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PHOTOSENSITIVE DOPANTS FOR LIQUID ARGON

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Abstract

Two photosensitive dopants for liquid argon are discussed. Trimethylamine is shown to be efficient for converting the excitons into collected charge, as well as approximately 40% of the recombination photons. A greater improvement in energy resolution, for 5.5MeV alphas, is measured which is greater than can be accounted for by the increase in charge collected. Possible applications are also discussed.

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1. Introduction

Although ionization chambers filled with a room temperature liquid are presently receiving much attention[1], liquid argon, LAr, is still a more practical material for large application such as calorimetry in high energy physics. Until recently, LAr has seldom been considered for experiments with heavy ions, because the high charge densities produced result in most of the electrons being lost to recombination[2,3] before they can be collected.

It was first suggested by Doke[4], in an excellent review on the properties of liquid noble gases, that a new type of LAr chamber for heavy ions could be constructed. The signals from ionization and from scintillation would be added to give the energy of the particle. The scintillation signal, collected by a photomultiplier, is first weighted to correct for light collection efficiencies. Since the recombination of electrons in LAr gives rise to scintillation, the two signals compensate each other.

The validity of this idea has been recently demonstrated by Doke et. al.[2] They have shown that a linear combination of the signals of scintillation and ionization is proportional to the energy deposited in LAr by all types of relativistic particles, at least for nuclear charge $Z \leq 26$.

In this paper we present a different approach to utilizing the scintillator light from LAr. By using photosensitive dopants, the recombination light can be converted to collected charge directly.

2. Doping LAr

To date there have been two types of dopants added to LAr. The first are molecules such as methane or ethylene. These molecules "cool" the excess electrons in the LAr and thus improve electron mobility, while reducing diffusion. Improvements of greater than a factor of two in mobility have been achieved in this way.[5]

The second type of dopant for LAr increases the ionization yield due to the existance of excitons in noble liquids. This is similar to the Penning effect[6] in gases where an admixture gas, with a low ionization potential, is added to a noble gas to convert excited states into free charge. In this case the addition of 1.6% xenon increases the collected charge by 13%[7].

LAr produces copious amounts of UV photons, particularly at low electric fields and for heavily ionizing particles such as alphas and heavy ions. These photons have a mean wavelength of 1300A (9.5eV)[4] and are due to excitons and to recombination. This will be discussed in greater detail later.

There are several potential dopants that are known to have low ionization potentials, I_g, in the gas phase and quantum efficiencies in the 50-80% range. The three that have been the most used in photosensitive proportional counters are tetrakis(dimethylamino)ethylene (TMAE), triethylamine (TEA), and benzene.[8,9] Trimethylamine (TMA) has also been used in photoionization detectors. These materials have I_g values of 5.36eV[10], 7.5eV, 9.13eV and 7.82eV[11], respectively.

When such materials are added to LAr, their photoionization thresholds, E_{TH}, become considerably lower than in the gas phase. The relationship between E_{TH} and I_g is given by:[12]

$$E_{TH} = I_g + V_o + P_+, \qquad (1)$$

where V_o is the ground state energy, with respect to the vacuum, of a free election in the liquid. P_+ is the polarization energy of the positive ion.

To evaluate P_+, the Born equation can be used to give[10,13]:

$$P_+ = (-e^2/2R)\ (1-1/\varepsilon_{op}), \qquad (2)$$

where ε_{op} is the optical dielectric constant of LAr, R is the radius of the sphere of the positive dopant ion, and e is the electric charge.

Since the thresholds for the materials under consideration are well below the emission energy of LAr, it is sufficient to know that the value of $V_o + P_+$ is on the order of -0.7eV. This is assuming V_o = -0.21eV[1] for LAr, and taking typical values of R from the literature.[10,14] Thus E_{TH} is about 0.7eV less than I_g for these photosensitive materials in LAr.

