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OBSERVATION OF THE 14 MeV RESONANCE IN $^{12}\text{C}(\text{p,p})^{12}\text{C}$
WITH MOLECULAR ION BEAMS*

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Abstract

The lowest $T = 3/2$ state in ^{13}N has been observed using H_2^+ and H_3^+ molecular ion beams incident on ^{12}C . The dependence of the shape of the observed anomaly on the molecular ion species and the target thickness is accounted for using the parameters of the molecular ions and a standard resonance shape determined using a proton beam. In addition to explaining the interaction of molecular ion beams with a thin target, these results provide a technique for extending precise energy calibration standards to significantly higher energies.

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1. Introduction

In the past decade the development of the isochronous cyclotron has extended the realm of nuclear spectroscopy investigations to considerably higher energies. The requirement of high-resolution experiments to obtain precise spectroscopic information has led to the construction at several cyclotron laboratories of magnetic analysis systems^{1,2,3)} which select a portion of the external beam with greatly improved energy resolution and emittance.

The problem of obtaining precise energy calibrations⁴⁾ for these high-energy beams remains. One technique⁵⁾ for calibration at higher magnetic rigidities involves measuring a neutron threshold, e.g. $^{16}\text{O}(d,n)$, using an analyzed ^{16}O beam with successively lower charge states. This extends the range of existing energy standards somewhat; however, it is limited to accelerators with heavy-ion capabilities. Another technique involves the measurement of a narrow proton resonance using protons bound in molecular ions. Parkinson and Bardwick²⁾, in a recent attempt to observe the narrow 14 MeV resonance in $^{12}\text{C}(p,p)^{12}\text{C}$ with an H_2^+ beam, conclude erroneously that the resonance cannot be observed in this way. The present observation of this resonance with both H_2^+ and H_3^+ molecular ion beams indicates that calibration points up to a magnetic rigidity equivalent to that of 128 MeV protons can be obtained. An explanation of the dependence of the shape of the resonance curve on the structure of the molecular ion and the target thickness is presented.

2. Apparatus

The present measurements were performed with proton, H_2^+ and H_3^+ beams obtained from the Berkeley 88-in. cyclotron. The beam analysis system¹⁾ consisted of two 110° uniform-field magnets operated such that the first magnet provided energy analysis and the second magnet removed the beam tail produced by scattering from the analyzing slit (located midway between the two magnets). For each beam the energy resolution (fwhm) was experimentally determined to be 0.015%. At the proton resonance energy (14.232 MeV) this corresponds to an energy resolution of 2.1 keV. The presence of a small proton contamination of the molecular ion beam which might arise from collisional dissociation following the last analyzing magnet was minimized by a set of small collimator slits located immediately in front of the target. This arrangement was particularly effective since such protons would be defocussed by a magnetic quadrupole located upstream from the scattering chamber. For the H_2^+ beam, measurements of the beam current with the target in and out of the beam place an experimental limit of $< 5\%$ on the proton contamination; the actual amount of contamination is estimated to be considerably less than this limit.

Self-supporting carbon targets with thicknesses in the range of $10\text{-}40 \mu\text{g}/\text{cm}^2$ were employed. The target thickness was compared directly to that of a standard target ($\approx 40 \mu\text{g}/\text{cm}^2$) several times during each run. An absolute thickness calibration was obtained by direct measurement of the weight of a second standard target. The targets were carefully selected for their uniformity and the possibility of appreciable carbon buildup was eliminated by using low beam currents ($\approx 100 \text{ nA}$) and by maintaining a vacuum in the scattering chamber of less than 10^{-6} Torr.

Scattered protons were detected by two Si(Li) solid-state counters located symmetrically at laboratory angles of $\pm 165^\circ$. This geometry was particularly insensitive to small changes in the beam direction, which was defined by an entrance beam collimator and a split Faraday cup. The counter resolution was sufficient to separate the elastic yield from ^{12}C from that due to small ^{13}C , ^{14}N , and ^{16}O contaminants which might produce a different energy dependence over the narrow range of the resonance measurements.

For the measurements with protons, H_2^+ and H_3^+ the magnetic field, as monitored by a NMR probe in the first analyzing magnet, was held constant to about 1 part in 10^5 (≈ 0.3 keV in the proton energy). The reproducibility of the measurements for protons and H_2^+ was found to be ≈ 0.5 keV in the proton energy.

3. Results

Figure 1 shows excitation curves for protons, H_2^+ and H_3^+ for a laboratory angle of 165° covering proton energies in the neighborhood of the lowest $T = 3/2$ level in ^{13}N ($E_p = 14.232$ MeV, $E_x = 15.066$ MeV).⁶⁾ The proton yields have been plotted as a function of the energy of an individual proton in the beam. In each case the anomaly due to the narrow resonance ($\Gamma = 1.1$ keV)⁷⁾ is clearly seen, although the size of the effect and its apparent width are obviously functions of the type of beam employed. The energy loss in the target is less than 0.5 keV. While the curve shown for protons corresponds to a slightly thicker ^{12}C target, other measurements with different targets have verified that for such thin targets the size and shape of the proton resonance curve is insensitive to the target thickness. This shows that energy loss and straggling effects are small when compared to the molecular-ion effects that are to be explained.

