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ANGULAR CORRELATIONS OF EXCITED  $^{41}\text{K}$  NUCLEI  
RECOILING IN ARGON GAS

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THE MEASUREMENT OF PRESSURE DEPENDENT PERTURBED ANGULAR  
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ABSTRACT

The time integrated perturbed angular distribution of the 1294 keV  $\gamma$ -transition, deexciting the recoiling  $^{41}\text{K}$  excited nuclei, was measured as a function of argon gas pressure between 20 torr and 1000 torr. The pressure dependence presents a strong minimum around 100 torr. The magnetic hyperfine interaction responsible for the perturbation changes randomly due to inelastic charge exchange collisions, with a pressure dependent and time dependent correlation time  $\tau_c$ .

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I. INTRODUCTION

The pressure dependence of the perturbed angular correlation (PAC) of excited nuclei recoiling in gas is a well established experimental fact<sup>(1)</sup>. In the early measurements, where energetic heavy ions recoil in gas, the number of possible charge states and atomic levels is so large, that a calculation of the expected perturbation was not feasible. For lighter systems, Hagemeyer et al.<sup>(2)</sup> and Brenn et al.<sup>(3)</sup> have tried to relate the observed dealignment to experimentally measured atomic collision cross sections. The observation time in most of these experiments is much shorter than the stopping time and constant velocity can be assumed. The correlation time  $\tau_c$ , defined as the mean time interval between atomic collisions is also constant in these situations. The perturbation factor calculations performed on these experimental data are usually based on randomly varying time-dependent hyperfine interactions, described by the Abragam-Pound<sup>(4)</sup> model in the very fast variation limit ( $\tau_c \ll \tau$ ), or by the Scherer<sup>(5)</sup>-Blume<sup>(6)</sup> model for an arbitrary, but constant  $\tau_c$ . The Scherer-Blume theory predicts a minimum in the pressure dependence of  $G_k$ , as shown by Zemel and Niv<sup>(7)</sup> and da Silveira and collaborators<sup>(8)</sup>, when the collision frequency  $\lambda = 1/\tau_c$  is the same as the mean hyperfine frequency.

In this paper we present our experimental results

on the time integrated perturbation factor, measured as a function of the gas pressure, where a clear minimum is seen in the pressure dependence. In our experiment a simple atomic system ( $^{41}\text{K}$ ) is recoiling at low velocity (27 keV) in a noble gas (Ar) so the atomic cross sections are all well-known, as well as the hyperfine fields and correlation times. Due to the low recoil velocity, the stopping time is comparable to the nuclear life time and the correlation time  $\tau_c$  is not constant.

In a recent paper<sup>(9)</sup> we presented the adiabatic extension of the Scherer-Blume theory, also applicable to non-stationary regimes with variable  $\tau_c$ . The atomic processes relevant to the perturbation mechanism and the theoretical calculations about perturbation factors in the context of these models will be treated in a subsequent second paper, in order to become more comprehensible. In this paper we present a description of the nuclear reaction and the experimental data, together with a simple physical interpretation of results based on some experimental results of atomic processes.

## II. EXPERIMENTAL METHOD

Due to the low recoil energy and the knowledge of the nuclear and atomic states and transitions and atomic collision cross sections, we choose the excited  $^{41}\text{K}$  nucleus

to study its perturbed angular correlation when recoiling in Ar gas. The second excited state of  $^{41}\text{K}$  nucleus at 1294 keV has a relatively long nuclear life-time ( $\tau = 10.7$  ns), spin  $7/2^-$  and  $g = 1.26 \pm 0.01$ <sup>(10)</sup>. It decays directly to the  $3/2^+$  ground state of  $^{41}\text{K}$  nucleus by the emission of an almost pure M2  $\gamma$ -transition.