Although there is some information available on the solubility of hydrocarbons in LAr[15], benzene is the only material in our list that is discussed in the literature. The only guide we have in selecting potential new candidates for dopants is that they must be non polar. Besides solubility, a second question that one is forced to ask experimentally is whether the photoelectric effect is efficient for these molecules in solution.

3. Experimental Setup

In order to test the behavior of the doped LAr, we used two radioactive sources. The first was an Am241 source providing 5.5MeV alpha particles. This source was in an ionization chamber with a 1.4mm gap.

The second source was a Ru106 source providing betas with a maximum energy of 3.5MeV. The chamber used for tests with betas had a gap of 1.9mm and was followed by a second ionization chamber used in

coincidence with the first. By triggering on betas that deposit a substantial amount of energy in the second chamber, only minimum ionizing electrons were studied. Unfortunately, we had a very weak source and thus were forced to use a large acceptance angle for the betas, giving a fairly wide pulseheight distribution. For this reason most of our data has been taken with the alpha source.

The dewar was made of glass with a 1cm vertical strip removed from the silvering so that the LAr could be observed. This proved very useful for determining if materials were freezing out, and how much was in colloidal suspension. A drain was also provided so that a fraction of the doped LAr could be removed before more argon was added.

The argon used was condensed directly from the gas, taken from dewars of LAr. We found that the performance was indistinguishable from gas first purified with a Hydrox purifier.

For all dopants, except TMAE, the evacuated dewar was filled with the dopants to a fraction of an atmosphere and then the argon was condensed. The volume of the system was 4.25 liter with 0.8-1.0 liter occupied by the LAr. TMAE was introduced by bubbling argon through it. The TMAE was condensed out onto a cold plate in the volume normally occupied by the LAr. After enough TMAE was transferred, the LAr fill was started.

4. Performance - Betas

There were six dopants tested: TMAE, TEA, TMA, dimethylamine (DMA), benzene and acetone. Of the above only TMAE, TEA, TMA and DMA showed an increase in charge collected with the beta source. From work done later with alphas, only TEA and TMA interacted through the photosensitive effect. TMAE and DMA are believed to give an enhanced charge yield due to excitons.

Figure 1 shows the charge collected as a function of electric field for pure LAr and LAr doped with TEA and with TMA. Betas, are minimum ionizing, and yield a low charge density. Thus, at fields greater than about 1.4 $kVmm^{-1}$ almost all the charge is collected, and the enhancement is primarily due to excitons which can yield ionization directly or through an intermediate photon.

The ratio of excited atoms, N_{ex}, to the number of initial ions, N_i, is:[16]

$$\frac{N_{ex}}{N_i} = 0.21. \qquad (3)$$

The charge collection enhancement at high electric fields is about 4.5% and 7.4% for TEA and TMA respectively. Enhancements similar to that of TEA were also seen for TMAE and DMA but were not seen for these materials with alphas.

5. Performance-Alphas

Alpha particles are more useful than batas for studying photosensitive dopants, because the high charge densities give rise to recombination and thus photons. At an electric field of $1kVmm^{-1}$, 90% of the electrons are lost to recombination.[17] One can make an estimate of the efficiency of photon production by recombination from alpha particles in LAr. For the calculation we assumed that the quantum efficiencies η_i and η_{ex} for, photon production and for recombination excitons are equal[16]m ie:

$$\eta_i = \eta_{ex} \tag{4}$$

For alphas there is an additional reduction in efficiency by a factor of 0.71 because of quenching of the light not seen for betas or heavy ions.[2] Thus the number of photons available for conversion in pure LAr is given by:

$$N_{PH}(E) = 0.71\ \eta_i\left[\frac{E\alpha(eV)}{23.6} - Q_c(E)\right] \tag{5}$$

where E_α is the energy of the alpha particle and $Q_c(E)$ is the electric field dependent charge collected measured in electrons.