For the H_2^+ and H_3^+ measurements the resonance anomalies occur at molecular ion energies of 28.46 and 42.70 MeV, respectively. Expressed in terms of equivalent proton magnetic rigidities, these correspond to protons of 56.9 and 128.1 MeV. These calibration points cover the range of magnetic rigidities of beams available from the Berkeley 88-in. cyclotron quite adequately. As will be apparent from the theoretical treatment in the next section, this technique can be extended to other proton resonances provided sufficiently thin targets are available.

4. Analysis

The preceding section has shown that when a nuclear reaction is induced by a beam of molecular ions instead of by a beam of atoms or bare nuclei, narrow resonances are broadened and diminished in amplitude. It must be presumed that these effects can be accounted for in terms of an increased energy spread in the beam due to the randomly oriented internal molecular velocities. If \vec{v}_0 is the velocity of the molecular ion beam and \vec{v}_m is the velocity of a proton with respect to the center of mass of the ion, then the proton energy shift is:

$$\Delta E = \frac{m}{2} |\vec{v}_0 + \vec{v}_m|^2 - \frac{m}{2} v_0^2$$

$$\approx m v_0 v_m \cos \alpha \quad , \quad \text{since} \quad |\vec{v}_m| \ll |\vec{v}_0| \quad ,$$

where α is the angle between \vec{v}_0 and \vec{v}_m and is distributed randomly. This simple expression shows clearly that, though the molecular energies are only of the order of a few eV, energy spreads a thousand times larger are possible for beam energies in the MeV range.

The internal velocities arise principally from two sources: the vibrational energy of the molecular ion⁸) and the acceleration of the nuclei in the molecule due to their mutual Coulomb repulsion after their binding electrons are stripped off at the surface of the target⁹). The first of these effects is independent of the nature of the target and depends only upon the structure of the molecular ion. The latter effect must, however, be strongly influenced by the thickness, density, and composition of the target material. This dependence arises because the nuclear reaction samples only the short time interval ($\approx 10^{-15}$ sec in this case) during which a beam proton is in the target. In this period the

Coulomb force can only accelerate the protons to a small fraction of their ultimate terminal velocity; thus, only a small fraction of the maximum spreading of the beam energy is achieved before the proton leaves the target.

In the next paragraphs we shall investigate the relative contributions of these two effects; however, the Coulomb "explosion" of the ion must, even at this stage, be suspected as the origin of the discrepancy between these results and the conclusions of Parkinson and Bardwick²).

In order to determine the shape of a nuclear reaction resonance observed using a molecular ion beam, we shall consider only diatomic molecules; more complicated molecules are dealt with by a two-body approximation. At the present time this approach is well justified by the limited precision of the data. In most of the derivation H_2^+ will be used as the example; however, the results are easily modified for any other diatomic molecule.

The form of the resonance, $N[E(p_0)]$, can be expressed in the following way:

$$N[E(p_0)] = \int_0^{t_E} dt \left\{ \int_{-\infty}^{\infty} f_{p_1}(p_1, t) Y'(p_0 + p_1) dp_1 \right\},$$

where: p_0 is the nominal momentum of a proton in the beam; p_1 is the net contribution to the total momentum from molecular effects; t_E is the transit time for a beam proton in the target; $Y'(p_0 + p_1)$ is the observed yield $Y(E)$ per unit transit time for a proton beam that has been expressed in terms of the proton momentum; and $f_{p_1}(p_1, t)$ is the probability density that a proton will have an increment p_1 to its momentum at the time t .

$N(E)$ has been expressed in this way because, by introducing Y' , the integrals over the intrinsic beam spread, target energy loss and straggling are

unnecessary. This method of decoupling the multi-dimensional integral has the further advantage of eliminating the necessity of knowing the intrinsic line shape of the resonance.

Since $Y(E)$ is obtained by scattering protons from a carbon target, we have only to concern ourselves with $f_{p_1}(p_1, t)$. The model used contains the following assumptions.

1. Before it hits the target the molecule can be characterized as a simple harmonic oscillator in its ground state.[†] (The energy associated with rotational excitation is an order of magnitude smaller and its effects can be neglected.)

2. The binding electrons of the molecule are stripped off at the surface of the target (by large angle e-e collisions) and move away fast enough that their influence on subsequent processes can be ignored.

[†]Note that for both H_2^+ and H_3^+ , electric dipole decay is forbidden for transitions among the vibrational levels. Thus, it is conceivable that the excited states populated on leaving the ion source are still partially occupied after the $\approx 4 \times 10^{-5}$ sec transit time to our target. Walters, et al.¹⁰) have shown experimentally that the population of such excited vibrational levels is sufficiently low that the results we present would not be affected appreciably. Since their experiment involved the short transit time characteristic of an electrostatic accelerator, we shall assume that the much longer acceleration period of the 88-in. cyclotron (\approx a factor of 100) brings us even closer to our assumed situation.