The  $^{41}\text{K}$  nucleus was formed at 8.88 MeV excitation energy in the  $^{40}\text{Ar}(p,\gamma)^{41}\text{K}$  reaction at  $E_p = 1.1$  MeV using the proton beam of the São Paulo Van de Graff accelerator<sup>(11)</sup>. This proton capture reaction presents isobaric analog resonances (IAR) at proton energies of 1.10, 1.58, 1.87, 2.45 and 2.90 MeV, corresponding to excited states in the  $^{41}\text{Ar}$  nucleus. The IAR at  $E_p = 1.1$  MeV has spin  $3/2^-$  and isospin  $T = 5/2$ <sup>(12)</sup> and is fragmented over many narrow resonances, the most intense of them at 1086,5 keV, 1101,8 keV and 1108,4 keV. The spins of these most intense resonances have been determined as  $3/2^-$ <sup>(12)</sup>.

In our measurements the Ar target was a gas cell limited by a Mo entrance foil (2.5  $\mu$  thick) and a Ta beam stopper. The incident protons lose  $250 \pm 40$  keV in the Mo foil as verified by the excitation curves extrapolated to zero gas pressure. All the fine structure components of the IAR were populated simultaneously. The gas pressure was varied between 20 and 1000 torr and the length of the gas cell was varied inversely with gas pressure in order to maintain the energy dispersion of the protons in the gas target less than 350 keV and guarantee that the IAR of 1.58 MeV is not populated

simultaneously with the IAR of 1.10 MeV. At the same time the length has to be sufficiently large for all the recoiling ions to stop in gas. Every angular distribution measurement was preceded by an excitation function to verify that the IAR was populated with the highest yield in the given experimental situation and no other IAR were populated in the reaction. In Fig. 1 we present our excitation curve measured with thick gas target, where the 1.10 MeV IAR is well resolved from the subsequent IAR at 1.58 MeV.

The IAR at  $E_p = 1.10$  MeV is located at 8.88 MeV excitation energy in the  $^{41}\text{K}$  nucleus and does not decay directly by  $\gamma$ -emission to the second excited state of  $^{41}\text{K}$  at 1294 keV. The deexcitation of the 8.88 MeV state has been well studied by several authors<sup>(11,13,14)</sup> and the population of the 1294 keV state ( $I^\pi = 7/2^-$ ,  $\tau = 10,7$  nsec,  $g = 1.26 \pm 0.01$ )<sup>(10)</sup> proceeds through several  $5/2^+$  spin states around 3 and 4 MeV excitation energy. The decay scheme populating the 1294 keV state is shown in Fig. 2. The first excited state of  $^{41}\text{K}$  at 980,4 keV has  $I^\pi = 1/2^+$  and its isotropic  $\gamma$ -transition to the ground state was used as an internal normalization of the spectra.

The  $\gamma$ -rays were detected in a true-coaxial Ge-Li detector ( $40 \text{ cm}^3$ ) with 2.4 keV resolution for the  $^{60}\text{Co}$  peak, located at 8 cm from the center of the target and the detection angles were  $0^\circ$ ,  $54^\circ$  and  $90^\circ$ . Corrections were made to account for absorption in the walls of the gas cell, finite extension

of the source, finite extension of the detector and the errors introduced by these corrections were less than 3%.

The angular distributions of all  $\gamma$ -transitions with energies between  $E_\gamma = 980$  keV and 1698 keV were measured, varying the gas pressure and the length of the gas cell, and consequently the energy dispersion of protons in the gas target. The pressure was measured directly by a mercury manometer, using as reference, the vacuum for low pressure measurements and the atmospheric pressure for high pressure measurements.

The unperturbed angular distribution was measured with  $^{40}\text{Ar}$  target implanted into Ta backing, where the hyperfine fields are zero due to cubic (bcc) crystal structure of Ta. Due to the reduced quantity of Ar present in these implanted targets, NaI(Tl) detectors were used to detect the  $\gamma$ -rays.