In order to evaluate Eqn. (5) we assumed a value of $\eta_{ex} = 0.4$, which may be high.[18] Thus with $E_\alpha = 5.5 \times 10^6 eV$ we have:

$$N_{PH}(E) = 1.32 \times 10^5 - 0.57\ Q_c(E). \tag{6}$$

5.1 Charge Collected

The charge collected as a function of electric field for pure LAr and for three levels of doping with TMA are shown in Fig. 2. For Curve B the dewar was filled with 12.2 Torr of TMA and then filled with 0.8 liter of LAr. This filling was very colloidal. The concentration was then reduced in several steps. No colloids were seen at a concentration of about 1.7×10^{16} cm^{-3}.

Curves B and C of Fig. 2 can be fitted reasonably well with:

$$Q_B(E) = Q_1(E) + 5.1 \times 10^3$$

$$Q_C(E) = Q_1(E) + 2.7 \times 10^3, \tag{7}$$

where $Q_1(E)$ is the extrapolation from the high field region. Likewise, Curve D can be fitted:

$$Q_D(E) = Q_2(E) + 2.4 \times 10^3. \tag{8}$$

We believe the constant, field independent term, is due to excitons, since a reduction in concentration reduces this efficiency while not affecting the field dependent term due to photons. From Eqn. (3) we can calculate that tne number of excitons available to be about 4.9×10^4. Thus the conversion efficiency, for excitons, is about 10% at the highest concentration of TMA.

By comparing Curve 1 with Curve A, and using Eqn. (6), we are able to estimate the conversion efficiency of photons into detected electrons. Since the range of the alphas is only about 40μm, we assume

that half the light is lost into the radioactive source so we use:

$$\text{Efficiency} = \frac{Q_1(E) - Q_A(E)}{0.5\ N_{PH}(E)} \qquad (9)$$

Figure 3 shows our calculated efficiency as a function of electric field, with a peak efficiency of 41%. The efficiency increases with electric field, as expected, and reaches a maximum at about 1.0 kV^{-1}. This response is similar to betas since both have low recombination. We do not know if the turnover in efficiency at high fields is real or an artifact. The estimate of efficiency depends heavily on our estimate of $\eta_{ex} = 0.4$. If it is lower, as the literature suggests,[18] the efficiency would be higher.

The difference between Curve 1 and Curve 2 in Fig. 2 is believed to be loss of photons to the other side of the chamber. The efficiency for Curve 2, with a concentration of 1.1×10^{15} cm^{-3}, is 70% of the efficiency for higher concentrations. Assuming that the reduction is due to photons being lost to the second electrode, and the charge reduced because the photoelectrons do not cross the entire gap, we calculate an absorption length for this concentration of about 1mm. For a concentration of 4.6×10^{15} cm^{-3}, the absorption length would be 0.18mm. If TMA saturates LAr at 1.7×10^{16} cm^{-3}, the absorption length would be about 65μm.

The problem with this calculation is that it yields a cross section of almost 10^3 Mb. This is 30 times larger than anticipated. We feel that our estimate of the first absorption length is good to a few per cent and our value for the concentration cannot be that far off. At present cannot resolve the discrepancy.

Figure 4 shows the charge collected for pure LAr and for two dopings of TEA ($9.8 \times 10^{17} cm^{-3}$ and $2.4 \times 10^{17} cm^{-3}$). The doped curves have a similar structure to those of TMA, but with only about 60% of the efficiency. This could be due to a poorer match between the absorption spectrum of TEA and the emission spectrum of LAr, or to very low solubility.

5.2 Energy Resolution

An unforseen advantage of photosensitive dopants is that the energy resolution of the 5.5 MeV alphas was improved, more than is accounted for by the increase in charge collected. This can be seen in Fig. 4, where the energy resolution is plotted as a function of charge collected for pure LAr and LAr doped with TMA (7.1×10^{17} cm^{-3}). The improvement is due to the fact that the light produced is related with the amount of charge lost to recombination. Thus fluctuations in the number of electrons that escape recombination are somewhat compensated by the photoelectrons. The fits to the pulse height spectra are 21% and 16% narrower for the doped LAr at $0.7 \times 10^4 e$ and $4 \times 10^4 e$ respectively.