3. The stripping of the electrons occurs sufficiently fast ($\approx 10^{-17}$ sec) that it does not disturb the "instantaneous" state of the protons, i.e. immediately after stripping the wave function of the protons is still that of a ground-state harmonic oscillator.

4. The molecular ion is randomly oriented with respect to the beam direction.

5. The interaction of the protons with the target material does not appreciably change p_1 . This implies that the Coulomb repulsion of the protons is not screened by a redistribution of the target electrons. That this assumption is valid has been checked crudely using a free electron model of the solid target. In the short time that the protons from the molecular ion remain near a given electron, that electron will not be displaced sufficiently to appreciably modify the repulsive interaction of the protons. (Because this approximation can be justified in this particular case does not imply its universal validity; at low bombarding energies (≈ 100 keV) such screening effects may indeed be important.)

The momentum, \vec{p}_t , of an individual proton in the beam is

$$\vec{p}_t = \vec{p}_0 + \vec{p}_v + \vec{p}_c(t) = \vec{p}_0 + \vec{p}_m,$$

where: \vec{p}_v is the momentum due to the zero-point vibration of the molecule;
 $\vec{p}_c(t)$ is the momentum arising from the Coulomb repulsion between the protons;
 and \vec{p}_m is the total molecular contribution to the momentum vector. If α is the angle between the beam direction and the common axis along which both \vec{p}_v and $\vec{p}_c(t)$ are directed, then:

$$p_t = (p_0^2 + p_m^2 + 2p_0p_m \cos\alpha)^{1/2}$$

$$\approx p_0 + p_m \cos\alpha, \quad \text{since } p_m \ll p_0.$$

From this we can identify p_1 as $p_m \cos\alpha$; using the above definitions this can also be expressed as $(p_c(t) + p_v)\cos\alpha$.

The probability density $f_{p_1}(p_1, t)$ can now be calculated by finding the probability densities of the individual momentum components and folding in the effects due to the distribution of α . Consider first $f_{p_v}(p_v)$, the probability that the zero-point vibration energy results in a residual momentum p_v when the molecule is dissociated. (Note that p_v is directed along or opposite to the direction of $p_c(t)$.) If p and x are the momentum and distance from equilibrium in the center-of-mass system, then the vibrational ground state of the molecule is described by:

$$\psi_0(x) = \frac{a^{1/2}}{\pi^{1/4}} e^{-a^2 x^2/2},$$

where $a = \sqrt{\frac{m\omega}{2\hbar}}$, m is the proton mass and ω is the vibrational frequency.

The Fourier transform of this wave function

$$\phi_0(p) = \frac{1}{(a\hbar)^{1/2}\pi^{1/4}} e^{-p^2/2a^2\hbar^2},$$

yields the momentum distribution

$$f_p(p) = |\phi_0|^2 = \sqrt{\frac{2}{m\omega\hbar\pi}} e^{-p^2/a^2\hbar^2}.$$

Since the momentum of either one of the protons is the same as the momentum of the reduced mass,

$$f_{p_v}(p_v) = \sqrt{\frac{2}{m\omega\hbar\pi}} e^{-p_v^2/a^2\hbar^2}$$

Now consider the momentum arising from the Coulomb explosion. The classical differential equation that governs the motion of the protons is:

$$\frac{d^2r}{dt^2} = \frac{\beta}{r^2},$$

where $\beta = \frac{e^2}{4m}$, and r is the distance between one proton and the center of mass. The solution of this equation is:

$$t(r) = \frac{r_0}{2\gamma} \left[\sqrt{\frac{2r^2}{r_0} - r} + \frac{1}{2} \sqrt{\frac{r_0}{2}} \ln \left| 2 \sqrt{\frac{2}{r_0} \left(\frac{2r^2}{r_0} - r \right) + \frac{4r}{r_0} - 1} \right| \right],$$

$$\frac{dr}{dt} = \gamma \sqrt{\frac{2}{r_0} - \frac{1}{r}},$$

where $\gamma = \sqrt{2\beta}$ and r_0 is the interatomic spacing of the protons in the molecular ion. These expressions can be combined numerically to give the desired quantity, $p_c(t)$. For the cases of present interest, however, the protons move apart only $\approx 10^{-9}$ cm in the time it takes them to transit the target, while r_0 is $\approx 10^{-8}$ cm. Therefore, $p_c(t)$ can be approximated to good accuracy by ma_0t , where a_0 is the initial Coulomb acceleration. The following derivations do not specify the functional form of $p_c(t)$, and the more precise form can be used where necessary.

The zero-point vibration corresponds to an uncertainty in the interatomic spacing at which the Coulomb explosion begins. Although $\psi_0(x)$ is used to obtain the distribution of these variations, the resulting probability density cannot be readily combined with $f_{p_v}(p_v)$, because x and p are not independent variables. To do the problem correctly would require quantizing the oscillator in states of definite $p_m(t)$ and then calculating the amplitude for the ground state to go to each of these states--quite a difficult undertaking. Fortunately, for the cases of current interest the uncertainty in x introduces at most a 10-20% uncertainty in the initial Coulomb acceleration. After the averaging over α , this results in a negligible 2-3% uncertainty in the part of p_1 due to the Coulomb explosion. Therefore, we shall ignore this effect.