### III. EXPERIMENTAL RESULTS

#### III.1. IMPLANTED TARGET MEASUREMENTS

The unperturbed angular distribution was measured only at  $0^\circ$  and  $90^\circ$  due to the low yield with implanted targets. With the use of thin implanted Ar targets, the fine structure of the IAR was easily separated due to the much better energy resolution in the entrance channel. The measurements were made at the fine structure resonance of  $E_p = 1102$  keV with spin  $3/2^-$ .

Due to the initial spin value, the subsequent  $\gamma$ -transitions should have  $A_4 = 0$  and measurements at two angles should be sufficient to fix the value of  $A_2$ . The 1294 keV  $\gamma$ -transition was easily separated from the others even with the NaI(Tl) detector. The  $A_2$  anisotropy coefficient obtained, assuming  $A_4 = 0$ , was:

$$A_2 = 0.29 \pm 0.04$$

in good agreement with the results of Levy<sup>(10)</sup>:

$$A_2 = 0.279 \pm 0.028 \quad \text{and} \quad A_4 = -0.045 \pm 0.30$$

### III.2. GAS TARGET MEASUREMENTS

In order to see the influence of the bad energy resolution in the entrance channel, when gas target is used, and the effect of the possible population of other fine structures of the IAR with spins different from  $3/2^-$ , the length of the gas cell and pressure were varied and the  $A_2$  and  $A_4$  of all  $\gamma$ -transitions were measured as a function of  $\Delta E_p$ , the energy dispersion of protons in the gas target. These results are shown in Figs. 3, 4 and 5. All the  $A_4$  values are practically zero, even when the proton energy dispersion is as large as several hundred keV, and the  $A_2$  values of all transitions, with the exception of the 1294 keV transition, are also constant with  $\Delta E_p$  and pressure, within their uncertainty. The coefficients

$A_2$  of the 1294 keV transition, measured with constant pressure (530 torr) and variable gas cell length, present no variation with  $\Delta E_p$ , even when  $\Delta E_p$  is as large as 350 keV (see Fig. 5).

The fact that the  $A_2$  values do not depend on the proton energy dispersion  $\Delta E_p < 350$  keV or with the number of fine structure components of IAR populated in the proton capture reaction, means that the components included have spin  $3/2^-$  and no intense components with spin different from  $3/2^-$  are populated, even when  $\Delta E_p$  is as large as 350 keV. We can use the initial spin as  $3/2^-$ , even populating many components of the IAR and have confidence in our  $A_2$ , even when  $\Delta E_p$  or the pressure is increased.

### III.3. PRESSURE DEPENDENCE OF THE ANGULAR DISTRIBUTION OF THE 1294 keV TRANSITION

The anisotropy coefficients  $A_2 G_2(\infty)$  of the 1294 keV transition show a strong variation with pressure, presenting values always smaller than the unperturbed value  $A_2$ . The attenuation of the anisotropy is represented by the time integrated perturbation factor  $G_2(\infty)$  obtained by dividing the measured  $A_2 G_2(\infty)$  by the unperturbed value of Levy<sup>(10)</sup>  $A_2 = 0.279 \pm 0.028$ . The pressure dependence of  $G_2(\infty)$  is shown on Fig. 6 and it presents a pronounced minimum around 100 torr, with a strong increase for high pressures and a slight increase for lower pressures. This variation cannot be due to

changes in the population of the IAR, as discussed above.

The attenuation of the anisotropy is due to the deorientation of the  $^{41}\text{K}$  nucleus in its long-lived second excited state, caused by the magnetic hyperfine interaction between the nuclear spin  $I=7/2$  and the electronic spin  $j$ . This hyperfine interaction is time dependent because the electronic spin  $j$  can change randomly due to inelastic collisions between the K and Ar atoms. The mean time interval between inelastic collisions, called correlation time  $\tau_c$  is inversely proportional to the pressure and so the time dependent behaviour of the interaction is transformed into a pressure dependence of the anisotropy.