6. Conclusion

The development of photosensitive dopants suggest several directions for futher effort. The first is for calorimetry for heavy ion physics. For relative heavy ions, there is 40% more scintillation light available, because of reduced quenching. With the events occuring deep in the LAr, light would not be lost to the structure as in our test. Thus, a substantial amount of the collected charge will be due to photoelectrons. This should further enhance the energy resolution yielding a massive calorimeter without the complications of photomultipliers and optical windows.

A more speculative direction of study would be to dope liquid noble gases in order to get charge gain around a wire. Since photosensitive dopants have been shown to work for LAr, it seems likely that they will also work with LXe. With a value of $V_o = -0.67eV$,[1] the thresholds of TEA and TMA will be around 6.3eV and 6.6eV, respectively. These are below the 7.1eV (1750A) mean photon energy from LXe. Since it has been shown that proportional scintillation is possible around a wire in LXe,[19] this light could be converted into charge giving substantial gain at much lower electric fields than in pure LXe. This would yield an interesting instrument for imaging α rays in such fields as α-ray astronomy and nuclear medicine.

Since the effect of photosensitive dopants is greater for heavily ionizing particles than for minimum ionizing particles, they may be an interesting probe for LAr calorimetry. By observing the change in charge collected for electromagnetic and hadronic events, one may be able to estimate the amount of charge lost due to saturation in hadronic events. This saturation may also be partially compensated for by the dopant.

Finally there should be a continuing effort to find better photosensitive dopants with higher efficiencies and higher solubilities. With a higher solubility, the dopant could both convert the scintillation light and speed up the electron drift. It seems unlikely that the best materials have been found in the first effort.

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Figure Captions

Figure 1 - Charge collected as a function of electric field for pure LAr, and LAr doped with TEA and TMA.

Figure 2 - Charge collected for 5.5 (5 FWHM) MeV alpha particles for pure LAr and LAr doped with three different concentrations of TMA. The concentrations of TMA. The concentrations are given in molecules cm^{-3}.

Figure 3 - Recombination photon conversion efficiency for TMA as a function of electric field.

Figure 4 - Charge collected for 5.5 MeV (% FWHM) alpha particles for pure LAr and for two concentrations of TEA. The concentrations are given in molecules cm^{-3}.

Figure 5 - Energy resolution for 5.5 MeV alpha particles as a function of collected charge for pure LAr and LAr doped with TMA (7.1×10^{17} cm^{-3}).