Since we now know the form of $p_c(t)$ and the distribution of p_v , we may express the momentum distribution in terms of p_m . This will allow us to eventually perform our integration over $p_1 = p_m \cos\alpha$ in a more straightforward manner than if we were to insist on averaging first over the momentum p_v of the oscillator.

The corresponding probability density for p_m is:

$$f_{p_m}(p_m, t) = f_{p_v}(p_m - p_c(t)) = \sqrt{\frac{2}{m\omega\hbar\pi}} e^{-(p_m - p_c(t))^2/a^2\hbar^2}$$

Since the breakup of the molecular ion is isotropic in its center-of-mass system and since $\cos\alpha$ and p_m are independent,

$$f_{p_m, \cos\alpha}(p_m, \cos\alpha, t) = \sqrt{\frac{1}{2m\omega\hbar\pi}} e^{-(p_m - p_c(t))^2/a^2\hbar^2} \quad \text{for } |\cos\alpha| \leq 1$$

where $f_{p_m, \cos\alpha}(p_m, \cos\alpha, t)$ is the joint distribution function of p_m and $\cos\alpha$. Consider now the mapping of the $(p_m, \cos\alpha)$ plane onto the (p_m, p_1) plane, defined by $p_1 = p_m \cos\alpha$.

$$f_{p_1, p_m}(p_1, p_m, t) = \sqrt{\frac{1}{2m\omega\hbar\pi}} \frac{1}{|p_m|} e^{-\frac{(p_m - p_c(t))^2}{a^2\hbar^2}} \quad \text{for } |p_1| \leq |p_m|$$

Thus,

$$f_{p_1}(p_1, t) = \int_{-\infty}^{\infty} f_{p_1, p_m}(p_1, p_m, t) dp_m$$

Returning to the original expression for $N(E(p_0))$ and interchanging the order of integration of p_1 and p_m , we obtain

$$N[E(p_0)] = \sqrt{\frac{1}{2m\omega\hbar\pi}} \int_0^{t_E} dt \left\{ \int_{-\infty}^{\infty} \frac{1}{|p_m|} e^{-\frac{(p_m - p_c(t))^2}{a^2\hbar^2}} g(p_m, p_0) dp_m \right\},$$

where

$$g(p_m, p_0) = \int_{-|p_m|}^{|p_m|} Y'(p_0 + p_1) dp_1$$

Expressing g in terms of E_{Res} , the nominal energy of a beam proton at the resonance

$$g(p_m, p_0) \approx \frac{m}{p_{Res}} \left[I(E_{Res} + \frac{|p_m| p_{Res}}{m}) - I(E_{Res} - \frac{|p_m| p_{Res}}{2}) \right],$$

where p_{Res} is the proton momentum corresponding to the resonance energy (assuming a narrow resonance) and

$$I(E) = \int_{-\infty}^E Y(E) dE .$$

$I(E)$ can be easily calculated by graphically integrating the observed yield curve for the proton beam, and $N(E_0)$ is now in a form that permits numerical integration.

The results are shown in figs. 2 and 3. For improved clarity of presentation a constant contribution to the yield was subtracted from the molecular ion results. Figure 2 gives the resonance shapes for H_2^+ and H_3^+ ions on a $10.3 \mu\text{g}/\text{cm}^2$ carbon target. It must be emphasized that in this figure as well as in fig. 3, there are no adjustable parameters in the theoretical analysis. The shape and size of the curve are completely determined from the accepted values of ω and r_0 for H_2^+ (ref. 11) and H_3^+ (ref. 12) together with the proton resonance curve.

Figure 3 provides a resonance curve for H_2^+ incident on a thicker target ($13.9 \mu\text{g}/\text{cm}^2$) and a family of curves calculated for H_2^+ corresponding to a range of target thicknesses (given in $\mu\text{g}/\text{cm}^2$). The latter clearly shows that near the front of the target (or for thin targets, $\lesssim 3 \mu\text{g}/\text{cm}^2$) the energy spread is dominated by the zero-point motion. At intermediate distances into the target (3 to $15 \mu\text{g}/\text{cm}^2$) the contribution from the Coulomb explosion dominates and soon the observed resonance width substantially exceeds that seen with a proton beam. This causes not only a further broadening of the resonance but also allows fewer resonant reactions to take place in the target volume, thus decreasing the

resonant yield per $\mu\text{g}/\text{cm}^2$. At greater depths the energy spread from the Coulomb explosion is so large that essentially no further resonant reactions can occur. The resonance thus retains its shape and magnitude as the thickness of the target is further increased, but the ratio of resonant yield to non-resonant yield decreases linearly with thickness.

