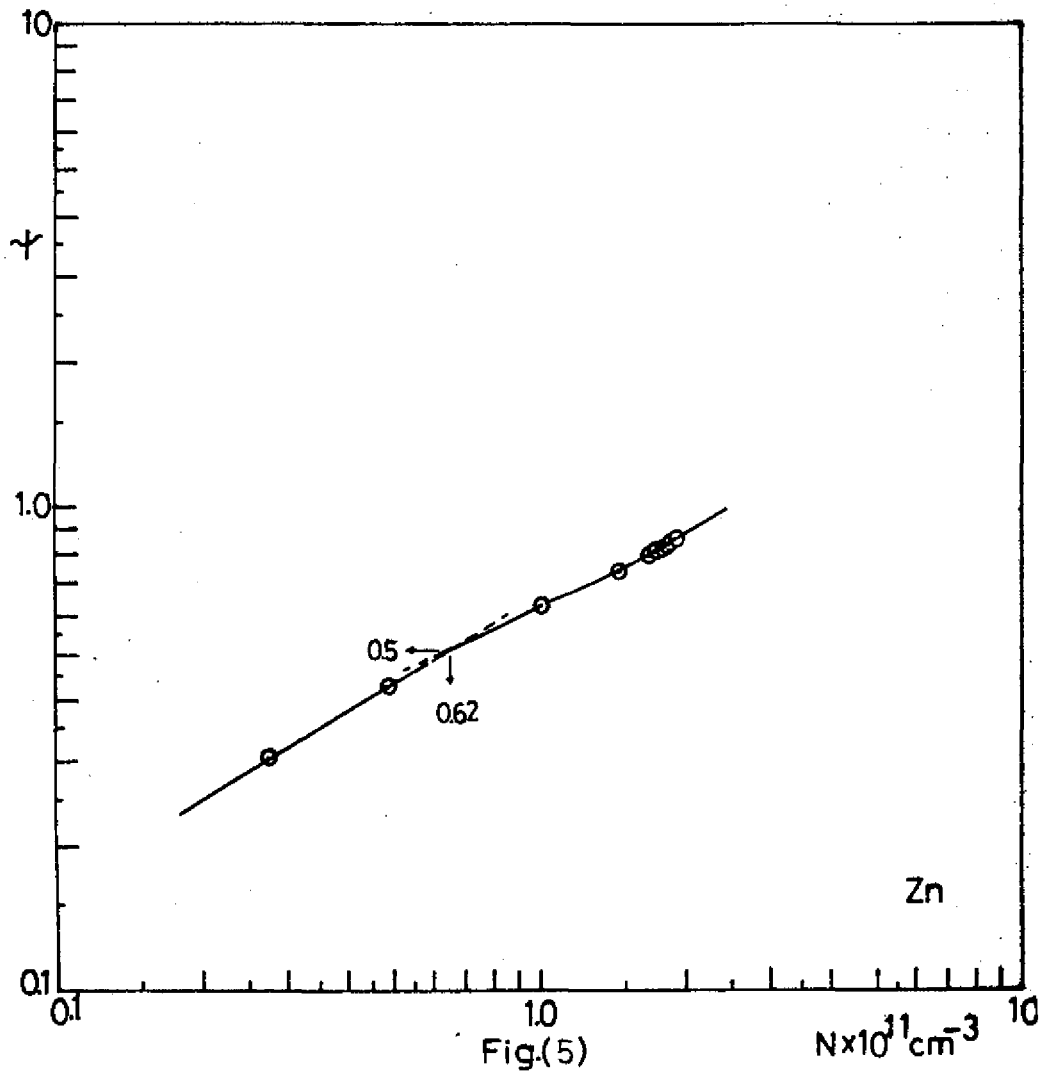

Fig.(5)

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