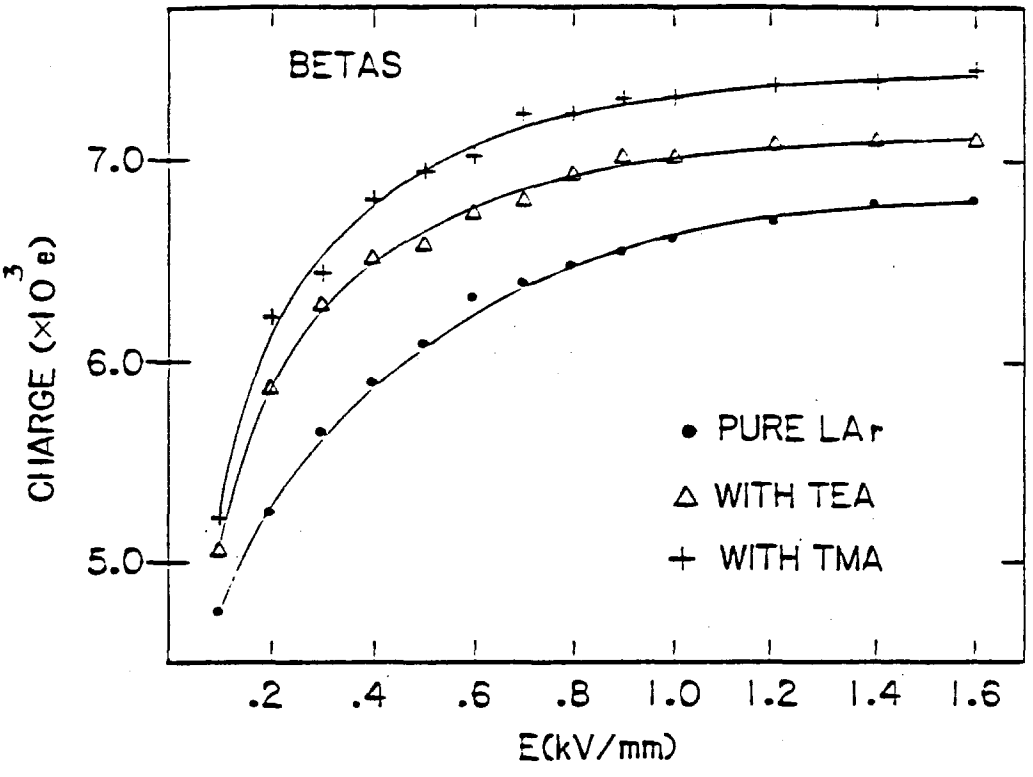

Figure 1

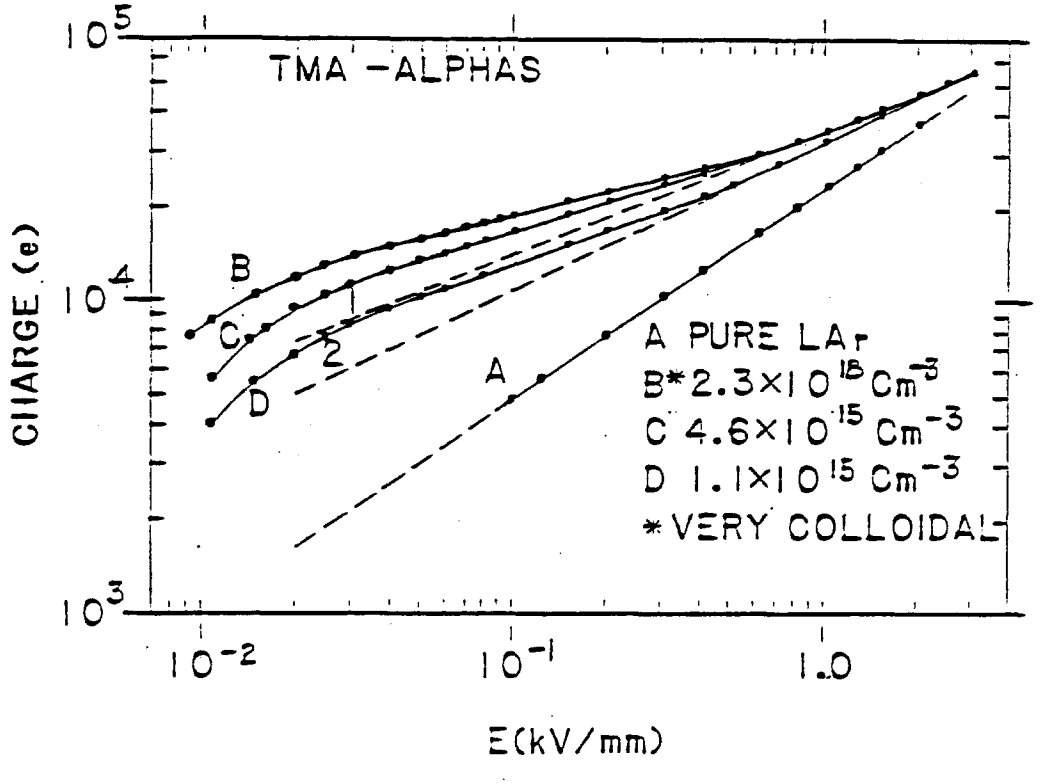

Figure 2