Figure 4 shows a smooth curve representing the proton data together with calculated curves for H_2^+ and H_3^+ beams on a $1 \mu\text{g}/\text{cm}^2$ target. The only appreciable contribution to these curves comes from the zero-point motion of the molecular ions. Thus, the curves represent the "intrinsic" resonance shapes observable with these beams.

5. Conclusions

The data obtained for the 14 MeV resonance in $^{12}\text{C}(p,p)^{12}\text{C}$ show clearly that by using beams of protons, H_2^+ ions and H_3^+ ions to observe the same resonance a range of calibration points corresponding to widely different beam rigidities can be obtained. The discrepancy with the conclusions of Parkinson and Bardwick²⁾ has been resolved. The energy spread of the breakup protons from a molecular ion beam observed at the focal plane of a spectrometer corresponds to the terminal velocity from the Coulomb explosion. This spread is necessarily much greater than that which can be attained in the $\approx 10^{-15}$ sec period sampled by observing the scattering from a thin target. These results yield a new appreciation of the role played by the spatial localization of the constituents of the incident molecular ion upon entering a solid target.

It must be noted that the close proximity in space ($\approx 1\text{\AA}$) and time ($\approx 10^{-17}$ sec) for the impact of the several protons in the molecule on the target allows, at least in principle, the observation of their successive or simultaneous interaction with a single atom of the target. This could permit the observation of the interaction of protons with excited atoms or the modification in the energy loss for the second of a pair protons due to the prior passage of the first.

These results have inspired the investigation of the proton backscattering when molecular ions are incident on aligned silicon crystals¹³⁾. In this case small, but significant differences have been observed when axial channels in the crystals are aligned with the beam direction.

Acknowledgments

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Figure Captions

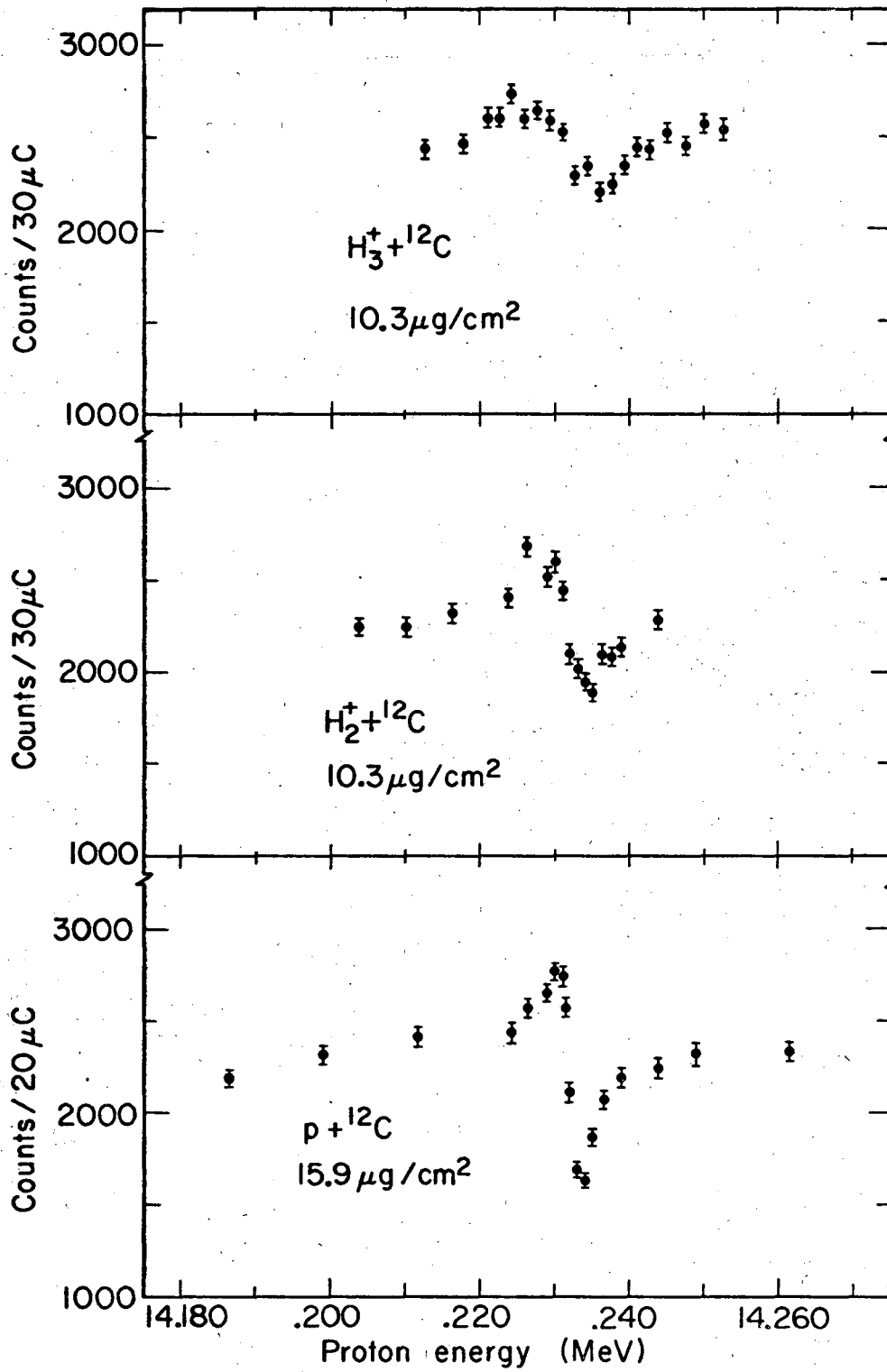
Fig. 1. Proton yield curves for a laboratory angle of 165° corresponding to the bombardment of carbon targets with beams of protons and the molecular hydrogen ions H_2^+ and H_3^+ . The energy scale shown refers to the energy of a proton in the beam; the target thickness and integrated charge (protons) are shown for each experiment separately. (The resonance shown corresponds to the $J^\pi = 3/2^-$, isospin $3/2$ level in ^{13}N at $E_x = 15.066$ MeV.)