This behaviour of  $G_2(\infty)$  can be understood taking into account the relevant atomic processes contributing to the perturbation mechanism. A detailed description of these processes and calculation of  $G_2(\infty)$  will be presented in a subsequent paper. However a simple explanation based on the most relevant facts will be given below.

#### IV. A SIMPLE EXPLANATION OF THE PRESSURE DEPENDENCE OF $G_2(\infty)$

The recoil energy of the  $^{41}\text{K}$  ion is of 27 keV and the stopping times were calculated using the Biersack-Ziegler<sup>(15)</sup> stopping power  $dE/dx$  and defining the time interval necessary

to lose the kinetic energy from the initial value  $E_0$  to  $\epsilon$  as

$$t_s = \sqrt{\frac{m}{2}} \int_{E_0}^{\epsilon} \frac{dE}{\sqrt{E} \frac{dE}{dx}}$$

This stopping time is inversely proportional to the gas pressure and its values are for  $E_0 = 27 \text{ keV}$  and  $\epsilon = kT$ :  $t_s = 10.5 \text{ ns}$  for 100 torr,  $t_s = 1.05 \text{ ns}$  for 1000 torr. This means that for low pressures ( $P < 100 \text{ torr}$ ) the ions are slowing down during the whole nuclear lifetime, presenting a strong time-dependence in the correlation time  $\tau_c$ , aside from the obvious pressure dependence.

The equilibrium charge distribution of K beams in argon gas, measured for recoil energies ranging from 52 keV to 135 keV<sup>(16)</sup>, shows that for low energies ( $E_0 < 82 \text{ keV}$ ) the only charge states present in the beam are the  $\text{K}^0$  (neutral K atom) with 18% of probability and the  $\text{K}^+$  (singly ionized K ion) with probability of 82%. Atomic charge exchange cross section measurements<sup>(17)</sup> confirm that the cross sections for capturing electrons from charge state +3 to +2, from +2 to +1 ( $\sigma_{32}, \sigma_{21}$ ) are much larger than any other and the ionization cross sections  $\sigma_{12}, \sigma_{02}, \sigma_{13}, \sigma_{23}$  are very small, so very quickly the ion recaptures its electrons and will spend its lifetime in neutral or singly ionized states. The ionization cross section  $\sigma_{01}$  is 4 times larger than the electron capture cross section  $\sigma_{10}$  and so any ion will spend 80% of its time in the singly ionized

state  $K^+$  and only 20% in the neutral state  $K^0$ . As the K atom is stopping,  $\sigma_{01}$  and  $\sigma_{10}$  fall to zero and the atoms cannot change their charge anymore, so one has 80% of the thermal atoms in the  $K^+$  state and only 20% in the neutral state, at least during a time interval of the order of the nuclear lifetime.

The hyperfine magnetic fields are also known:

$$B(K^0, 4^2S_{3/2}) = 5.8 \times 10^5 \text{ G} \quad \omega_{FF'} = 28 \text{ GHz}$$

$$B(K^0, 4^2P_{1/2}) = 0.72 \times 10^5 \text{ G} \quad \omega_{FF'} = 3.5 \text{ GHz}$$

$$B(K^0, 4^2P_{3/2}) = 0.46 \times 10^5 \text{ G} \quad \omega_{FF'} = 0.81 \text{ GHz}$$

$$B(K^+, 3P_6) = 0 \quad \omega_{FF'} = 0$$

We assume for the moment that due to their much higher excitation energy, the  $K^+$  ions remain mostly in their ground state with zero hyperfine field.

With these assumptions the increase of  $G_2(\infty)$  with pressure can be easily explained. With increasing pressure the atoms stop more rapidly and during their slowing down are less perturbed due to the rapid variation of hyperfine field and lower  $t_s$ . After the stopping time  $t_s$  the majority of ions do not feel the perturbation mechanism due to zero field. The higher the pressure, the more quickly the ions stop and are less perturbed, increasing  $G_2(\infty)$ .