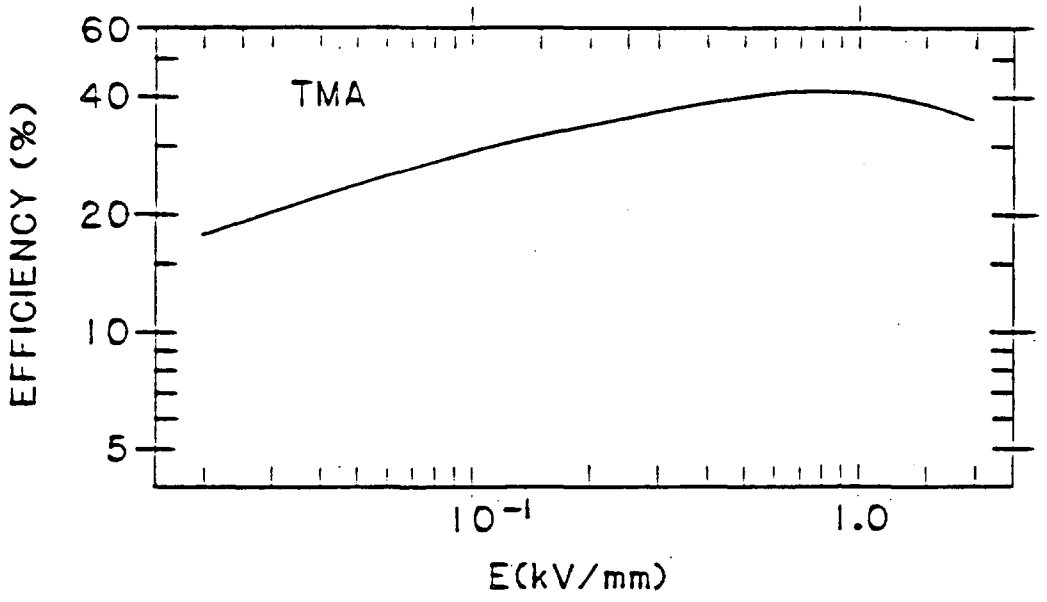

Figure 3

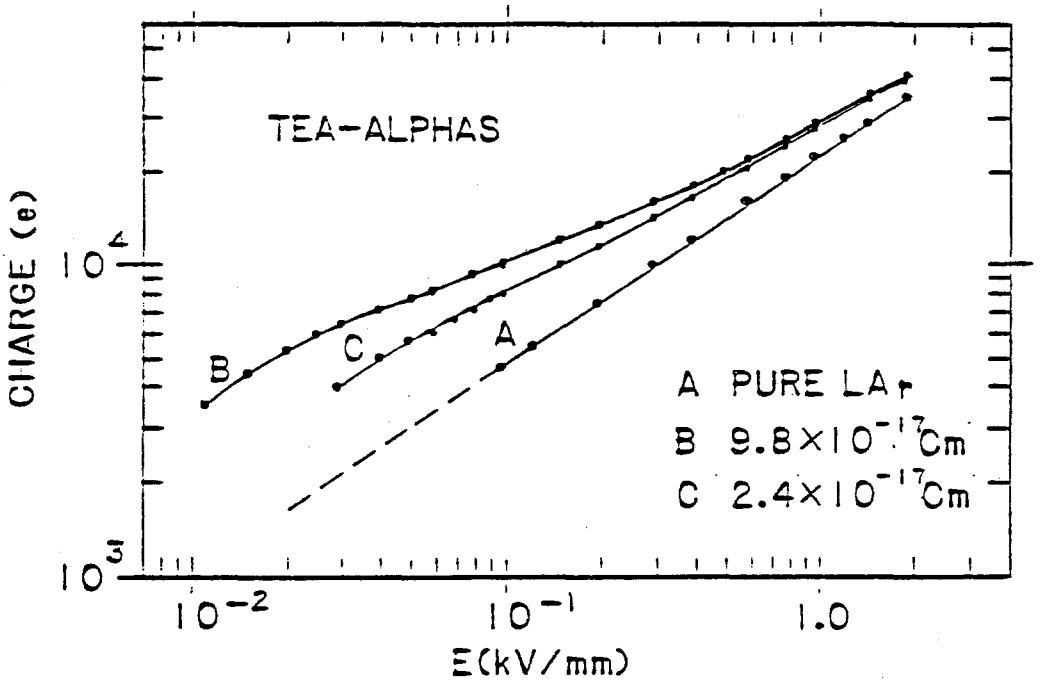

Figure 4

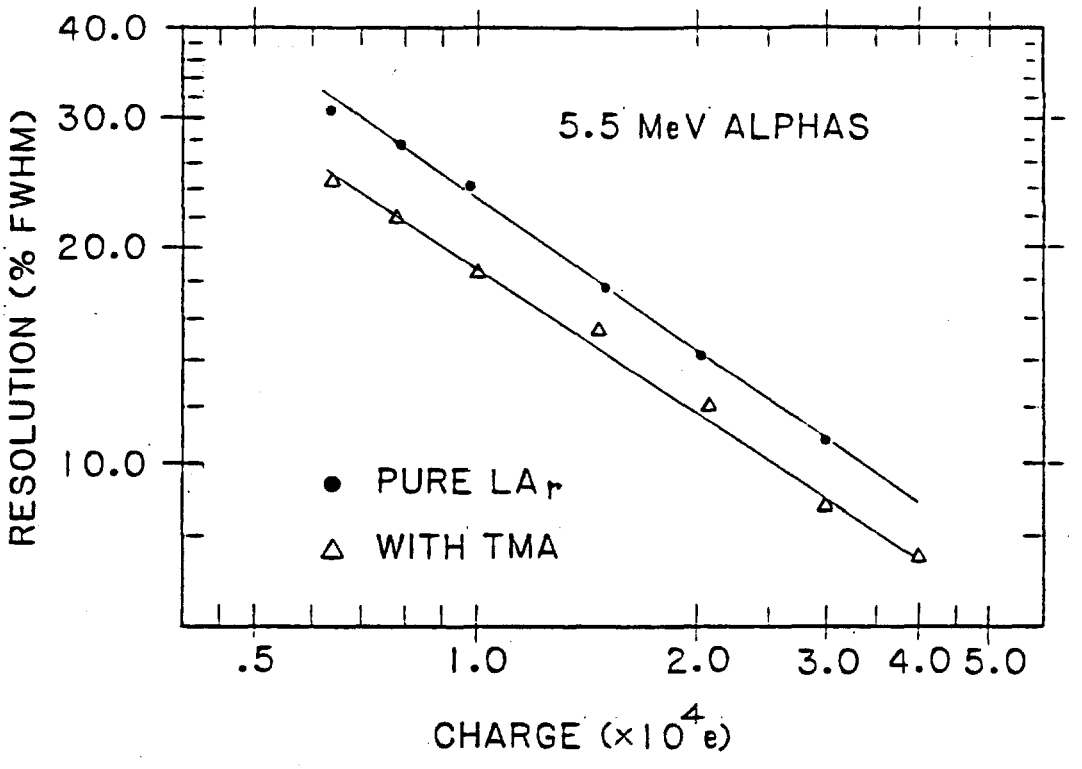
40.0
30.0
20.0
10.0
RESOLUTION (% FWHM)
5.5 MeV ALPHAS
PURE LAr
WITH TMA
.5
1.0
2.0
3.0
4.0
5.0
CHARGE (×10⁴e)

Figure 5