Fig. 2. Resonant yield curves for the bombardment of a $10.3 \mu\text{g}/\text{cm}^2$ carbon target with beams of H_2^+ and H_3^+ . The energy scale shown indicates the proton energy displacement from the resonance energy, E_{Res} . A constant, non-resonant contribution to the yield has been subtracted from each point for greater clarity of presentation. (Note that the theoretical curves do not involve free parameters.)

Fig. 3. (Bottom): A resonant yield curve for the bombardment of a $13.9 \mu\text{g}/\text{cm}^2$ carbon target with an H_2^+ beam.

(Top): A family of resonant yield curves calculated for the bombardment of carbon targets of various thicknesses (shown in $\mu\text{g}/\text{cm}^2$) with H_2^+ ions. The curve labeled with "p" is a smooth line drawn through the proton data.

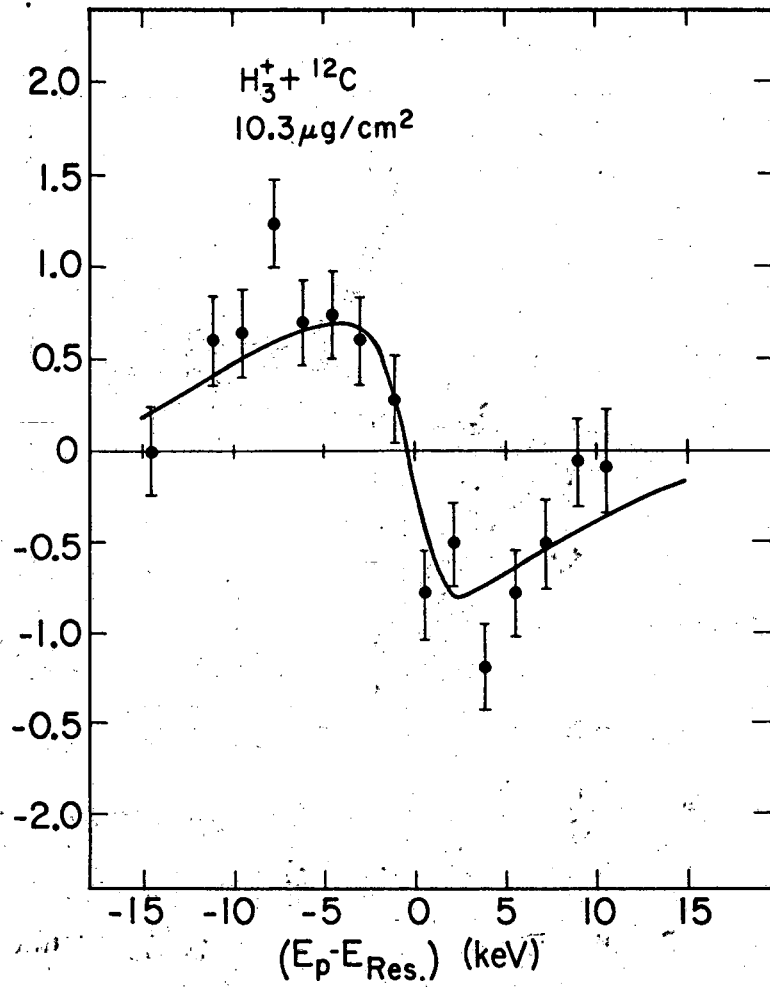
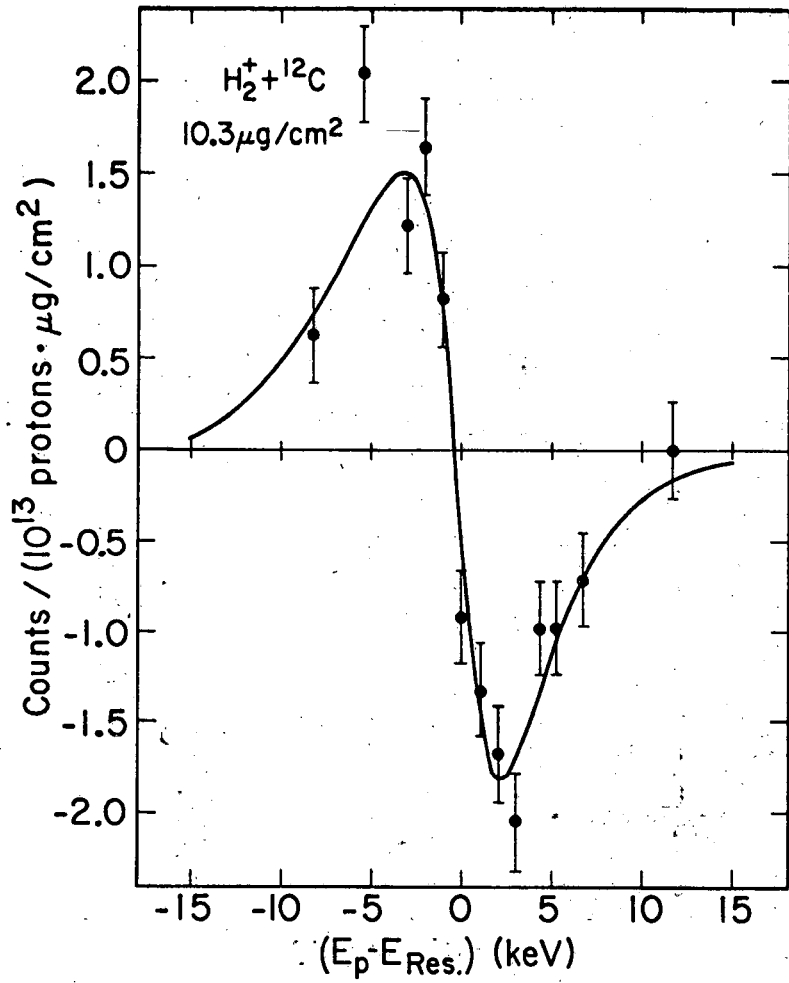
Fig. 4. The curves shown correspond to the proton data (labeled with a "p") and calculated curves for the bombardment of a $1 \mu\text{g}/\text{cm}^2$ carbon target with H_2^+ and H_3^+ ions. The dominant contribution to these calculated curves comes from the zero-point motion of the incident molecular ion.