The presence of a minimum and the increase for lower

pressures can also be understood with these assumptions. The knowledge of the charge exchange cross sections  $\sigma_{01}$  and  $\sigma_{10}$  allows the calculation of  $\tau_c$ , the mean time interval between charge exchange collisions. For a pressure of 100 torr:

$$\tau_c = 2 \times 10^{-10} \text{ s}$$

for energies between 27 keV and 10 keV, while the period of the hyperfine precession in the  $K^0$  state is also:

$$T = 2\pi/\omega = 2 \times 10^{-10} \text{ s}$$

If the charge exchange collision frequency  $\lambda = 1/\tau_c$  is the same as the hyperfine frequency  $\nu = 1/T$ , a resonance effect occurs, with the collisions (time variation of the hyperfine field) inducing transitions between the magnetic substates. These transitions will modify the initial alignment and will decrease  $G_2(\infty)$ . This minimum is also predicted by Zemel and Niv<sup>(7)</sup> when  $\bar{\omega}\tau_c \sim 1$ .

For pressures lower than 100 torr, the collision frequency will be lower than the hyperfine frequency and the perturbation of the alignment will be less efficient, resulting in a higher  $G_2$  value.

## V. SUMMARY

The pressure dependence of  $G_2(\infty)$  was measured for the 1294 keV transition deexciting the second excited state of  $^{41}\text{K}$  ( $\tau = 10.7$  ns). The  $^{41}\text{K}$  was produced by the capture of 1.1 MeV protons by  $^{40}\text{Ar}$ , it has 27 keV initial energy and recoils in the Ar gas colliding with Ar atoms. The perturbation factor  $G_2(\infty)$  presents a strong minimum around 100 torr. The presence of the minimum and the increase of  $G_2(\infty)$  for higher and lower pressures can be understood with the use of atomic charge exchange processes, responsible for the time variation of the hyperfine magnetic field. Due to the behaviour of these charge exchange cross sections, 80% of the K ions are in singly ionized charge state after their slowing down. With increasing pressure the atoms stop more rapidly, are less perturbed during their slowing down and after their stopping the majority feels a zero field, resulting in an increasing  $G_2$  with pressure. The minimum in  $G_2(\infty)$  can be understood as a resonance effect, occurs when the collision frequency is the same as the hyperfine frequency, inducing transitions between the different magnetic substates of the atom.

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TABLE CAPTION

Table 1 - The time integrated perturbation factors  $G_2(\infty)$  of the 1294 keV transition were obtained dividing the measured  $A_2 G_2(\infty)$  values by the unperturbed  $A_2$  value<sup>(10)</sup>. The  $G_2(\infty)$  values are presented as a function of the Ar pressure.

TABLE 1

Pressure (torr)	$G_2(\infty)$
19	$0.30 \pm 0.11$
52	$0.142 \pm 0.061$
97	$0.101 \pm 0.050$
198.5	$0.292 \pm 0.048$
301	$0.419 \pm 0.033$
401	$0.543 \pm 0.039$
448	$0.467 \pm 0.082$
530	$0.627 \pm 0.020$
630	$0.601 \pm 0.054$
710	$0.647 \pm 0.070$
835	$0.636 \pm 0.090$
1008	$0.700 \pm 0.11$

FIGURE CAPTIONS

Fig. 1 - Excitation function of the  $^{40}\text{Ar}(p,\gamma)^{41}\text{K}$  reaction, detecting the 1294 keV  $\gamma$ -rays, deexciting the second excited state of  $^{41}\text{K}$ . The well defined peak in the yield curve corresponds to the Isobaric Analog Resonance at  $E_p = 1.10$  MeV.

Fig. 2 - The decay scheme of the IAR at 8.88 MeV excitation energy in  $^{41}\text{K}$ , showing only the population of the second excited state at 1294 keV.