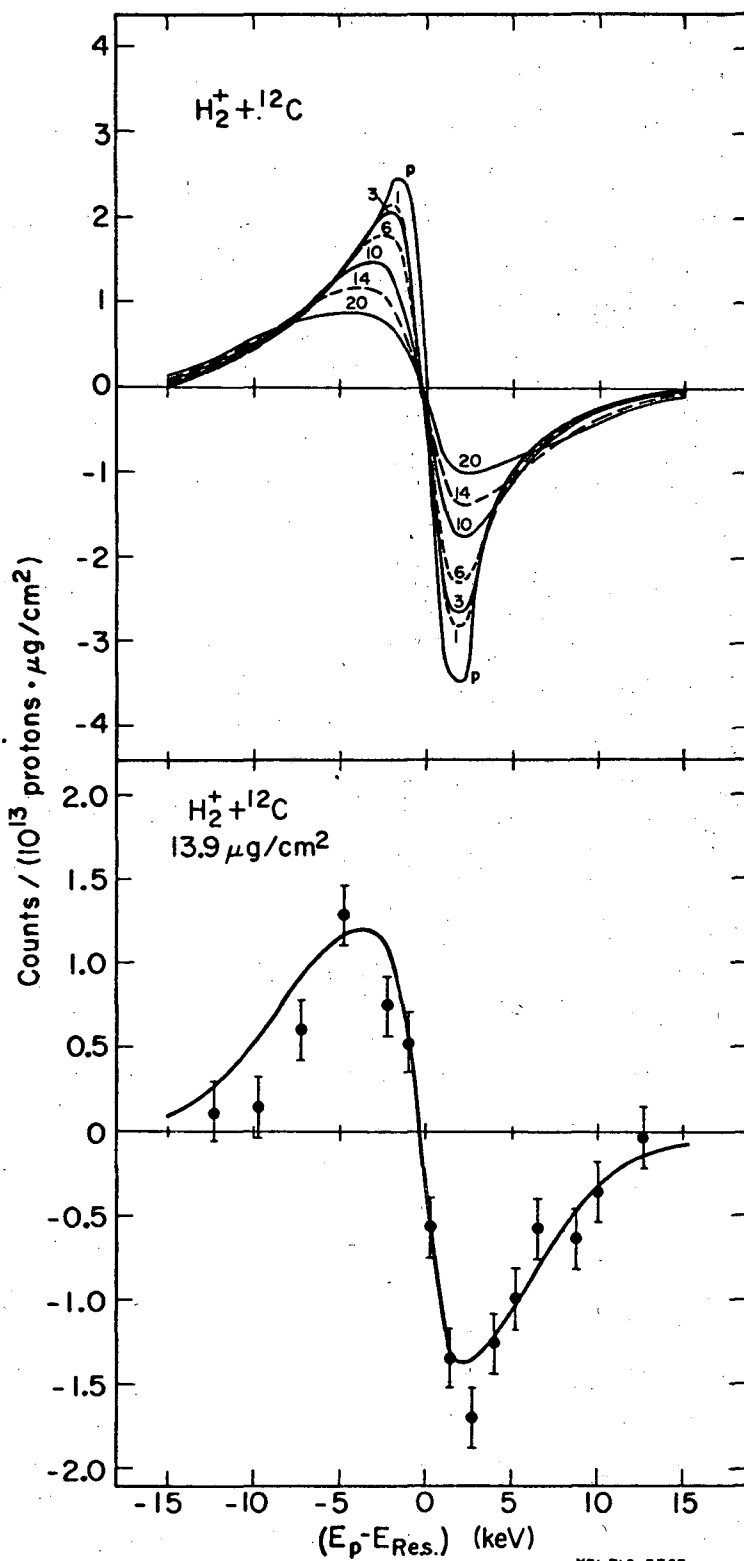
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Fig. 1