Fig. 3 - The angular distribution coefficients  $A_4$  of some  $\gamma$  transitions deexciting the  $^{41}\text{K}$  nucleus, measured as a function of the energy dispersion of protons  $\Delta E_p$ .

Fig. 4 - The angular distribution coefficients  $A_2$  of some  $\gamma$  transitions deexciting the  $^{41}\text{K}$  nucleus, measured as a function of the energy dispersion of protons  $\Delta E_p$ .

Fig. 5 - The angular distribution coefficients  $A_2$  of some  $\gamma$  transitions deexciting the  $^{41}\text{K}$  nucleus measured as a function of the energy dispersion of protons  $\Delta E_p$ . The coefficients of the 1294 keV transition were measured with constant gas pressure and variable gas cell length.

Fig. 6 - The time integrated perturbation factors  $G_2(\infty)$  of the 1294 keV transition, measured as a function of gas pressure.

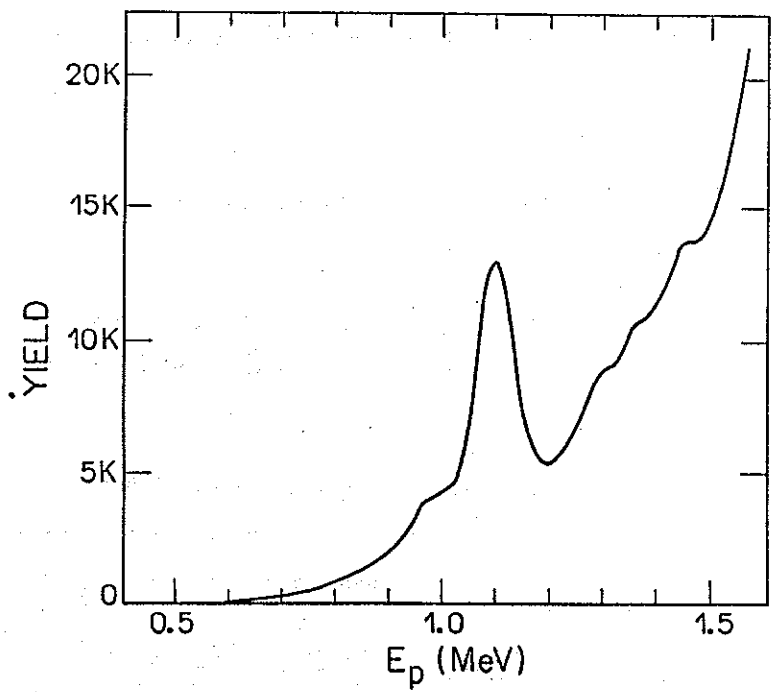


FIG. 1

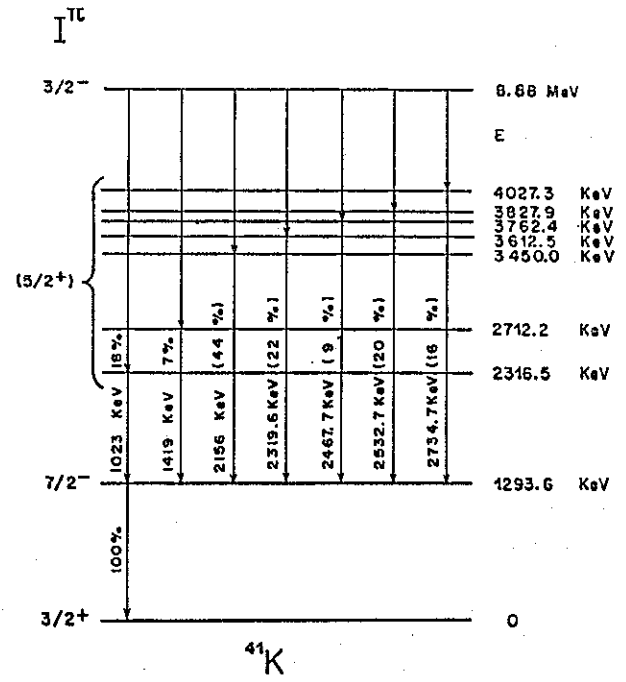


FIG. 2

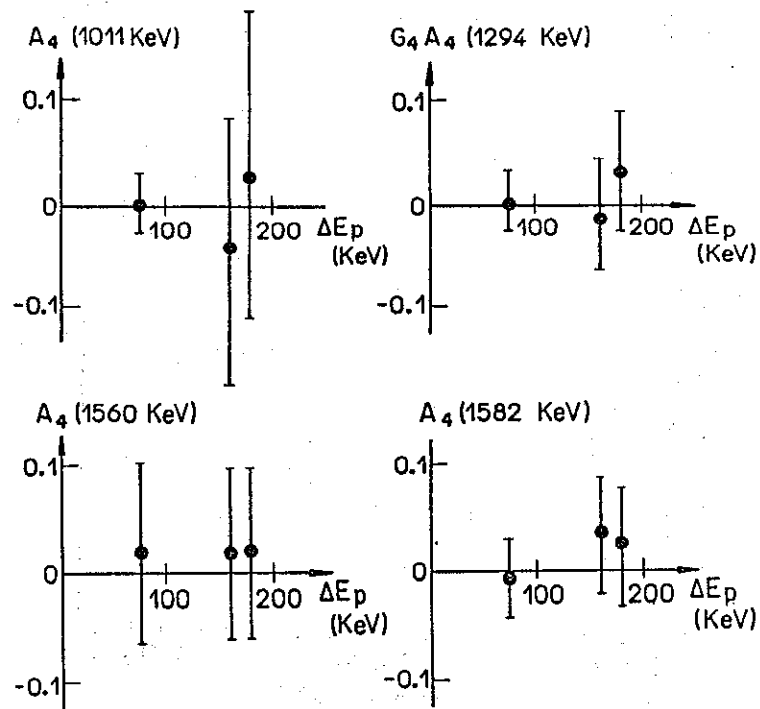


FIG. 3

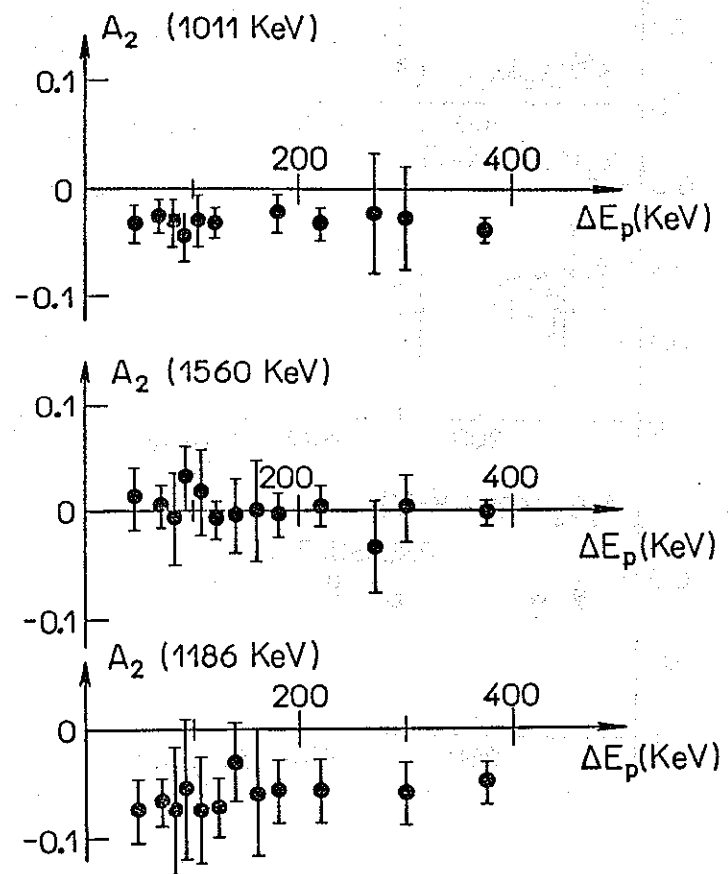


FIG. 4

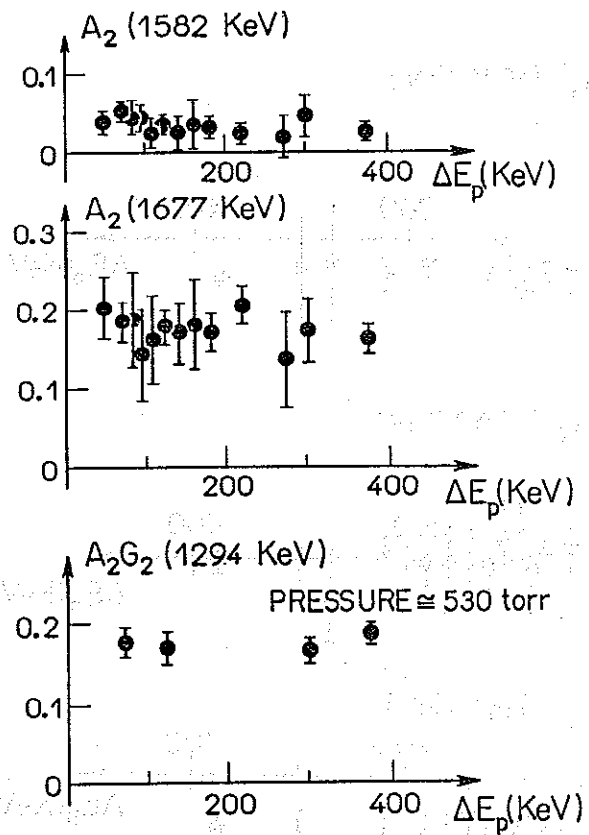


FIG. 5

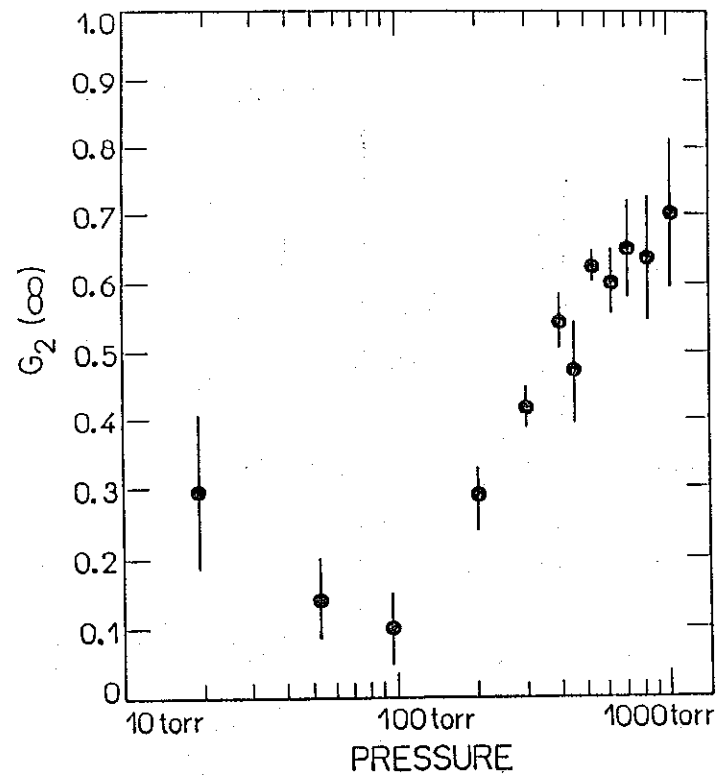


FIG. 6