Fig. 2

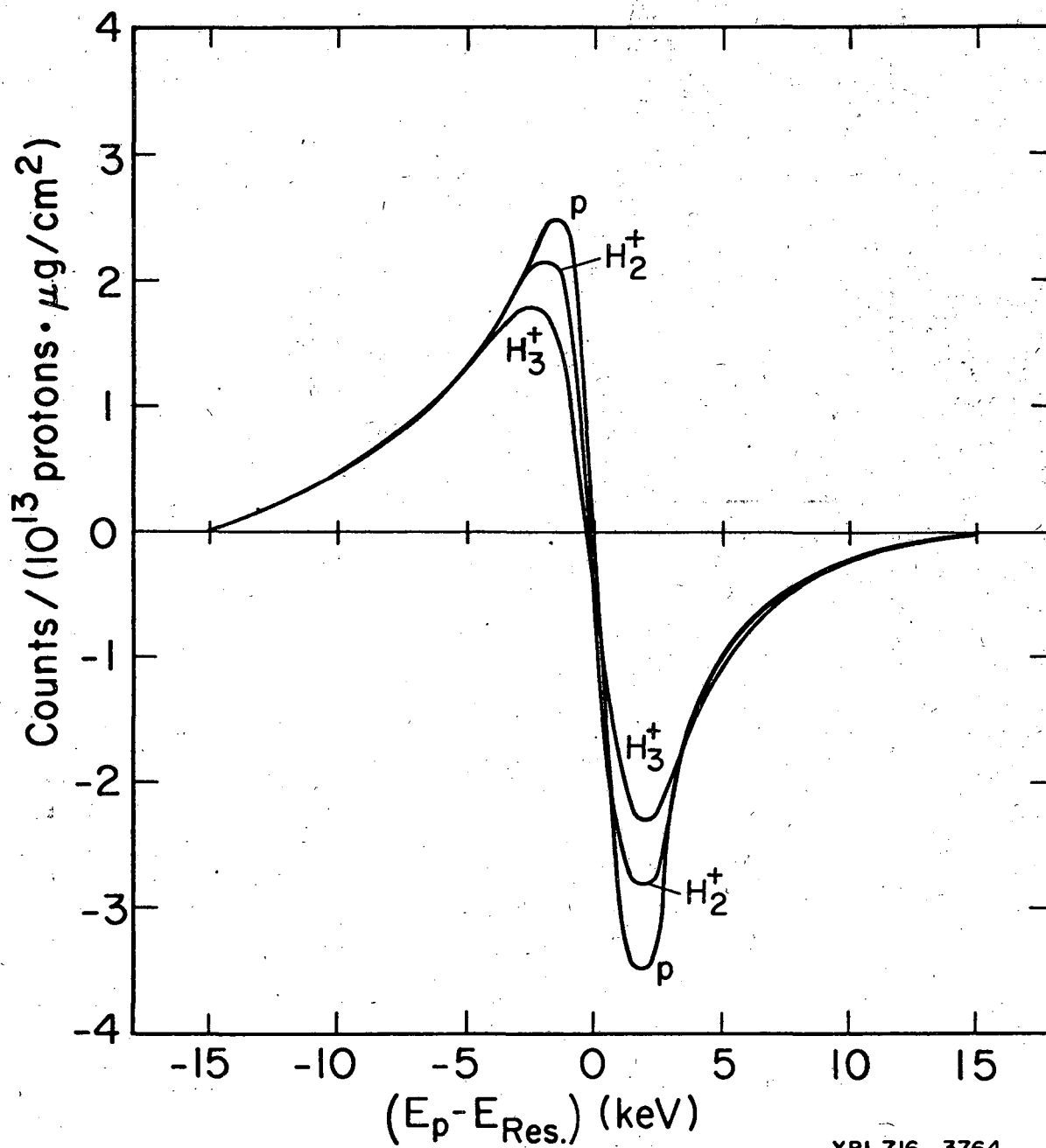


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Fig. 3



XBL 716 - 3764

Fig. 4

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